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# Contents

# An Historical Review and Perspective of AINSE

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## Abstract

AINSE was formed in 1958 as a cooperative venture of modest scope, involving the newly established Australian Atomic Energy Commission (AAEC) (created by the Atomic Energy Act of 1953 with facilities at Lucas Heights being formally opened in 1955) and the nine universities that existed at the time. Research emphasis was very much nuclear, with the two reactors MOATA and HIFAR and possible future nuclear energy developments defining it. Two accelerators, added in the early sixties - the 3 MV Van de Graaff and the 1.3 MV electron machine, were to sustain the original focus of the AAEC. It would be true to say that AINSE in those early days placed much importance on the general support of nuclear science throughout Australia, whereas now of course the facilitation of research by University staff and students at ANSTO has become the main function.

Thereafter, both AINSE and the AAEC have undergone dramatic change. The number of universities expanded to 19 during the sixties, along with more support and encouragement for research at both the new institutions and the original group of nine. University use of Lucas Heights facilities, through the agency of AINSE, increased and began to diversify somewhat into other disciplines - a trend that has continued apace ever since. In the nineties, the Dawkins revolution led to a doubling of the number of Universities, so that once again AINSE experienced a quantum jump in size, with of course matching complexity. In parallel, the AAEC broadened its activities to embrace a wide range of nuclear and energy related areas, though basic research began to taper off. Finally, the organisation was given a new charter in 1985 and re-named the Australian Nuclear Science and Technology Organisation (ANSTO).

A much expanded university system, the "new" ANSTO, the rise of economic rationalism and the creation of the ARC have combined to provide a succession of challenges to AINSE. From the original small, club-like beginning with narrow interests, AINSE has emerged with a four-fold increase in membership, to foster research within many of the Faculties of those members.

### In the beginning

The Australian Institute of Nuclear Science and Engineering came into being formally on December 4 1958, when a group of representatives from eight of the nine universities and four from the AAEC accepted a draft Constitution. Those present included D. J. Allan-Williams (UWA), J. P. Baxter (AAEC), G. H. Cheesman (TAS), D. O. Jordan (ADE), L. H. Martin (MEL, but representing the Commission; a nomination from Melbourne was anticipated), A. D. McKnight (AAEC), H. Messel (SYD), R. A. Smith (UNE), E. W. Titterton (ANU), C. N. Watson-Munroe (AAEC), H. C. Webster (QLD) and L. C. Woods (UNSW). The meeting was held in the temporary headquarters of the AAEC in Beach Street, Coogee. No attempt has been made to establish the background preceding that inaugural meeting [1], though clearly the Government of the time was fully aware of the venture. A lengthy apology was received from Senator Spooner, the Minister for National Development, who had hoped to be able to attend. In it, he confirmed that the Federal Government would contribute £50,000 per year to AINSE, inter alia for the purpose of awarding scholarships and fellowships. For some years before then, the Commission had operated a similar scheme within the same allocation. A so-called Research and Training grant was thus to be transferred from the AAEC to AINSE. There was to be a second allocation directly to AINSE of £60,000 to establish a building at Lucas Heights. It was to be May 1960 before Council was able to meet in its permanent quarters.

AINSE's modus operandi was to evolve over a number of years, influenced of course by the installation and development of facilities at Lucas Heights. At that first meeting though, the objectives had one central theme - uranium and atomic energy, consistent with the prevailing optimistic vision for nuclear energy and other peaceful uses of "the atom". Within the Constitution, the rules to be followed by AINSE were :

- (a) to carry out research and investigation in connection with matters associated with uranium and atomic energy.
- (b) to arrange for the training of scientific research workers and the establishment and award of scientific research studentships and fellowships in matters associated with uranium and atomic energy.
- (c) to collect and distribute information relating to uranium and atomic energy.

Two final rules were more general, relating to the publication of scientific and technical reports and the stricture "to do all things incidental or conducive to the attainments of the Institute".

Initial subscriptions to be paid by members were minuted without recorded discussion. The Universities of Sydney, Melbourne and New South Wales, and the Australian National University, were nominated for £2,000 each, the Universities of Adelaide and Queensland for £1,000 and the Universities of New England, Western Australia and Tasmania for £500. The AAEC subscription of £11,000 matched the total University contribution.

#### Now what?

Appointment of an Executive Officer to run the fledgling organisation became a pressing matter. Prolonged discussions sought to define the essential qualities and a job description; the preference between a scientist and an administrator was not always well-defined though. On one point, that of salary, there was more but not unanimous agreement. Many of the new Councillors supported a somewhat unworldly view that it might be necessary to pay as much as £4,000 pa, essentially a professorial salary, to get the right person, and indeed one Councillor stated that the guiding hand of AINSE should be "of professorial calibre".

No suitable applicant emerged from a widely promulgated advertisement. Thereafter, Councillors proposed a number of potential candidates, though there is nothing to suggest that any of them evinced any interest. Without preamble, Bill Palmer's name appeared suddenly in the Minutes of a November 12 1959 meeting as the newly appointed Scientific Secretary. The University of Melbourne was pressed to release him so that he could begin duties on February 2 1960. The appointment was a watershed for AINSE. Over the next 28 years, Palmer was to mould the Institute into an effective and pro-active organisation, and to give it much of the character that it retains to the present time.

Though the search and appointment of Palmer had occupied most of 1960, the ways in which AINSE would begin to fulfil its obligations with respect to "uranium and atomic energy" were under way by the end of the year. Beyond the scholarship and fellowship functions which were straightforward enough, it would seem that, at the outset, most Councillors saw AINSE as either or both the means by which University researchers could gain access to the facilities at Lucas Heights, as and when they became available, and as a source of research funding both for there and at the Universities. Less explicit, though implicit from early decisions, was to be the role of AINSE in assisting with the development of facilities at Lucas Heights for mutual benefit. At least one Councillor had a different viewpoint, suggesting that AINSE should become involved "to push out from the Commission staff, facilities and finance for atomic energy work to be carried out in the Universities themselves". Such a reversal never did occur, although some contract work for the AAEC was undertaken in the Universities in the following years.

Three requests for research grants were considered at a meeting in April 1959. Two were for travel expenses; the other from Eric Hall of the Newcastle University College was to enable "the neutron irradiation of materials". The latter was deferred pending further details being supplied, so that the first AINSE grant awarded was for £100 to allow an Honours student from the University of Queensland to work at Lucas Heights for a week during a vacation period. In November 1959, seven PhD scholarships were awarded [2]. At the same meeting, it was decided to hold a Radioactive Dating conference early in 1960. Here AINSE was being pro-active, while in another sense it was marking time until research facilities were available with the HIFAR reactor (the first neutron diffractometer was still being developed by Terry Sabine with AINSE support in mid-1960). Indeed, most of the grants awarded between 1959 and 1963 were for the support of research and PhD scholarships at the Universities and for conference travel. Radioactive dating was to become a dominant area, followed by thermonuclear (plasma) research for which modest support was provided to Watson-Munro, who had moved to the University of Sydney. Surprisingly perhaps, the first of the continuing AINSE conferences that were to become an important contribution to Australian science was that of Plasma Physics in 1962.

After the Radioactive Dating conference (April 1960), it was resolved to allocate an amount not exceeding £10,000 over three years to support two ventures claiming the ability to make radio-carbon measurements "within a few months" and to provide for a Senior Fellowship to develop dating techniques at Lucas Heights. Thus the Victorian Museum of Applied Science was granted £695 for equipment and the University of NSW provided with funds to allow appointment of a professional scientist (£1800 pa) and a technician (£1200 pa) as well as £2950 for equipment. G. H. Riley took up the Fellowship in 1961, working at Lucas Heights on both rubidium/strontium and rhenium/osmium techniques. The group at NSW was surely ambitious if nothing else - J. H. Green reported in 1961 that age studies of meteorites were using  $^{26}$ Al,  $^{10}$ Be and  $^{36}$ Cl as well as  $^{14}$ C. Though radio-carbon counting was implemented successfully over the following years at several centres, including

AAEC, the first-named, three cosmogenic isotopes could be exploited effectively only when accelerator mass spectrometry was introduced many years later [3].

The first new member, Monash, was recruited in 1961.

### Fast forward

Clearly a complete history is beyond the scope of the present review. It is instructive though to scan quickly over the subsequent years to gauge the growth of AINSE and the changes of scope and research fashions. Two years are chosen. The first is 1970, a decade after the Institute began to function with Palmer at the helm. and the second is 1990, just prior to the emergence of "the new AINSE" in 1992.

James Cook University joined AINSE in 1970 to bring the University membership to 15. The University of Newcastle (1965), Flinders, La Trobe and Macquarie (1966) had joined in the intervening years.

The pattern of activity that had developed is apparent from the conference schedule for 1970/71 - the Fifth Radiation Chemistry (1970), the Eighth Plasma (1971), the Fifth Heat Flow and Fluid Transfer (1971), the Third Radiation Biology (1971) and the Third Nuclear Physics (1970) conferences.

An impressive inventory of AINSE funded research equipment available to users at Lucas Heights included powder and single crystal neutron diffractometers, a polarised neutron single crystal diffractometer, a spectrophotometer, modular electronics, a liquid scintillation spectrometer and a high resolution, lithium-drifted germanium detector, along with a multi-channel pulse height analyser.

About 63 projects were funded in 1970, a sharp increase from the 9 of 1960. Now though almost all of them [4] involved use of AAEC facilities, including the HIFAR reactor, the 1.3 MV electron accelerator that probably became available in 1963 [5], and a 3 MV van de Graaff accelerator (installed 1962/3 and still operating effectively). As had become the custom, AAEC facilities were provided without charge [6]; AINSE paid the travel and accommodation costs needed for their access, with modest grants for equipment and expendables.

To some, 1990 was the pinnacle, but also the end, of the golden days of AINSE. Outwardly at any rate, the Institute was flourishing. University membership had grown to 21 [10], with the addition of the University of Wollongong and Griffith University (1975), Murdoch (1985), the University of Technology and RMIT (1988), and Curtin (1989). The AAEC had been re-named ANSTO in 1987, complete with new charter, although the consequences to AINSE of the change still lurked in the wings. Roger Gammon, appointed Scientific Secretary following Palmer's retirement in 1988, presided over a secretariat of three and two groups of AINSE scientific and technical staff, totalling as many as seven in number. Lindsay Davis (1975) and later Shane Kennedy (1989), backed usually by two technical staff, assisted users of the neutron scattering equipment, while one and sometimes two scientists and a technician provided similar support to users of the two small accelerators. David Cohen was an AINSE appointment in the latter area (1976). When he joined ANSTO, Mike Hotchkis (1990) was taken on in his stead.

Some 100 applications for grants received the traditional AINSE support and encouragement. By the end of that year, 61 Fellowships and almost 100 PhD scholarships had been awarded by AINSE since its inception.

### The new AINSE - 1992 onwards

In 1992, a Memorandum of Understanding was signed by ANSTO and AINSE to determine a much altered way in which AINSE would continue to operate, albeit with the same overall objectives. A leaner if not meaner Institute was to operate with a reduced secretariat, with all accounting functions to be undertaken by ANSTO, and with AINSE scientific and technical staff subsumed by ANSTO. AINSE funds, freed of a substantial salary burden, were to be used to pay ANSTO for the use of facilities, at rates discounted from those established for commercial use. ANSTO would continue to contribute a subscription that subsidised the total University contribution on a 2 for 1 basis. This arrangement had evolved with the demise of the original Research and Training grant when the introduction of one-line budgets meant that the grant was no longer a designated item in ANSTO budgets. Council meetings were to be reduced from three (four until 1989) to one each year.

Needless to say, the changes were not introduced without a considerable upheaval and many concerns within AINSE. The MOU and the subsequent implementation of it were thrashed out at many lively meetings of sub-committees appointed to negotiate the details of both. The reasons that the changes were forced onto AINSE (and onto ANSTO for that matter) are many and complex. A brief summary of them stands the risk of being overly simplistic - or worse, of being based in part on ill-informed opinion and hearsay, but one is attempted nonetheless.

A facile explanation could be provided by Government decisions in 1988 and 1990 that the CSIRO and ANSTO respectively should aim to attract, within about four years from the nominated date, at least 30% of their operating funds externally, beyond that of the normal recurrent institutional grants. Subsequent efficiency dividends [7] were likely to increase this external fraction. The new AINSE arrangements meant that ANSTO stood to claw back much of its subscription and those of the Universities to be counted as external income. While such creative accounting [8] may well have been a factor, it was by no means the only one. Both AINSE and ANSTO were beginning to show the effects of the passage of time. In the case of ANSTO, there had been a mid-life crisis in the early seventies when the decision to build a power reactor at Jervis Bay was reversed. The original decision had seemed the climax of much of the AAEC effort in the preceding years, confirming the raison d'etre of the Commission. Suddenly, the AAEC was without a mission. Thereafter, it drifted into a variety of projects, but with aging plant and infrastructure. By the mid-eighties, the vigour of the sixties was no longer evident, nor did there seem to be strategic direction beyond the belief that a replacement reactor loomed somewhere in the future. Many University users complained that facilities had become unreliable. For its part, AINSE too was drifting, with Council showing signs of being an old boys club, taking perhaps more interest in the social arrangements for the several Council visits each year to far-flung member Universities than in the affairs of the Institute. Be that as it may, the burgeoning size of Council meant that organisation of the away meetings was becoming complex and expensive. Users were taking AAEC assistance for granted, often complaining that AAEC staff "stole" their results [9]. What had begun as a partnership between the Universities and AAEC was degenerating into "them" and "us" divisions.

Whatever the truth of the above, due review of the AAEC in 1986 led to a new charter, with emphasis on the practical application of nuclear science and technology, focussing on the needs of industry. ANSTO came into being in April 1987 as a consequence of the ANSTO Act 1987. The Director of the former AAEC, Terry Walker, retired on April 30 1988. He was replaced by David Cook. Cook set about the job of establishing a new image for ANSTO immediately, addressing problems of run-down fabric and facilities. The issue of upgrading accelerator facilities to establish AMS was decided quickly, although significant changes occurred between August 1988 and March 1989 in the way in which it was to be achieved [3]. Almost as quickly, he made it clear that ANSTO proposed to play a much more pro-active role in the affairs of AINSE than had been the case in the past. At a July 1991 Council meeting in Newcastle, an ANSTO position paper "The future of AINSE" was presented, outlining alternatives considered to be more cost-efficient for the conduct of AINSE affairs. Further, AINSE was urged to apply for ARC grants jointly with ANSTO. The "reforms" within the MOU were obviously the outcome, driven more by the perceived need to streamline AINSE, and to make ANSTO sections more reliant on satisfying AINSE users (in blunt terms, "no measurements - no money"; in the language of the times, to develop a service culture with improved delivery), than by need to derive nominally external earnings. Cook had also believed that AINSE Council should operate at a higher level, with most, if not all, Councillors being Pro-Vice-Chancellors (Research) or the equivalent. Fortunately, this concept was not part of the MOU, allowing AINSE to retain much of its unique character in the face of the other changes.

By and large, the new AINSE has functioned well, managing by dint of successful applications to the ARC as a peak body, to sustain a user base that began to expand substantially when AMS measurements, principally radio-carbon, became possible at ANSTO from about 1994 onwards. Available funding for the ANSTO facilities cannot support the total demand by users, but is in fact reasonably well-matched to the time available on the facilities. In 1998, 216 grant applications were received. Of these, 173 were supported in part or fully, and 5 listed as provisional, dependent on the uptake of the first category of grants. Post-doctoral fellowships are no longer awarded. The numbers involved tapered off because of financial restraints; then the nature of them was varied in 1990 to place more emphasis on work at Lucas Heights, before the scheme was finally terminated several years later. Post-graduate student awards have continued, not as full scholarships, but as top up grants to holders of APA or equivalent scholarships. In 1997, 22 students were being supported in this way, with designated ANSTO staff as co-supervisors.

Gradually the new Universities became members, largely as the result of an active recruitment program by the President, Bob Breakespere, during 1995 and 1996. All 35 Australian Universities involved with research [11], and the University of Auckland [12] are members at the present time. Nonetheless, such complete membership has been threatened occasionally (and is likely to continue to be) by the unreasonable and short-sighted insistence by some Universities to devolve the subscription, that plainly should be centrally paid, to their current users of AINSE.

For completeness, the conference schedule for 1997/8 included the Tenth Nuclear Techniques of Analysis (1997), the Twenty-first Plasma Science and Technology (1997), the Seventeenth Nuclear and Particle Physics (1998), the Fourth Radiation

Science -incorporating the Nineteenth Radiation Chemistry and the Sixteenth Radiation Biology (1998) conferences, as well as a workshop in SIMS (1997) and involvement with the Sixth Archeometry Conference (1997). AINSE Winter Schools were held in both years at Lucas Heights to give final year students from all member Universities hands-on experience with equipment at ANSTO. Most of the conferences have continued since inception, although the Heat Flow and Fluid Transfer component was dropped when reactor development was no longer a priority. The relative newcomer, Nuclear Techniques of Analysis, has become an important one, attracting the largest attendances. Presently, it is held in conjunction with the conference of the Vacuum Society of Australia. The Nuclear and Particle Physics conferences have been part of the Institute of Physics Congresses since 1988, becoming an essential, central component of them.

Cook resigned in 1993, to be replaced by Helen Garnett, who had been a central figure in the MOU negotiations. She has been sympathetic in the interpretation of the new agreement. For example, it quickly became clear that one Council meeting a year was sustaining neither adequate continuity nor the past effective networking between Councillors and member Universities. There are now two meetings each year, usually one at Lucas Heights, and the other away. A major review of ANSTO was undertaken in 1994. The report recommended closure of some facilities used by AINSE (most critically, the 1.3 MV accelerator), urging ANSTO to concentrate on core areas. Again, the post-review ANSTO has endeavoured to ameliorate any effects of the review on AINSE, in particular by supporting the transfer of radiation chemistry equipment to the Australian Radiation Laboratory in Melbourne. The spirit of partnership has returned, under-pinned by the new arrangements, and the realisation, perhaps belatedly on both sides, that ANSTO and University researchers collectively must be mutually supportive.

AINSE can look to the new millennium with confidence - a new neutron, small- angle scattering facility, developed with substantial support from AINSE, became available recently, and the Government has agreed that HIFAR should be replaced. The MOU was renewed in 1998 for a further 5 years, without amendment. AINSE will be steered into Y2K by Dennis Mather, who was appointed as Scientific Secretary in 1998 after Gammon retired.

#### Notes

- [1] The historical information presented was gleaned from the formidable collection of Minutes that are available in the AINSE library. Archived correspondence files of some of the more prominent attendees of the inaugural meeting undoubtedly harbour much useful information.
- [2] At least two of the names amongst the seven should be familiar to many Robin Storer, now of Flinders who has attended many Council meetings and Erich Weigold, presently Director of the Research School of Physical Sciences and Engineering at the ANU.
- [3] Accelerator mass spectrometry (AMS) began in Australia in 1985 at the ANU as a joint AAEC/ANU/CSIRO venture. A measurement capability for radiochlorine was successfully implemented using the 14 UD accelerator, though the joint venture was terminated in 1991 as ANSTO set about

establishing independent facilities. Thereafter, techniques were developed at ANU for a number of the other long-lived cosmogenic isotopes and for the anthropogenic planetary elements - plutonium and neptunium. An account of the sometimes turbulent early days of AMS can be found in "A Tower of Strength - a history of the Department of Nuclear Physics (ANU)" by T. R. Ophel (1998).

- [4] Palmer was ever vigilant on the issue of ensuring that all members were able to derive some benefit, in order that University administrations could be assured that AINSE subscriptions were good value. Research at New England was graciously included within the Plasma umbrella and sustained with grants for equipment at Armidale. AINSE had adopted a philosophy (that continues) whereby virtually all valid and scientifically acceptable applications were at least partially funded. Such an enlightened approach contrasts starkly with the ARC criterion of excellence that is supposedly demonstrated by the small fraction of applications funded.
- [5] The 1.3 MV machine was installed under the guidance of David Sangster, in the first instance for radiation chemistry studies related to the use of beryllium as a reactor moderator. A delightful, informal history of the accelerator, that was finally closed in 1996, has been prepared by Sangster. Some of the dates within it should be treated with caution though.
- [6] One interesting exception was the IBM 1620 computer purchased by the AAEC in 1962. A request to use it for calculations was approved, subject to "the payment of the full prime rate hiring rate and of the operator's time"
- [7] A fiendish bureaucratic device. If an organisation managed to survive with reduced funding, then it was rewarded with even less.
- [8] An assertion once made that "of the 37% external income claimed by ANSTO, it was likely that about 9% was genuinely external" could well have been an ill-founded, throwaway line to an audience cynical about bureaucratic imposition of such arbitrary (and arguably, counter-productive) targets. Alternatively, it may have been based on a narrow interpretation of "external", excluding all attracted income that could be traced back to tax-payer funded sources.
- [9] Some involved with AAEC collaborations complained that "their" measurements and projects were cited at conferences and in publications. In fact the complaints reflected more on the shortcomings or at least the lack of nous, of many of those getting measurements done at Lucas Heights. The reputation (and the continuance) of such programs, as well as the advancement prospects for the AAEC staff involved, were not determined by the number of measurements made, nor how well they were made. Instead, contributions to the science had to be evident. If the users were dilatory in preparing work for publication, then AAEC collaborators had to fill the breach for mutual benefit.
- [10] In the early eighties, some facilities at Lucas Heights were transferred to the control of the CSIRO as a result of a Government decision. Thereafter, colocation of the two organisations at Lucas Heights proved somewhat less than harmonious. CSIRO became a member of AINSE in 1982, but withdrew in 1992.

- [11] The two remaining Universities, the Australian Catholic University and Bond University, do not undertake research.
- [12] The University of Auckland finally became a member in 1995, with its subscription paid by the NZ Government (but not subsidised by ANSTO). Involvement by NZ was a long time in the making. A first proposal was discussed in 1963.

# Archaeometry and AINSE

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This AINSE meeting coincides with the 40<sup>th</sup> anniversary of the founding of the journal Archaeometry. When first published by the Oxford University Research Laboratory for Archaeology and the History of Art, the term "Archaeometry" remained undefined. In action, the ensuing volumes focussed on the application of physics-based dating and analysis to archaeological materials and sites. The Archaeometry journal is testimony to more than 40 years of successfully addressing those archaeological questions amenable to physics based enquiry. Another achievement of the Oxford Laboratory is the steady output of AMS dates on archaeological specimens since 1984, a decade before similar facilities became available to archaeologists in Australia. Martin Aitken, a co-founder of the laboratory, later gave a minimalist definition of 'Archaeometry- [as] measurements made on archaeological material' (1961:v). At the same time he seemed doubtful about the way archaeologists use the results generated from physics-based measurement, by saying: A physicist is not a machine that, when fed with material, regurgitates an archaeological answer acceptable without qualifications'. This is a fair warning that instrumental analyses alone does not automatically transmute the base materials of archaeology into higher order explanations of past events.

In their early experiments Libby and deGrosse relied on a wide range of independently dated archaeological materials to assess their newly proposed radiocarbon dating system. As Rainey reported-'[*they*] proposed an experiment with radioactive carbon-14 in an attempt to develop a system for absolute dating of prehistoric events. That was shortly after atomic explosions became reality and before atomic-nuclear research developed a new order of technology. It is no wonder that the two languages of physics and archaeology were mutually almost unintelligible at that time' (Rainey 1971:vii). But, any mutual unintelligibility must have been short-lived because there was a boom in the establishment of radiocarbon dating laboratories soon after Libby's success.

The radiocarbon laboratory of the Applied Science Centre for Archaeology, at the Archaeology Museum of the University of Pennsylvania had, since 1951, relied on dendrochronology for radiocarbon date calibration (Ralph *et al.* 1965). The Pennsylvania laboratory, and others like it, is an institutionally based centre specifically for the support of field archaeology and museum studies. The British Museum Research Laboratory houses similar analytical facilities including radiocarbon dating and NAA. Other museums in Europe and the United States also have long-established laboratories for the analysis of archaeological materials using nuclear physics based analytical systems. The University of London Institute of Archaeology has had well-developed laboratory and analytical facilities established since 1937.

Apart from dedicated archaeological science centres there are a group of institutions where archaeological research is supported as part of their other specialised functions. An important contribution from nuclear physics to archaeological research began around 40 years ago with the US Brookhaven National Laboratory with Neutron Activation Analysis of pottery (Sayre & Dodson 1957). Sayre and his colleagues continued the Brookhaven connection with archaeological research on a wide range of materials from many countries up to recent times.

When it comes to new techniques for observation, analysis or dating, archaeologists have to be unrequited borrowers. This involves the integration of field and laboratory studies, so the idea that everything can be solved at a laboratory bench is generally not supported. The traditionally wide purview of archaeology was reasserted in 1974 when the Journal of Archaeological Science appeared, almost as a response to the more physics-oriented agenda of the Oxford publication Archaeometry. JAS was less coy in its aims by calling for contributions: 'covering material which combines archaeology and other branches of the sciences, including the physical sciences, biological sciences, earth sciences and mathematics'. Since the mid-eighties this call has been reduced to: contributions 'covering the interaction between the sciences and archaeology'; with archaeology dropping from an august place among the sciences to a polar position apart from them. This is an important distinction that suggests the problem of mutual unintelligibility persists. For example Aitken, the co-founder of the term 'archaeometry', recently shared the view, that: '..archaeological studies have not evolved in the same manner as have, for example, physics or chemistry. There has not developed an agreement on a single model of what constitutes the basis of how archaeologists are to "understand" the phenomenon they study since they cannot even agree on what constitute the primary unit(s) of cultural analysis.'(Taylor & Aitken 1997:xiv).

To archaeologists these words may be true but fairly meaningless since archaeology has not developed with the experimental methodologies of physics or chemistry. The reconstruction of social function and cultural context are vital elements in any archaeological research, but remote from nuclear physics. A more acceptable view to archaeologists is that: 'archaeology can be seen as an organised system of analysis of the human past, based on the systematic study of the material manifestations of human actions. Hypotheses derived from analyses of field and laboratory data must be testable, that is, capable of refutation by reference to new data.' (Jones 1982:25).

Australia's nuclear science research facilities at Lucas Heights were set up in 1953 at a time when, elsewhere, nuclear weaponry with the death of countless thousands was contemplated. Future archaeologists may endeavour to reconstruct a virtual site from the exiguous evidence by using every analytical device, but they would also need to account for its purpose. If we need any reminder of the difficulty of the social sciences to operate like physics and chemistry, in an experimental-predictive sense, we need only to contemplate the failure of econometric modellers to successfully include social behaviour, ethics and belief systems in their futurology. These distracting comments emphasise the differences that exist between our operations when we attempt to analyse the content and context of archaeological remains. Meanwhile the term 'Archaeometry' will have to hobble on under an increasing burden of definitions applied by different practitioners.

Radiogenic dating is a good example of why archaeologists have to be cautious, in Aitken's terms, of the verities of physics-based analysis. The constraints arise from the nature of archaeological materials. We all know that a dating system is more than an instrumental procedure. Apart from laboratory factors of sample preparation there

is the Pandora's box of field based weathering effects in the diagenesis of archaeological materials, and this is a specialised study in its own right. Joint research is therefore essential in overcoming possible cross-disciplinary conflicts of interest.

The Archaeometry Laboratory at the Missouri University Research Reactor, otherwise known as the Nuclear Archaeology and Geochemistry Group, is a good example of the interests of archaeology and nuclear physics operating jointly in one institution. Set up in 1988, the laboratory anticipates having Neutron Activation Analyses completed on 40,000 specimens by early 1999 (Glascock 1998). These items include obsidian, pottery and a range of other archaeological materials. This begins to look like the work that has been so successfully undertaken over the last 24 years by Roger Bird and his group on the analysis of archaeological materials at Lucas Heights through the co-operation of ANSTO and AINSE.

### Australian archaeology, ANSTO and AINSE

Forty years ago there was no university-based prehistoric archaeology in Australia. Most of what we now recognise as the astonishing prehistory of our region, has been revealed since the early 60s when the first academic appointments in Prehistory were made at the University of Sydney and the Australian National University. In the pre and post-war eras elsewhere in the world, when archaeological links to centres of nuclear physics were being formed, Australian archaeology was absent from the scene. Even New Zealand had established a radiocarbon laboratory and university-based prehistory research nearly a decade earlier. This meant that for the organisers of nuclear physics research facilities at Lucas Heights archaeology in Australia in 1974 when an archaeological approach to the Australian Atomic Energy Commission was met with polite interest. A main source of the greeting was Roger Bird, who was unstinting in his efforts to generate co-operative work for over twenty years, until well past his retirement.

The work undertaken by Roger Bird and his colleagues was more than simple assistance with the task at hand. They were instrumental in sustaining a major project based on access to the accelerator based PIXE-PIGME facility. Their co-operative research effort has had a significant influence on archaeological research in our region, particularly for the analysis of thousands of obsidian artefacts and their related geological sources, as well as the analysis of pottery and metal artefacts. Some of this contribution can be outlined but a fuller appreciation is to be found in dozens of joint publications. The names of Bird, Clayton, Cohen and Duerden appear often in this early literature and created lasting procedures for work to the present day. Roger Bird in fact undertook his own fieldwork, while on holiday, to increase the source samples of obsidians from Easter Island and northern NSW.

I was privileged to be part of the first prehistory project to receive AINSE cooperative support in 1974. We aimed to chemically characterise all the known sources of obsidian from Papua New Guinea and the south west Pacific, and to relate obsidian artefacts to them using NAA. For reasons of economy, and the need to analyse many hundreds of artefacts, the project adopted accelerator-based prompt nuclear analysis, using PIGME analysis of Na, Al, and F (Coote *et al.* 1972, Bird *et al.* 1981). This first stage analysis of around 700 specimens, yielded a remarkable picture of obsidian transfer over thousands of kilometres from sources in the Bismarck Archipelago to settlements in New Ireland, Vanuatu, New Caledonia and the New Guinea mainland, as well as its carriage from the D'Entrecasteaux Islands to sites around coastal New Guinea. The demonstration that this movement was achieved around 3500 years ago, and was associated with the spread of distinctive Lapita style pottery, other artefacts, and changed settlement practices, generated a great deal of interest. PIXE analysis, with its greater range of around 23 elements, was then developed to allow finer and more unequivocal subdivisions and source allocations for obsidians, and pottery (Rye and Duerden 1982).

This first phase of work was influential in promoting the first Australasian Archaeometry conference in Sydney, opened by the Premier of NSW in 1982. The organisers were from AINSE, ANSTO, The University of Sydney, The Australian Museum, Macquarie University and the ANU. The group adopted the generalist agenda of the *Journal of Archaeological Science* while appropriating the name 'Archaeometry' from the narrower scope of the Oxford Group's publication. This hybridisation, like many such unions, was probably a result of accident rather than design

We attempted to define 'Archaeometry' in a way that could justify our wide-ranging collection of papers. For this purpose we accepted the view that 'archaeometry is archaeology or it is nothing' (Jones 1982). This can be compared with the view that 'archaeology is science or it is nothing' (Taylor and Aitken 1997:xiv). The complete Jones paper, with its epithetic remark, was commended by an American reviewer who says that 'This paper should be required reading for [archaeology] method and theory students as well as archaeometry students everywhere' (Young 1998). But although courses leading to an 'archaeometry' degree exist elsewhere, there are none in Australia.

The success of the Sydney meeting led to the second Archaeometry conference at the National Gallery in Canberra in 1985 where the scope was widened to include the dating and analysis of Aboriginal art. It was also the year that a major effort was made to fill out the picture of the origins of the elusive Lapita culture complex. For nearly a year, archaeologists from Australia, New Zealand and the United States took part in the so-called Lapita Homeland Project. The products of this exercise included collections of obsidian that promised to provide a detailed picture, over time, of the connectivity of settlements in the region over the last 25,000 years. This second phase of PIXE-PIGME analyses at Lucas Heights dealt with about 1500 obsidian artefacts and geological sources.

The continued success of this project is aided by continuing AINSE support. A truly remarkable picture has emerged of obsidian being transported from the Bismarck Archipelago to Borneo in the west and Tonga in the east, giving a total distribution range of more than 6000 kilometres. Add to this the more limited range but carriage across a sea barrier to New Ireland by 20,000 years ago from New Britain. In Manus obsidian from known and unknown sources was being canoed across 30k of ocean from 12,000 years ago until early this century. These are only some of the bare indicators of remarkable happenings in the history of the archipelagos to our north (Allen *et al.* 1989).

A third phase of work since the late 80s on the prehistory of our region is supported by AINSE with research projects undertaken in Papua New Guinea by archaeologists at the Australian Museum, and universities of Sydney, La Trobe, and ANU. The 1100 analyses have targeted detailed information on obsidian distribution and resource use in West New Britain (Summerhayes *et al.* 1998). While this is occurring others working in Australia have benefited greatly over the last couple of years from the ARC/AINSE support for AMS dating of archaeological materials.

From the perspective of archaeology's long term development some cumulative trends in its association with other disciplines can be seen. If its initial 19<sup>th</sup> century collaborators were mainly geology and art history, the first half of this century saw the crucial underpinning of chemistry and biological sciences to studies of sites and organic remains. Since the Second World War Nuclear Physics has had an increasingly important impact on archaeology through the development of systems for radiometric dating and the non-destructive analysis of materials. The most recent advances in understanding our past come from other major developments in the biological sciences, which allow the identification of residues associated with ancient implements, pottery vessels and site deposits, and DNA analyses on human, animal and plant remains. It appears that the total repertoire now available for understanding the past is converging to produce an increasingly detailed picture of human history, with nuclear physics based research being an essential component.

AINSE co-operation has been invaluable for archaeological work in our region over the last 25 years and will be essential in the future. The joint work at ANSTO is more a paradigm of archaeology at work, rather than a new prospect requiring new definitions. On the other hand ANSTO is a primary research organisation that pursues its own agenda. In this context archaeology is not a programmed part of ANSTO's inhouse activities, but it could be. This raises an important issue for archaeologists who rely on data generated over the long term by the instruments of science based research. The successful work on the distribution of obsidians is only one aspect of Australian archaeological research that has been supported by AINSE.

When compared with university and museum research centres in Europe and the United States, Australia has very limited archaeological science resources. The ANU dismantled its Division of Archaeology and Natural History and curtailed access to radiometric dating. It is unreal to expect that AINSE could accommodate even a small fraction of these lost opportunities for archaeology from just one University research group. Any future development of the field will require a realignment of University priorities, but whatever develops it is clear that AINSE should remain an important partner to archaeology through nuclear science based research in Australia.

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# **Nuclear Physics Achievements Via AINSE**

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### Abstract

From its establishment, the Australian Institute of Nuclear Science and Engineering has supported a strong program in nuclear physics studies. This was especially so in the earliest days of the Institute. The studies made use of the Van de Graaff accelerator, the neutron beams from the HIFAR reactor and more recently the Tandem Accelerator on the Lucas Heights site. There were other studies which used some of the supporting infra structure there. Within the university sector, major facilities such as the Betatron and Pelletron at Melbourne University and the EN Tandem, Cyclotron and 14 UD at the Australian National University were involved in various collaborations. The activities covered many areas of research. The more prominent were studies of the nuclear fission process, neutron capture and astrophysics.

## AINSE's Role in Australian Nuclear Physics

Since its inception in 1958, AINSE has played a major role in the development and exploitation of studies in Nuclear Physics. This activity of AINSE has proceeded in two important ways.

- The AINSE Nuclear and Particle Physics Conference Series
- Collaboration and Use of the Lucas Heights Facilities.

### The AINSE Nuclear and Particle Physics Conference Series

AINSE has been the grandfather organisation for a regular series of meetings in Nuclear and Particle Physics which have brought the community together and stimulated exchange of ideas between Australian scientists working at different institutions. In addition, these conferences have attracted some overseas interest, notably from New Zealand, and have also served as a forum to allow visiting scientists at one institution to extend their experience and knowledge publicly to other Australian institutions. The first conference was held in 1962 and succeeding conferences have been held approximately every two years since then. The most recent conference, held in Fremantle, was the 17<sup>th</sup> in the series. In 1988, it was decided to hold the AINSE conference series in conjunction with the larger AIP conferences.

At the beginning, the conference attendance was about 70 delegates although details of the first two conferences are not available in the AINSE archives. The numbers gradually increased and by 1976 the conference size was x and included a secondary stream of nuclear applications. Subsequently, the more applied studies were catered for within the AINSE Nuclear Techniques of Analysis Series which is now bigger in size than its former parent. In recent years, the attendance at the Nuclear and Particle Physics has returned to about 70 attendees reflecting a more modest attention to nuclear physics studies in Australian Universities.

## Collaboration and Use of the Lucas Heights Facilities.

In the early days of AINSE (early 1960), nuclear physics studies in Australia were dominated by groups at the three universities, Australian National University, Melbourne University and Sydney University. At that time, the principal experimental effort was in nuclear structure studies and interactions at energies that would now be described as relatively low energy. There were also some cosmic ray studies. Complementing these experimental studies was a significant theoretical program.

With the expansion of the AAEC Physics Division in the early 1960s, a number of new research facilities were installed in Australia principally for neutron physics studies and to generate accurate nuclear data for nuclear reactor design. This newly established infrastructure had some attractions to the university community as the neutron studies provided complementary data to the studies within the university sector. For example Melbourne University had a significant program in photo nuclear reactions and the facilities at Lucas Heights offered the opportunity to study the reverse process. Nuclear shell structure plays a major role in the systematics of neutron cross sections and thus neutron physics studies provide additional nuclear structure data to the more commonly studied charged particle reaction studies within the universities.

The policy within the AAEC Physics Division was to foster this relationship as any interaction with the university community provided critical mass to the priority projects within Physics Division.

AINSE provided the mechanism for the efficient promotion and management of this interaction.

#### **Research Facilities Used**

The research facilities on the Lucas Heights site that were involved in the interaction in nuclear physics were

- HIFAR which first went critical in 1958. Most studies were performed on the collimated neutron beams eg 6 HGR8,9 and 10, and 4 H1 and 4 H2. There were some minor irradiations in the reactor itself.
- 3 MV Van de Graaff installed in 1964. This facility was the mainstay of the AINSE nuclear physics sponsored research.
- Oak Ridge Electron Linear Accelerator-The AAEC had a major involvement with the pulsed neutron beams from this facility and attracted members of the university community to become involved. These studies terminated towards the end of the 1970s.
- 9 MV Tandem Accelerator This facility was purchased second hand from Rutgers University in early 1989 and arrived on the site in mid September of that year. The facility was refurbished in subsequent years and there have been several charged particle nuclear structure studies since.
- Laboratory Facilities –DLTS, Cameca 5F.

# Major Areas of AINSE Sponsored Research in Nuclear and Particle Physics

The principal areas of AINSE sponsored research have been in the areas of

- Fission Studies. These included measurements of fission cross sections, fission fragment angular distributions, neutron emission, fission barriers, mass yields, fragment kinetic energies and excitation, fission cross section widths and triple humped fission barriers. They involved the use of the Van de Graaff accelerator, neutron beams on the HIFAR reactor, MOATA and some supporting studies at overseas accelerators. Studies of fission neutron spectra made use of AINSE funded experimental equipment on the EN tandem at ANU. The experimental work was supported by theoretical studies.
- Fission Product Yields. These included studies of the symmetric region, and more general compilations.
- Neutron Cross Sections –Total Capture, Inelastic. Important aspects of these studies were nonstatistical processes, such as the valence process, doorway states eg. two particle one hole states. An important target of the studies was the establishment of correlation between neutron widths and radiative capture widths. Both types of cross section studies were carried out at Oak Ridge and analysed at Lucas Heights.
- Neutron Capture Spectra. These were carried out on both the Van de Graaff accelerator and the neutron beams on the HIFAR reactor. They were performed to provide complementary data to those obtained in the cross section studies.
- Solid State Studies. These were mostly concerned with studies of various semiconductor materials such as GaAs and CdTe and their preparation for use in high resolution photon detectors..
- Charged Particle Reactions,
- High Energy Physics. These studies involved consideration of GaAs detectors for the vertex detectors in the Atlas experiment at CERN. They involved extensive studies of the systematics of neutron irradiation damage.
- Astrophysics. The astrophysics studies were related to the neutron capture studies mentioned previously and involved measurements of 30 keV neutron cross sections in support of the s-process studies.

### Joint ANSTO/AINSE Proposals

Over the last 40 years, relevant nuclear physics research has required the use of expensive experimental infrastructure. One of the very positive aspects of the AINSE – AAEC and subsequent AINSE- ANSTO interaction have been carefully prepared joint proposals for the establishment of valuable experimental infrastructure. These proposals sought to address both the university needs and those of ANSTO. Most were ultimately unsuccessful but there was one notable success which led to the installation of the 9 MV tandem accelerator. The various proposal listed in order were

• 1970 - Medium Energy Cyclotron - 150 MeV. This was driven principally by Melbourne University and the AAEC. In effect it aimed at a facility similar to the

Indiana Cyclotron Facility. It is interesting to speculate on the subsequent course of the AAEC research program had this facility been funded.

- 1973 High Current Linear Accelerator 100 MeV. This facility would have been a high intensity pulsed neutron source which would also have been able to drive the critical facility.
- 1981 Applied Tandem Accelerator 8 MV. The first attempt to install a tandem accelerator on the Lucas Heights site began in mid 1981. The principal use of the proposed facility was to be the newly established technique of Accelerator Mass Spectrometry but this was to be complemented by extensions of the previous nuclear physics studies.
- 1983 University of Washington FN 9 MV. An opportunity to satisfy the need for a tandem accelerator presented itself in 1983 when the University of Washington decided to upgrade their accelerator complex and one of two FN tandem accelerators became surplus to their needs. The accelerator was inspected but any prospective deal was unattractive.
- 1986 Texas EN. A second opportunity to purchase a second hand accelerator arose in late 1986. The possibility of acquiring the Texas EN was been considered when the facility was purchased by another group.
- 1989 Rutgers FN. An accelerator facility based on an 8 MV horizontal Pelletron had been developed when the opportunity arose to purchase the Rutgers FN. This proved to be a much more attractive option than those for previously offered second hand facilities and the tandem was purchased for \$US 250,000.

In more recent years this collaboration between ANSTO and the universities via AINSE has been particularly rewarding. A considerable number of AINSE-ANSTO ARC grant applications have been successful beginning with the area detector in 1992 and subsequently a succession of proposals for neutron scattering infrastructure and AMS studies.

### Summary and the Future

This paper has provided an outline of the course of AINSE's sponsorship of nuclear physics research in Australia. No attempt could be made to detail the scientific successes that resulted because of the limited space and the wide range of topics studied. A summary of these can be found in the reports of the 17 AINSE sponsored Nuclear and Particle Physics conferences. However it should be pointed out that the effort was recognised in international circles and the scientific output in neutron physics was ranked with the best in the world.

# AINSE - More than 20 Years of Ion Beam Analysis

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#### 1. Introduction

AINSE has a strong association with people, personalities, scientific training and research spanning 40 years. From the early 1970's it operated a small but efficient Accelerator Group, attached to the ANSTO Physics Division. This Group assisted in the national promotion of ANSTO's accelerator based capabilities throughout all Australian Universities. It was mainly associated with research related to the use of the 3 MV positive ion Van de Graaff accelerator, the 10 MV Tandem accelerator, the 1.3 MV Van de Graaff electron accelerator and a 3 ns pulsed electron Febetron, together with a range of ion implantation and  $\gamma$  irradiation facilities. The Febetron went to Melbourne University, Department of Chemistry in 1988 and the 1.3 MV electron accelerator is currently on display in ANSTO's main reception centre.

During the mid 1970's physicists at ANSTO and AINSE picked up on the world-wide move towards the use of accelerator based ion beam analysis (IBA) methods. These IBA techniques were for the main part fast, non-destructive and able to be applied across an extremely broad range of scientific disciplines. This particularly suited both AINSE's role, with a need to be relevant to as broad a range of Australian universities as possible, and ANSTO's need at the time, to become more relevant to outside research groups including industry.

With the advent of significant new facilities at ANSTO throughout the 1990's the role of AINSE into the next millennium should continue to strengthen and the multiplier benefit to ANSTO of improved research linkages to all Australian Universities should continue to grow.

### 2. Facilities Associated with IBA

The initial focus for ion beam work within AINSE was on a 3 MV Van de Graaff accelerator. This High Voltage Engineering KN-3000 machine was purchased in 1963 and commissioned in January 1964 by the Australian Atomic Energy Commission (AAEC). In a letter to the then director, K. Alder, dated 11 April 1962, it was stated that this accelerator was needed for basic neutron cross section data, the study of subcritical assemblies using pulsed neutrons and as a source of monoenergetic neutrons that were relatively free of  $\gamma$ -rays for neutron dosimetry and health physics research.

Despite this emphasis on neutrons the accelerator was less than 2 years old when, in 1965, the first IBA experiments using (p,n) and  $(p,\alpha)$  reactions for analysis of metals and other materials were performed. The IBA work continued to grow rapidly through the 1970's, 80's and 90's, reaching a plateau in the late 1980's with up to 12 different operational beamlines. A total operating time on the 3MV accelerator of 50,000 hours was reached in January 1976 and 100,000 hours in October 1996. Currently, there are five functional beamlines and the accelerator operates 2,500 hours per year which corresponds to 10 hours per day Monday to Friday every week with AINSE groups at present using around 40% of the available time.

As early as 1979 ANSTO was looking at the feasibility of using mass spectrometry for <sup>14</sup>C and <sup>36</sup>Cl measurements for its Alligator Rivers and Artesian Basin Programs. Between 1982 and 1983 the Australian Quaternary Science community approached ANSTO for help to establish an accelerator mass spectrometry (AMS) facility in Australia. Between 1982 and 1988, when ANSTO purchased the Rutgers FN Tandem, considerable effort was put into obtaining a high energy accelerator facility. This included a visit to Seattle in May 1983 to look at the University of Washington's HVEC FN Tandem and a close inspection of the University of Texas FN accelerator in December 1986. Finally on 14 September 1989 the Rutgers FN Tandem arrived at ANSTO from the USA to start a new life in AMS and high energy IBA research. The FN series was designed for 8 MV on terminal using N<sub>2</sub> and CO<sub>2</sub> gases, with SF<sub>6</sub> the ANSTO Tandem reached 9.5 MV on terminal in September 1998 and has operated for more than 11,800 hours up to November 1998.

The first nuclear physics experiment on the new Tandem at ANSTO started in December 1991 it was proposed by University of Melbourne, Physics Department and funded by AINSE. Proton beams of 5 to 8 MeV were used to study the excitation function of the 6<sup>-</sup> excited state, corresponding to a 17.16 MeV excitation energy, in <sup>28</sup>Si. The results were published in Physical Review C [1].

The Tandem currently operates one IBA line, one high energy heavy ion microprobe line and three AMS lines with typically 25% of its current running time being used for high energy accelerator based IBA research.

### 3. People and Statistics

AINSE has a long tradition of interfacing external university researchers with ANSTO facilities. In the main this was done by dedicated people employed both by AINSE and within ANSTO. Table 1 shows the AINSE employees associated with AINSE's Accelerator Group from its formation in 1970 to the Group's absorption into ANSTO in January 1993. These people worked within and as an integral part of the ANSTO Physics Division. Today the IBA research aspects of the AINSE operations are carried out by ANSTO Physics Division staff and as figure. 1 shows this has not affected the number of accelerator based AINSE projects completed each year.

Personnel	Position	Year
Peter Lloyd	Technical	1970 - 76
Sid Kannard	Technical	1971 - 82
David Cohen	Professional	1976 - 89
Alex Katsaros	Technical	1983 - 91
Stan Newman	Professional	1989 - 90
Mike Hotchkis	Professional	1990 - 93
William Kyu	Technical	1992 - 93

**Table 1.** AINSE Acceleratorstaff from 1970 to January1993 when acceleratoroperations were transferred toANSTO, Physics Division.

Over the years there have been hundreds of university students, lecturers and professors come through the AINSE system to perform IBA related research. During the five year period 1985 to 1989, when records were kept, there were around 700 person day visits each year to ANSTO by researchers to use the accelerator facilities.

Since the shutdown of the 1.3 MV electron accelerator this number has been reduced by about 30%.

University researchers, through AINSE, have been involved in over 550 projects since 1964 related to accelerator based methods. Figure. 1 shows the annual distribution of these projects, starting with only 5 in 1964 and currently running at 36 separate projects for 1999 involving most Australian Universities. The 36 accelerator projects for 1999 include 28 on the 3MV and 8 on the Tandem accelerators. Some projects last only 12 months, many run over several years and some have been successfully producing for more than a decade. Since 1967, research on the accelerator facilities at ANSTO has produced 35 PhD and 8 MSc theses through AINSE. Since 1976 over 700 journal and conference research publications have been produced in collaboration with Australian Universities covering a broad range of scientific disciplines.



**Figure. 1.** The number of accelerator based AINSE projects completed each year between 1964 and 1999.

The range, depth and scope of AINSE's unique involvement in IBA research at ANSTO has produced significant benefits to Australian research community generally and considerably raised the profile of ANSTO.

### 4. IBA Methods

The growth in research associated with the applications of accelerator based IBA methods within Australian has been extremely well documented in the Proceedings of the ten national Conferences on Nuclear Techniques of Analysis. These conferences, held every two years, commenced in May 1976 and the eleventh one is due to be held at Lucas Heights in November 1999. They have been sponsored by AINSE, Australian Universities and ANSTO all of this time and hundreds of papers have been presented by students, lectures, professors and researchers. Most Australian universities have presented research at this Conference at some time.

It is interesting to note that at the first Conference in 1976 a range of IBA methods were presented, including; ion channelling, Rutherford backscattering (RBS), proton nuclear microprobes, particle induced X-rays (PIXE), particle induced  $\gamma$ -rays (PIGME) ion beam sputtering, ion implantation, forward recoil scattering (FRS), charged particle reactions and nuclear reaction analysis (NRA). The wide range of samples analysed included; bones, muscles, teeth, glass, rocks, soil, Th/U ores, coal, obsidians, air particulates, solar absorbers, wheat grains, silica and ceramics. At the last Conference in November 1997, some 22 years on, all of these research areas and

sample types were still well represented through a range of medium energy accelerators around Australia.



**Figure. 2.** The current IBA set up on the 3 MV Van de Graaff accelerator. It has 4 simultaneous legs for PIXE, PIGME, FRS and RBS analysis.

The current IBA beamline on the 3 MV Van de Graaff accelerator is shown in figure 2.

In more recent times the IBA field has been considerably expanded by the inclusion of both low energy (<100 keV) and high energy (>10 MeV) heavy ion facilities at various institutions around Australia. This includes the world class 10 MV heavy ion Tandem accelerator facility installed at ANSTO in 1989 which regularly produces 100 MeV iodine beams for elastic recoil detection (ERD) experiments. Also during the 1990's researchers at the ANU 14 UD accelerator have moved into more applied research using the IBA methods of accelerator mass spectrometry (AMS) and heavy ion recoil spectroscopy.

### 5. A Few Projects

As mentioned earlier there have been over 550 AINSE accelerator based projects over the last 35 years. It is obviously impossible to mention all of them. Instead we will mention just one or two key areas that hopefully will help reflect the broad range of disciplines and research topics covered by IBA methods.

#### 5.1. The First Proton Microprobe

In 1965, which was only the second year of operation of the 3 MV Van De Graaff accelerator, Physics Division had discussions with Materials Division and others which led to tests of several methods of proton microanalysis to determine concentrations of light elements in metals and other materials. One of these tests included the use of alpha particles from ( $p,\alpha$ ) reactions in oxygen irradiated with 0.86 MeV protons to determine the oxygen concentration in small spots at the surface of  $\alpha$ Ti-O alloys. By focussing the proton beam to 250 µm diameter and scanning the beam across the sample surface it was possible to measure the oxygen distribution in the metal sample. This work was aided by an MSc. student from UNSW. The first results were published in Nature [2] in 1966. Some measurements were also made with 100 µm spot sizes.

Thus was born the concept of a proton microprobe which has since been developed into a versatile analytical tool with dozens facilities installed throughout the world, a special dedicated international conference series, and a wide range of applications which extend into many scientific disciplines.



Figure. 3. The current high energy , heavy ion microprobe at ANSTO available to AINSE users. It produces spot sizes down to 5  $\mu$ m for ions such as p,  $\alpha$ , C, Cl, and I.

AINSE uses at ANSTO currently have access to a world class high energy, heavy ion microprobe on the 10 MV Tandem which produces spot sizes of energetic heavy ions down to about 5 µm diameter for IBA related research (see figure. 3).

#### 5.2. Archaeometry

In 1975 a collection of obsidian (a volcanic glass) from the ANU Department of Prehistory arrived at ANSTO for neutron activation analysis. Physics staff suggested that accelerator based IBA methods on the 3 MV Van de Graaff accelerator may be more appropriate. Early tests using  $(p,\gamma)$  reactions (PIGME) for F, Na and Al determinations were followed by later tests using (p,X) reactions (PIXE) for elements from Al to U. These PIXE/ PIGME methods were reliable, fast, sensitive and non-destructive for a broad range of archaeological samples including obsidian glasses[3] and pottery samples. Furthermore, they required no special sample preparation. The multi-elemental analysis capability of these techniques proved to be a tremendous advantage in distinguishing various sources of these materials and thus provided unique information to researchers on such things as historical trade patterns in the South Pacific. Figure. 4 shows the results obtained for early traders (3,000-11,000BP) in obsidian by sourcing various artifacts to particular volcanoes in the region. To date thousands of archaeological samples, including Aboriginal ochres, have been analysed by AINSE groups from around Australia and the work still continues today.



**Figure. 4.** Early trade patterns obtained from the multi-elemental IBA techniques on obsidian artifacts.

## 5.3. Biological

Some of the earliest biological research on the 3 MV Van de Graaff was performed in the early 1970's in collaboration with the University of Adelaide and the Waite Agricultural Institute using the  ${}^{12}C(p,\gamma){}^{13}N$  reaction as tracer to study the mechanisms employed by nitrogen fixing bacteria. A 200  $\mu$ A 2.5 MeV proton beam was used on a carbon target to produce the shot lived  ${}^{13}N$  isotope.

During 1978-79 studies on trace elements in human teeth in collaboration with Flinders University were commenced using PIXE. Trace elements in healthy and decayed enamel, dentine and cementum were compared. These experiments were particularly significant since, when published in 1981 [4], they showed, for the first time, that theoretical thick target PIXE yields could be successfully used to predict experimental yields in thick biological matrices. Prior to this many PIXE users, requiring quantitative analysis, were analysing thin targets only. Figure. 5 shows the theory/ experiment comparison in an apatite matrix for trace elements from Si to Sn taken from this work. The agreement was  $\pm 3\%$ . This success was the start of quantitative thick target PIXE analysis at ANSTO which continues today as a routine analysis facility with codes that were written being sold commercially worldwide.



**Figure. 5.** Theory/ experiment comparison for K shell X-ray yields for an apatite matrix for 2.26 MeV PIXE and a detector solid angle of 0.06 msr.

Over the years teeth and bones of all shapes and structures have been analysed. Between 1985 and 1987 in collaboration with Murdoch University the teeth of limpets, chitons and other marine invertebrates were studied using IBA methods. The mineral composition of the teeth of rock eating marine limpets was shown to have large concentrations of Fe (up to 30%) very early in their development phase. Experiments on the differences in trace elements in human teeth of children 150 years ago and today are still progressing in collaboration with Sydney University.

Well characterised neutron beams free of  $\gamma$ -rays are an important tool for radiobiological studies. The 3 MV Van de Graaff accelerator is an ideal tool for the production of medium flux neutron beams of known energies with low  $\gamma$ -ray contamination. In 1979, a facility, known as the Standard Neutron Irradiation Facility (SNIF), was established to provide neutron fluxes up to  $3x10^8$  ncm<sup>-2</sup>s<sup>-1</sup> at well characterised energies from a few tens of keV to 6 MeV. Dose rates of around 50 Gy/ hr were routinely produced from mono-energetic neutron beams of 5.4 MeV from the <sup>2</sup>D(d,n)<sup>3</sup>He reactions [5]. Dose rates up to 150 Gy/ hr were possible using a deuterium gas target at three atmospheres and deuteron beams of around 30  $\mu$ A with  $\gamma$ -ray contamination reduced to less than 5% of the total dose. From mid 1979 to the early 1990's the SNIF facility was extensively used by researchers from the University of

Queensland and Queensland Institute of Medical Research, producing several PhDs, MScs and journal publications related to DNA damage mechanisms in cells by MeV neutrons [6].

The field of biological research is well suited to accelerator based IBA methods and the long running International IBA Conference series always has a major section on biological applications.

## 5.4. Environmental

One of the longest running and most successful AINSE projects involves the Zoology Department of the University of Western Australia, AINSE, ANSTO and IBA methods developed on the 3 MV accelerator. It commenced in 1982 and involves the measurement of water turnover and carbon dioxide production in small lizards in the arid Pilbara region of Western Australia. The animals are captured, a blood sample taken then injected with 3mL/kg mixture of <sup>3</sup>H<sub>2</sub>O and 97% enriched H<sub>2</sub><sup>18</sup>O, marked with red acrylic paint and released at the exact site of capture. After about a week the same animal was recaptured and another blood sample taken. The difference in isotopic content of the two blood samples enables one to estimate the total H<sub>2</sub>O and CO<sub>2</sub> exchange and hence their metabolic rates. The technique of extracting small samples of water from blood and preparing thin well characterised Ta<sub>2</sub><sup>18</sup>O<sub>5</sub> targets for analysis via the <sup>18</sup>O(p, $\alpha$ )<sup>15</sup>N reaction was successfully developed at ANSTO [7]. The project is continuing today using nuclear reaction analysis on the 10 MV Tandem looking at nitrogen in muscles of small animals in the wild.

From the very first PIXE measurements done in Sweden in the mid 1970's it was appreciated that fine atmospheric particles collected on filter papers were ideal targets for characterisation by IBA methods. However it was not until the late 1980's and early 1990's that fine particulate atmospheric pollution became a significant public issue worldwide, generating a window of opportunity for IBA techniques to come to the fore. In 1989 ANSTO developed an Aerosol Sampling Program (ASP) through significant external funding. This was picked up, through AINSE, by environmental departments within Australian Universities and has resulted in several PhD and MSc theses and numerous publications. A particularly large study, using the ANSTO ASP facilities, was carried out by Griffith University during 1993-94 in the Brisbane area [8]. They used four simultaneous IBA methods of PIXE, PIGME, PESA and RBS to fully characterise 2.5 and 10  $\mu$ m diameter particles collected on filters. This work was done in collaboration with the Queensland Department of Environment and Heritage who today continue to commercially fund fine particle characterisation at ANSTO.

# 5.5. Materials

The unique contributions of accelerators to the characterisation of thin films, surfaces and interfaces has been appreciated internationally and national for decades. This aspect has been well documented in the International Conference on IBA series held every two years since 1973. The 5<sup>th</sup> IBA Conference was held in Sydney at UNSW in February 1981 with assistance from ANSTO and AINSE officers. The acquisition of the Tandem accelerator at ANSTO in September 1989 and attaining more than 7 MV on terminal by mid 1992 opened the possibility for Australian universities through AINSE to considerably expand the ion and energy range of IBA techniques available for materials research. One of the major long term international materials research collaborations on the Tandem to date involved RMIT, UNSW, ANSTO and the University Lund in Sweden. The total group involved more than 15 researchers from the two countries. It commenced in mid 1992 when the Swedes lent ANSTO their timing mirror system to perform heavy ion recoil time of flight (RToF) experiments using 77 MeV iodine beams. These beams were used to study a variety of thin metallised

films on GaAs and reactions in metal-III-V semiconductor systems in order to develop more stable electrical contacts to these materials. The first RToF spectrum was obtained at ANSTO in September 1992 using the Swedish mirrors, by June 1993 ANSTO had constructed its own timing mirrors based on the Swedish design. Figure. 6 shows the ANSTO RToF system attached to the IBA chamber in the Tandem target hall.



**Figure. 6.** The Recoil Time of Flight system attached to the heavy ion IBA leg on the 10 MV Tandem at ANSTO.

Between 1992 and 1998 the joint collaboration has produced more than 65 journal and conference publications (see for example Ref [9]) and six PhDs directly related to experiments performed on the Tandem RToF facility. The collaboration, through AINSE, is continuing looking at ferroelectric materials and studying heavy ion straggling and multiple scattering in thin films and interfaces.

More recently the RToF collaboration has been expanded to include Auckland University and the Institute of Geological and Nuclear Sciences (GNS) in New Zealand with research on solar cell surfaces and in March 1997 the first joint IBA experiments since Auckland University joined AINSE, were successfully completed. The collaboration continues to date.

# 6. Summary

Accelerator based (IBA) methods are ideally suited to the study of thin films, interfaces, surfaces and multi-layers. The large number of linkages and networks generated by AINSE and ANSTO officers, both nationally and internationally over the past two decades or so, demonstrates the extremely broad range of scientific disciplines into which these techniques have been applied. Essentially one is limited only by how laterally one can think!

The multiplying factor that operates between the facilities at ANSTO and the researchers in Australian universities through AINSE interactions is enormous. This is an efficient liaison and provides mutual benefits to all parties by considerably raising the research profile of ANSTO and by providing easy access and training to external researchers using ANSTO's unique facilities. The quantity and quality of accelerator based IBA interactions over the past 20 years is direct proof as to how successful such a partnership can be.

# 7. Acknowledgments

Throughout this short paper we have purposely avoided using individuals' names, since there have been too many to mention and it would be unfair to include just a few and to omit others. However, we would like to acknowledge the enormous help and support of all AINSE

Executive Officers, the AINSE Council and ANSTO Division Directors and staff between 1970 and 1993 when the AINSE Accelerator Group was functional and the much of the work discussed here was performed.

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# The Ever Expanding Field of Ion Beam Analysis

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The field of Ion Beam Analysis has steadily developed over the past forty yields to provide more detailed information for the analyst from a wider range of materials analysis tools. The first technique to be well developed was Rutherford Backscattering Spectrometry which relied on the elastic and inelastic scattering of H and He projectiles to provide composition and structural information in the near surface region. From this grew Ion Channelling which provided a valuable structural tool for the crystallographic location of impurities in solids.

The field expanded to include Proton Induced X-ray Emission (PIXE) and Nuclear Reaction Analysis (NRA) which pushed the detection limits beyond the range of RBS for most elements. Apart from different techniques the energy range was extended to develop Medium Energy Ion Scattering (MEIS, 100-500keV) which has been shown to probe the first 3-10 atomic layers with almost layer by layer resolution and Low Energy Ion Scattering (LEIS) which is exceedingly sensitive to the outermost one or two atomic layers.

By pushing the limits of techniques we can push the detection limits. This applies for MEIS and for recoils. The use of recoils over a wide range of energies provides a range of applications and versatile techniques. At the low energy end this technique has the flexibility to detect  $10^{-4}$  of a mono-layer of O on a clean surface and locate it crystallographically. At the high energy end it also allows multi-element depth profile analysis with a uniform detection sensitivity over half the periodic table.

The power and capacity of these scattering and recoil methods as well as future developments will provide an insight to the future expansion of techniques and applications.

# The Contribution of AMS to Geosciences

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The advent of Accelerator Mass Spectrometry (AMS) has heralded investigation of a variety of important problems in the geosciences, previously not considered possible for technical or conceptual reasons. Some of the advances in AMS methods have been made using the accelerators at ANSTO and the Australian National University.

Perhaps the best known of these techniques is the application of AMS <sup>14</sup>C dating which has the advantage of needing much smaller amounts of sample (typically  $\leq 1$ mg C), and having a potentially somewhat older age limit (to approximately 55,000 yr), than conventional <sup>14</sup>C determinations by  $\beta$  counting. AMS <sup>14</sup>C has been applied to dating an enormous array of materials including archaeological samples and sites, tree rings, ice cores, banding in coals and circulation and ventilation changes in the world's oceans.

An exciting application of the measurement of the rare long-lived isotopes <sup>10</sup>Be, <sup>26</sup>Al and <sup>36</sup>Cl is in the relatively new field of cosmogenic exposure dating. Accumulation of these cosmogenically produced nuclides formed in-situ in exposed rock surfaces is used to estimate both the time of exposure of the rock surface and mean erosion rates. A large variety of landscape-related processes have been successfully addressed including weathering and sediment-transport rates and the ages of glacial retreat, tectonic uplift and lava eruptions.

In the field of hydrology, <sup>36</sup>Cl studies of dissolved chloride have been used to successfully estimate the ages of ground waters and trace their origins. The tracing of atmospheric air masses that deliver rain and the origin of Australian salt lakes and continental salinisation using <sup>36</sup>Cl lead to important conclusions on the origin and residence time of chloride in the Australian landscape. The ultimate origin of the bulk of the surficial chloride in Australia is shown to be meteoric, and for the western part of the continent, a mean residence time of about 0.75 Ma pertains. The realisation of the long-term and continuing delivery of salts to the landscape needs recognition in planning strategies to combat salinisation of agricultural areas.

# AMS at Lucas Heights: past, present and future

Claudio Tuniz

Australian Nuclear Science and Technology Organisation

"On June 23, 1989, a huge crane picked up a large yellow tank out of the bowels of the earth where it resided for twenty-five years, and placed it on a large platform truck ..." [1]

# The past

Accelerator Mass Spectrometry (AMS) [2] was first applied to radiocarbon analysis in 1977 [3] and soon extended to the analysis of other rare radionuclides of cosmic origin. Lucas Heights researchers perceived since the early days the importance of long lived cosmogenic isotopes, such as <sup>36</sup>Cl (301 ka) [4], and hence the value of the AMS method for applications in ground water studies [5] and in radionuclide migration near uranium ore bodies. Collaborative projects involving the Australian Atomic Energy Commission (AAEC), the Bureau of Mineral Resources (BMR) and US institutions were developed for applying <sup>36</sup>Cl to study the hydraulics of the Great Artesian Basin. In absence of an Australian facility, AMS measurements were made at the University of Rochester [6]. A <sup>36</sup>Cl dating capability based on the 14UD accelerator in Canberra was developed a few years later, in a collaboration between AAEC, Australian National University and CSIRO [7]. AAEC researchers were also involved in the early studies aimed at evaluating the potential of <sup>129</sup>I (16 Ma) as an environmental tracer [8].

During the 1980's Lucas Heights scientists attempted several avenues to acquire an AMS facility for fulfilling the pressing needs of the quaternary science community [9]. This objective was achieved only in 1989, when ANSTO was advised on the availability of the 8-MV Rutgers tandem [10]. This accelerator, model FN, serial number one and nicknamed the *King*, had been used for 25 years at the Nuclear Physics Laboratory of Rutgers University in New Jersey, in measurements of nuclear lifetimes, time reversal violations, studies of forbidden isobaric analog resonances and in many other nuclear physics studies. The *King* had been also utilised in the first AMS studies of extraterrestrial materials. One of the early <sup>10</sup>Be experiments performed on this tandem helped to unravel the origin of Australian tektites [11], a sign of the destiny for the Rutgers accelerator, re-born several years later on the Australian continent as ANTARES, the Australian National Tandem for Applied RESearch [12].

# The present

### Status of the facility

The transformation of the Rutgers tandem into a high-performance AMS spectrometer required the installation of several new components, including: spirally-inclined accelerator tubes, Pelletron charging system, gas stripper, 60-sample ion source and fast sequential isotope system. Purpose-designed beamlines for the detection of <sup>14</sup>C, <sup>10</sup>Be, <sup>26</sup>Al, <sup>36</sup>Cl, <sup>129</sup>I have been constructed using high resolution selectors for momentum, energy and velocity and versatile detector systems [13].

A facility designed for the detection of heavy radionuclides, such as <sup>236</sup>U, <sup>229,230</sup>Th and <sup>244</sup>Pu, in natural samples, has been recently added to the suite of ANTARES beamlines. An electrostatic quadrupole doublet on the high-energy end of the accelerator provides mass

independent focussing of the heavy-mass beam. Momentum and E/Q analysis is performed with a high-rigidity double focussing magnet (mass-energy product = 250 MeV.amu) and a high-resolution double focussing electrostatic analyser (E/Q = 7.6 MV and energy dispersion of 5000 in the image plane).

The chemistry laboratories for target preparation are an integral part of the ANSTO AMS facility. The current <sup>14</sup>C accelerator and chemistry backgrounds correspond to 50 and 60 ka BP respectively. With the increasing demand for measurements of extremely small samples, ANSTO developed methods for the preparation of targets containing as little as 10 micrograms of carbon. During the last 12 months the capabilities of the chemistry laboratories were expanded to encompass the preparation of <sup>10</sup>Be, <sup>26</sup>Al and <sup>36</sup>Cl from a variety of environmental samples. Procedures for the extraction of iodine, uranium and plutonium from water, sediments, soils and biota are being developed for the nuclear safeguards program.



# The scientific program

### **Quaternary Science**

A national research program in Quaternary science based on the AMS analysis of <sup>14</sup>C, <sup>10</sup>Be, <sup>26</sup>Al, <sup>36</sup>Cl and <sup>129</sup>I started at Lucas Heights in 1993. This program, funded by the Australian Research Council and AINSE, has involved collaboration with 36 Australian universities and AMS analysis of nearly 3000 samples from more than 100 projects covering a wide range of disciplines: archaeology, geomorphology, hydrology, palynology, glaciology and paleoclimatology. Some selected projects are described below to give the flavour of this research program.

### Carbon cycle and palaeoclimatology

<sup>14</sup>C was used to give the essential framework for interpreting information locked in the ancient dwellings of stick-nest rats which are built in arid areas from local materials and preserve plant and animal material for up to 10 ka. The material found in nests from Western Australia, Northern Territory and South Australia includes bones and hair from extinct species and pollen from plants growing thousands of years ago in the arid centre of the continent. AMS dating was used to reconstruct the past distribution of Australia's arid zone mammals and provide much needed information about the last few thousand years (*Universities of Western Australia, Wollongong and Newcastle*).

Ancient conifer logs exhumed from the bed and banks of the Stanley River in western Tasmania have been sampled for dendrochronology and atmospheric <sup>14</sup>C studies. They range in age from 35 ka to the present. High precision <sup>14</sup>C measurements were performed on 90 consecutive, single-ring samples from a celery-top pine log dating from *ca*. 12,700 <sup>14</sup>C years BP. This high-resolution annual record of atmospheric <sup>14</sup>C provides insights into production variations and ocean CO<sub>2</sub> release during the last glacial transition (*University of Sydney*).

Analyses of <sup>14</sup>C bomb-pulse curves in tree rings from tropical regions and the southern hemisphere help also to improve our understanding of the carbon cycle and air-sea interactions, important processes for the global climate [14, 15]. (University of Sydney and Monash University)

### Geomorphology and landscape evolution

Long-lived cosmogenic radionuclides produced *in-situ* in exposed rocks and surfaces can be used as geochronometers and tracers of various geochemical and geophysical surface modification processes. A measurement program of in-situ radionuclides relating to the Australian and Antarctic environments has been supported by AINSE. An example from this program follows.

The Victorian Plateau region of Western Australia contains extensive upstanding coastal reliefs of sand regoliths overlying sandstone bedrock. The formation process of this landscape may be explained either through an internal chemical modification process (laterisation of pre-existing rock) or contrastingly, a deposition process of external material onto bedrock. The time scales for these two processes are believed to be quite different. *In-situ* <sup>10</sup>Be profiled from surface to bedrock has been able to distinguish between the alternatives and provide information of deposition rates (*Murdoch University and University of Western Australia*).

#### Archaeology

Several archaeological projects based on radiocarbon dating have been funded by AINSE and ARC. These projects include detailed research into the probable antiquity of Australian rock art. A variety of materials have been analysed, including pigments, oxalate minerals, silica coatings, plant fibres, carbonised plant matter, fatty acids, beeswax and mud-wasp nests. Different sample processing techniques have been explored in these studies including low-pressure plasma techniques and laser extraction methods. Intercomparisons between <sup>14</sup>C dating and other dating techniques such as thermoluminescence (TL) and optically stimulated luminescence (OSL) have been carried out within some of the aforementioned projects. Some examples from this program follow.

The Chillagoe region of North Queensland has been under intensive archaeological scrutiny since the early 1980's. To date, a series of excavations have revealed cultural deposits dating back to at least 26,000 years BP, with dramatic increases in the quantities of archaeological materials recovered during the mid to late Holocene. In order to test the chronological model constructed via indirect evidence, the ANTARES laboratories dated several charcoal paintings and drawings from Chillagoe. The Chillagoe pictographs show ages of 2000-3000

a, in support of the existing model of a mid to late antiquity for most of the regionalised art of the region [16] (*University of Queensland*).

The Kimberley rock art sequence is likely to be one of the longest and most complex in the world. On the basis of superimpositions and differential weathering, a very detailed rock art sequence has been reconstructed. This sequence depicts major changes in Aboriginal culture, ideology and local fauna over time. Fieldwork in the region started in 1994 with the aim of providing absolute dates for the Kimberley rock art sequence. Small samples of pigments, beeswax and associated mineral crusts have been collected. AMS dating of these samples has provided the first age estimates for the well-known Bradshaw painting style. Mudwasps, which overlie or underlie Kimberley rock paintings, can be dated by OSL, providing minimum or maximum ages. Radiocarbon analysis of the pollen allowed comparison between results from two very different dating techniques [17] (*La Trobe University*).

Ngarrabullgan Cave, in north Queensland, is one of the earliest radiocarbon dated archaeological sites in Australia (37,000 yr BP). The deposits at this site show very low erosion and a near-total absence of territorial vertebrate fauna. Our study shows that intensive use of the mountain started around 5000 yr BP, after 27 millennia of total abandonment. In this work we have also obtained the first paired <sup>14</sup>C/OSL determination for pre-30 ka archaeological deposits [18] (*University of Queensland*).

AMS was used to date the carbon bearing substances contained in a microstratigraphic sequence deposited on a bolder in a limestone rock shelter near Chillagoe, north Queensland. A time span of 30 ka was discovered in a rock surface accretion of about 2 mm. These results demonstrate that that materials such as oxalate crusts and silica skins could provide new environmental archives for palaeoclimate studies. This method was originally developed to identify ancient aboriginal rock art concealed within encrustations [19] (*James Cook University*).

The radiocarbon dating of charcoal fragments from the rock shelter at Jinmium in the Northern Territory demonstrated that this site was occupied by Aboriginal people only during the Late Holocene [20] (*La Trobe University*).

# Global Climate Change

### Long-lived radionuclides in Antarctic ice

Antarctic ice cores provide the best source of preserved air from which to reconstruct levels of greenhouse gases over recent centuries to millennia. One of the problems is that recent  $CO_2$  growth variations are difficult to interpret due to the smearing of ice-core signals induced by the diffusion of air in the firn. To calibrate the CSIRO models describing the diffusion of air in ice we used the <sup>14</sup>C "bomb spike" to determine the age spread and age of  $CO_2$  in Antarctic ice and firn [21] (*with CSIRO and Australian Antarctic Division*).

#### **Southern Hemisphere Glaciation**

The objective of this project is the application of in-situ cosmogenic radionuclides <sup>10</sup>Be, <sup>26</sup>Al and <sup>36</sup>Cl to the study of glacial histories during the Quaternary in the Southern Hemisphere. Over 50 <sup>10</sup>Be and <sup>36</sup>Cl cosmogenic exposure ages have been already determined by AMS in erratic, moraine and bedrock samples from sites in New Zealand, Tasmania and Antarctica. Exposure ages range from 15 to 20 ka at young sites associated with the Last Glacial Maximum in Tasmania to the extremely old exposed surfaces of 1 to 2.3 Ma at the Southern Prince Charles Mountains in Antarctica. The suite of exposure ages from Tasmania appear to be generally consistent with periods of glacial maxima inferred from the oxygen isotope record, though a number of differences have generated interesting inferences. The Antarctic
ages apparently are younger than anticipated and lend support to the model of a dynamic and recently active East Antarctic Ice sheet [22] (*with University of Tasmania, La Trobe University and University of Auckland*).

#### Ultra-sensitive monitoring of nuclear activities

Nuclear activities introduce into the environment long-lived radionuclides such as <sup>129</sup>I and <sup>36</sup>Cl [23]. AMS is the analytical technique of choice for the analysis of these radionuclides in natural specimens. Isotopic concentrations of 10<sup>6</sup> atoms per gram can be detected in samples taken from a variety of environmental materials such as water, air, soil and biota. The ANSTO AMS group has analysed <sup>129</sup>I in waters and sediments collected by IAEA inspectors at various distances from a nuclear reprocessing plant. ANSTO researchers have also analysed <sup>129</sup>I, <sup>14</sup>C and <sup>36</sup>Cl in water specimens from the Mururoa lagoon, contributing to an international project aimed at determining the environmental impact of the underground nuclear tests in the Pacific atolls [24].

#### Biomedicine

AMS provides a method for analysing long-lived isotopes of elements for which metabolic and toxicological information is not available. Aluminium, for example, is now considered to be a toxic element, whose accumulation has been identified as the cause of disease states in chronic renal failure patients. Detection via AMS of the long-lived radioisotope <sup>26</sup>Al administered at ultra-trace levels and thus with negligible radiation damage can provides a new avenue to understand the role aluminium plays in biological systems. The ANSTO AMS group has detected the presence of <sup>26</sup>Al in the brain tissue of Wistar rats gavaged with drinking water containing 70 Becquerel doses of <sup>26</sup>Al [25].

#### The future

Applications of AMS methods in quaternary and environmental science at ANSTO will expand in the foreseeable future with <sup>14</sup>C, <sup>10</sup>Be, <sup>36</sup>Cl and other radionuclides contributing to a growing body of knowledge on processes involving atmosphere, oceans, ice sheets, biosphere, soils and sediments. The AMS program based on *in-situ* produced radionuclides is expected to expand from glacial chronology to landscape evolution of the Australian continent during the Quaternary. One example is the study of complex processes such as the transport and deposition of eroded material to understand the evolution of desert sands. Less traditional applications of environmental <sup>14</sup>C will involve the identification of synthetic additives in food products or wine.

Biomedicine is set to become a major component in future AMS applications at Lucas Heights. The most significant advantage of AMS in biomedical applications is that the biological response to toxins can be obtained at environmentally relevant dose levels. In fact, AMS enables the detection of carcinogens labelled with <sup>14</sup>C at concentrations of one attomole per milligram of body tissue. Furthermore, the use of long lived radionuclides make negligible the radiation hazards to subjects and researchers. For example, the metabolism of calcium and aluminium in humans can be studied using <sup>41</sup>Ca and <sup>26</sup>Al, respectively. The capability recently achieved at ANTARES for the AMS detection of actinides will offer new opportunities in studies related to the toxicology of plutonium.

#### Summary

The ANTARES AMS facility presently provides an essential resource for much of the Australian research on climate, environment, national heritage and archaeology. The enormous potential for applications of AMS in detecting long-lived tracers in fields such as toxicology and pharmacokinetics has not been yet exploited. AINSE could have a key role in promoting the use of AMS in biology and medicine.

#### **Acknowledgments**

A world-class AMS facility is now available at Lucas Heights thanks to the foresight and perseverance of people like Roger Bird and John Boldeman, the skilful work of John Fallon and his group and the generosity and enthusiasm of the ANSTO AMS scientists, including David Fink, Ewan Lawson, Michael Hotchkis, Geraldine Jacobesen and Andrew Smith.

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### **Evolution of materials Research Within the AINSE Portfolio**

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The main materials research interactions between ANSTO/AAEC and the AINSE member universities are reviewed and linked to the main thrust of contemporary ANSTO/AAEC programs.

The AINSE portfolio encompasses the previous AAEC research contracts, which represent an earlier example of public sector outsourcing, until re-discovered during the present decade, as well as AINSE studentships and Research and Training Projects. Collectively these mechanisms did much to foster the maintenance of effective materials research teams in Australian universities. Selective examples will illustrate the success of the AINSE family in training to "help provide engineers and scientists of high ability for the future".

#### The National Centre for Neutron Scattering

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#### Abstract

As neutron scattering can be done nowhere else than at Lucas Heights in Australia, both ANSTO and AINSE have been heavily involved in its support for the past forty years. This paper traces the involvement of AINSE, highlighting significant developments in instrumentation and organisation. The prospect of a replacement reactor promises a new era for neutron beam science in Australia. I will outline the present plans with emphasis on the opportunities for new science for participating universities.

Neutron scattering, X-ray photon scattering and electron scattering are the three main particle probes of condensed matter. The neutron, because it has zero charge, can penetrate materials and reveal truly bulk behaviour. Neutron scattering has been a powerful tool especially for the basic scientific understanding of bulk condensed matter. Not only has it exposed atomic and magnetic structures but it has revealed the whole gamut of atomic and magnetic dynamics which are inaccessible by any other technique. The enviable record of neutron scattering, and its continuing usefulness, in the science of materials has overshadowed its achievements in the technology and development of materials where it also has some notable successes. That is why thousands of scientists visit reactor and spallation sources of neutrons each year world-wide.

#### Introduction

The centre for neutron scattering in Australia is at Lucas Heights and its form has been shaped by the Australian Atomic Energy Commission (AAEC), latterly the Australian Science and Technology Organisation (ANSTO), by AINSE, by the Australian Research Council (ARC) and by all the participants and their organisations. These have all contributed to "The National Centre for Neutron Scattering"; for that is what it is, and what it has been for almost forty years.

In recounting the history of the National Centre for Neutron Scattering I am mostly guided by historical method of '*1066 and all That*' which, if you remember, says "history is what can be remembered". Well, this is what I can remember and what I can remember to find.

#### Prehistory, 1958-60

AINSE was inaugurated on the 4<sup>th</sup> of December 1958. The new organisation was about national sharing of the facilities of the AAEC, particularly by the universities.

From the beginning AINSE requested applications for research grants and provided credits for the use of Lucas Heights facilities and for travel and accommodation. In the case of neutron scattering AINSE was unique in this activity. I remember a similar but more restricted scheme operating at Harwell in the late 1960s, initiated, I believe, by their knowledge of the Australian operation. The first service institute for external use of neutron beams was the Institut Laue-Langevin in Grenoble which was first operational in the early seventies.

#### Startup, 1960-64

HIFAR attained full power during 1960. The famous powder diffractometer based on a naval gun mount was operational in 1961 along with a single crystal diffractometer. The group leader was Terry Sabine who had returned from gaining experience with British neutron scatterers at Harwell. He was joined by David Browne, imported from the Metallurgy Division at Harwell, and Suzanne Hogg as computer programmer, as well as two technicians.

The first crystal structure study, which was of *p*diphenylbenzene, involved collaboration between the University of Western Australia, via AINSE, and Terry Sabine. It was published in 1961 in *Nature* under the names of Clews, Maslen, Rietveldt and Sabine [1]. Apart from being the first neutron diffraction study out of Lucas Heights, it was also significant because of the association with Rietveldt, then a PhD. student at UWA, whose least squares refinement of powder neutron and X-ray diffraction data Terry Sabine was later to promote as "The Rietveldt Method", and which remains today as the premier analysis tool for powder diffractionists.

The collaboration with AINSE sponsored institutions not only involved provision of neutron diffraction facilities to the universities but in small measure equipment and effort were supplied by universities and university personnel.

An early example of this is provided (figure 1) by the simple variable temperature liquid nitrogen cryostat for the powder diffractometer, designed by me and made by the Department of Physics workshop at Monash University [2]. Later Gordon Cox, who had recently joined the neutron group, and I assembled the first primitive triple axis spectrometer on HIFAR by the simple expedient of mounting a single crystal diffractometer (6H, figure 2).



Figure 1. Liquid nitrogen cryostat designed and made for the neutron powder diffractometer in the Department of Physics, Monash University in 1963..

I remember spending a night with this beast manually resetting the angles after each counting period. In addition to the second powder diffractometer there was a variable wavelength total cross section spectrometer associated with the name of Brian Hickman built for the specific purpose of looking at defects in BeO, at that time a favoured material for prospective power reactors in the Australian context. I understand that it played a significant role in the rejection of BeO for that purpose.

#### Expansion, 1964-73

AINSE took the decision to put troops on the ground. Frank Moore, a former D.Phil. student of the famous Oxford crystallographer, Dorothy Hodgkin, was appointed as leader of the new group. David Wheeler, with considerable equipment and scientific experience from Harwell, and two technicians, Peter Lloyd and Roy Ebdon were also appointed. The purpose of the new group was to expand the suite of equipment available for AINSE users.



Figure 2 –Powder spectrometer converted to triple axis spectrometer

This included much needed ancillary equipment such as magnets and cryostats as well as a further single crystal and a polarised neutron diffractometer.

A small angle spectrometer for the measurement of excitations (magnons) in ferromagnetic materials was also built. Figure 3 shows Frank Moore and David Wheeler with the original single crystal and gun mount powder diffractometer. Note that the powder diffractometer, which overhangs the spent fuel rod storage sports a new electromagnet.



Figure 3 Frank Moore and David Wheeler working on 4H1 powder diffractometer, based on a naval gun mount, at Lucas Heights

Meanwhile there were further appointments and an instrument for the AAEC group. The new triple axis spectrometer (figure 4) was the most ambitious instrumental development at Lucas Heights at that time. Triple axis spectrometers were the workhorses for the measurement of all types of excitations in condensed matter the measurement of which is the province of neutron scattering alone. Arthur Pryor, from the AAEC and Margaret Elcombe, from Cambridge joined the AAEC group particularly for this instrument. Arthur Pryor was later to continue his interest in lattice vibrations by joining the laser uranium enrichment program. Chris Howard joined the group and Gordon Cox left for a computing career.

This period also saw the initial development of LONGPOL (the long wavelength polarisation analysis diffractometer). At that time it was one of only three instruments worldwide with polarisation analysis capability and the only one on a medium flux reactor. While the concept

came from Monash University, the building was done by David Wheeler and the AINSE group. Figure 5 shows the early instrument for which the detector setting was not automatic but had to be pushed from angle to angle. This instrument's first task was to directly observe

the glassy nature of the magnetic structure in the archetypal spin glass Cu-Mn, something which the instrument at ILL was not capable of at the time.



Figure 4. The triple axis spectrometer sometime in the seventies.



Figure 5. The original LONGPOL in about 1974.

#### Struggle, 1973-86

At this point Terry Sabine left the AAEC to join the New South Wales Institute of Technology which was to become the University of Technology Sydney (UTS) and David Wheeler left for the Institut Laue-Langevin. Cliff Ball was appointed as Leader of the X-ray *and* Neutron Diffraction Group and Lindsay Davis was appointed to assist Frank Moore. While the AINSE group maintained its it strength of four (including two technicians) that part of the AAEC group still doing neutron scattering shrank to two (Margaret Elcombe and Chris Howard with no technicians). From 1977 to 1980 Chris Howard was the only AAEC staff member doing neutron scattering as Margaret Elcombe was in administration. At that time the AINSE group outnumbered the AAEC group 4 to 1.



Figure 6. The second LONGPOL on a higher intensity hole with the curved scavenged shielding.

AINSE was still able to develop LONGPOL which after the demonstration of its usefulness needed more intensity to maintain its competitiveness. There was no money for new shielding so Bill Palmer and I scoured the junkyard near stores to find a piece of discarded shielding which could serve. That found, the AINSE group carved out a space for the monochromator assembly and punched through a new hole below the original for the exit beam. Such was the improvisation of that time. Figure 6 shows LONGPOL installed on the new more intense beam with the scavenged shielding.

In this period one of the most important instruments was commissioned on HIFAR. That is the High Resolution Powder Diffractometer (HRPD). For its construction, Chris Howard relied very heavily on the AINSE group because of the technical backup that group had. Figure 7 shows a mid-eighties picture of HRPD. The power of HRPD is illustrated in Figure 8 which shows the diffraction pattern from a composite powder of several minerals. Unlike X-ray patterns, for which the peak intensity falls off with angle, neutron patterns have peaks of similar intensity at all scattering angles. Because of this more peaks can be collected as long as they can be resolved at high angles. The pattern in Figure 8 shows that with HRPD peaks can be resolved up to 160° scattering angle. The line through the pattern is the "Rietveldt" fit which allows the relative contributions to the total diffraction pattern to be quantified.



Figure 7. HRPD, looking into the detector bank with specimen in the centre, in the mid 80s.

The history of this period would not be complete without a mention of The Australian Neutron Beam Users' Group (ANBUG) representing Australian neutron scatterers (about 80 including some marginal users and some overseas). During the late seventies and the eighties ANBUG lobbied for improved facilities and staffing for neutron scattering at Lucas Heights. In addition there was a realisation that for neutron scattering to be useful in new areas, including soft matter and applied areas, the spectrum of neutrons available had to be widened towards longer wavelengths. This had already been done on all HIFAR's sister reactors overseas. This resulted in ANBUG's parallel push for a cold source in HIFAR.

ANBUG tried to influence the AAEC and prepared submissions to government agencies. One of these was a submission to The Australian Science and Technology Council (ASTEC)

which in its 1985 report `Nuclear Science and Technology in Australia' recommended that \$10m be spent on "HIFAR refurbishment and neutron beam instrumentation". Meanwhile ANBUG felt itself a strong enough organisation to host the International Neutron Scattering Symposium, a satellite of the International Crystallography Congress, in Sydney in 1987.



Figure 8. Diffraction pattern for a mixture of minerals with Rietveldt fit which makes phase analysis possible.

#### Looking up, 1986-97

Things have looked up in the last ten years or more. It is hard to pick the exact point because it all happened so gradually. Certainly Margaret Elcombe coming back to the group in the early 80s was important as was the decision of The Australian Nuclear Science and Technology Organisation (ANSTO) and AINSE to proceed with The Australian Small Angle Scattering Instrument (AUSANS) at the end of that decade. It seems to me however that about 1986 was significant for the turn around because there was a lot of discussion in the AAEC about new instruments and the refurbishment of old. Much of this was prompted by the ASTEC report. Unfortunately the amount recommended would not cover the cost of a cold source. But still AAEC/ANSTO and AINSE money flowed into refurbishing and enhancing instruments.

ANSTO also increased the number of personnel in the ANSTO group at various times with interfacing and computer technicians. After the amalgamation of the two groups ANSTO continued to increase the number of personnel, adding some scientific positions.

AINSE has, in this period, been able to assist with further capitalisation of the neutron instrumentation through its access to Australian Research Council (ARC) funds. Previously some ARC funds had flowed to particular instruments via personal ARC funding to

individuals in universities but with the advent of the Infrastructure Grants, AINSE, as a majority university organisation, was able to tap these funds for the development of neutron instrumentation for university use. This has helped ANSTO in its role as supplier and operator of the neutron scattering instrumentation.

#### New horizons, 1997-

During 1997 Federal Government plans for a replacement research reactor were announced. Broad specifications indicated that the reactor envisaged would be similar to that proposed to the Research Reactor Review by ANSTO a few years previously. The neutron scattering community was delighted. There had been some concern that if a replacement reactor were approved it would be a token reactor to serve the public relations needs of the government and useless for neutron beam work.

Broadly, the replacement reactor is expected to have a flux two or three times that of HIFAR, have a cold source, guide tubes and associated hall. Provision is included in the budget for a substantial suite of instruments and ancillary equipment. Significantly ANSTO has consulted widely in the scientific community and has formed a Neutron Beam Facilities Consultative Group representing scientific associations, CSIRO, AINSE and industry. The group has advised ANSTO on the range of science and technology appropriate for the new neutron source and the suite of instruments necessary. The group has also specified the level of staffing required to operate the new facilities effectively. This certainly promises to be a new era in Australian neutron scattering and will hopefully bring Australian facilities up to the standard of other medium flux reactor sites.

#### The future

I have outlined the various influences which have shaped 'The National Neutron Scattering Centre'. In the long run, if maximum effectiveness is to be obtained from the projected replacement facilities, these influences need to be recognised. The Neutron Scattering Group has always been buried deeply in the AAEC/ANSTO organisation and this has made it vulnerable. The AINSE group, while it existed, was much more stable and was vital for the survival of neutron beam work in Australia. ANSTO has made a fine start with the wide consultation it has initiated via the Beam Facilities Consultative Group. It would be excellent if an organisational structure for neutron scattering in Australia which recognises the inevitable wide interest in the technique is established.

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### Neutron Scattering in the 21st Century

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#### Abstract

The watchwords for neutron scattering in the 21st Century will be complementarity and computing. Despite the enormous impact that is coming in condensed matter science, chemistry and biology from synchrotron light sources, the essential complementarity between X-ray and neutron scattering is likely to be not only preserved but enhanced. This will be achieved by current and foreseen investments in new neutron sources of high intensity and by technique developments involving neutron polarisation, neutron optics and neutron detection.

The complementary relationship between neutrons and X-rays - especially using isotopic and spin polarisation contrast will be illustrated along with some challenges which might well be met in the next ten to 15 years.

#### *Complementarity*

The structure of films and interfaces is of fundamental interest in areas as diverse as polymer science, adhesion and lubrication, colloid and emulsion science and the operation of biological membranes and the new generation of sensors which mimic them. This structure can now be determined at molecular resolution from specular reflectivity measurements using neutrons and X-rays. The two methods are complementary, the X-ray method from the tremendous intensity now available from synchrotrons and the potential for anomalous contrast. The neutron method is indispensable because of the contrast variation available for hydrogenous systems by H/D isotopic replacement. Recent reviews of these rapidly advancing areas of research are available in the literature (eg. Dietrich and Haase 1995, and references therein; Als-Nielsen *et al.* 1994; Thomas 1997)

Australia has a significant stake in the newly commissioned neutron reflectometer known as SURF at the Rutherford Appleton Laboratory in the U.K. as well as easy access to the higher-resolution instrument known as CRISP. At the Research School of Chemistry, Australia's first X-ray reflectometer for liquid surfaces is operating and through the Major National Research Facility access to the Consortium for Advanced Radiation Sources (CARS) at the world's most powerful synchrotron, the Advanced Photon Source (APS) in Chicago X-ray reflectometers for high intensity and time resolved studies are planned. We thus have access to the essential complementarity at the world's best radiation sources.

This paper gives examples of that complementarity and where the neutron reflectivity measurements were decisive for determining film structure.

#### THE STRUCTURE OF THIN FILMS AT INTERFACES

#### Polydispersity and Respreading

The vast majority of our current knowledge of thin films at the air-water interface is based on the interpretation of surface pressure-area ( $\Pi$ -A) isotherms measured on the Langmuir trough.  $\Pi$ -A isotherms for simple short-chain surfactants generally show features characteristic of two-dimensional gaseous, liquid and solid phases with smooth transitions between them. At sufficiently high surface pressures, monolayer films of such materials will buckle and collapse to form multilayer structures.

The two isotherms of Figure 1 differ in that for one of them the ( $\Pi$ -A) cycle is of small area (Polyisoprene-OH) and the other, (Polystyrene-OH), shows extensive hysteresis upon compression and expansion (Saville, P. M., Gentle et al. (1994). The first is said to respread quickly the other not. This process is very important in biology and industry. For example the surfactant mixtures in the lung must respread quickly as the lung surface area increases upon inhalation. A  $\Pi$ -A isotherm like that of the polystyrene surfactant would result in respiratory failure whilst something like that for the polyisoprene would work well. Nature achieves this by mixing surfactants but the structural correlates of this behaviour are at present obscure.



Figures 1  $\Pi$ -A Isotherms (a) and Structural formulae (b) for polyisoprene-OH and polystyrene-OH surfactants at the air-water interface.

The neutron reflectivities from a perdeutero-polyisoprene-OH film are shown in Figure 2, and illustrate the change in fringe pattern with relative contrast to the substrate. Measurements at different contrasts are essential to constrain the model fit to the data (Saville, P. M., Gentle et al. (1994).

#### Some Hindrances to Respreading

The behaviour of the polystyrene surfactant (PS-OH) with an average molecular weight of 973 g.mol<sup>-1</sup> whose  $\Pi$ -A isotherm is shown in Figure 1 illustrates some of the time dependent phenomena involved in respreading. To establish structures at equilibrium, neutron reflectivity profiles were measured at a number of surface pressures for fully deuterated PS-OH spread on two different subphases: D<sub>2</sub>O and air contrast matched water (ACMW, a mixture of D<sub>2</sub>O and H<sub>2</sub>O with scattering length density the same as for air).



Figure 2 Neutron reflectivity vs momentum transfer perpendicular to the surface, Q, for a polyisoprene surfactant film on air contrast matched water and on  $D_2O$  showing substrate contrast effect.

Modelling of the data indicated that the film is initially composed of a monolayer of surfactant with a thickness of 13.3Å at a surface pressure of 0.5mNm<sup>-1</sup>. As the film is compressed to higher surface pressures it becomes thicker as the polymer chains are forced to become closer packed and more linear in conformation. At a surface pressure of 10mNm<sup>-1</sup> the film ruptures and undergoes a structural transition to a monolayer overlaid by islands of trilayer. When the reflectivity profiles were fitted for time intervals after the initial compression and the thicknesses of the various layers calculated, it was found the trilayer hydrated over a period of about 100 minutes (Figure 3). Faster kinetic phenomena on the time scale of seconds to minutes have also been identified(Saville, P. M., Gentle et al. (1994). These will be studied using X-ray reflectometry in the millisecond reflectometer (White *et al.* 1999) instrument proposed by us for the Chem Mat CARS beamline at the APS. Neutrons will never achieve such a short time scale – once again we see complementarity.

#### **Chemical Reactions at Interfaces**

The announcement (Beck *et al.* 1992) that mesoporous materials with internal structures in the tens of nanometre scale could be produced by using the liquid crystal mesophases of surfactants as templates for was a major step upward in materials science. The scale of structures that could be induced by templates was increased by a factor of ten. Recently the effect of constraining growth to a surface - the air liquid interface, has produced excellent films whose growth mechanism has been revealed by the combined application of X-ray and neutron reflectivity (Brown *et al.* 1997; Brown *et al.* 1998). These films have considerable potential for practical applications but also fundamental interest in relation to the mechanisms of biological mineralisation.



Figure 3 Time dependence of the thickness of the three layers in the model for the polystyrene surfactant film after trilayer formation,  $d_3$  head group layer in water,  $d_2$  bilayer region,  $d_1$  diffuse top layer.

Such films may be grown (Aksay *et al.* 1996; Brown *et al.* 1997; Yang *et al.* 1998) by slowly hydrolysing tetra ethoxy silane in the presence of an aqueous surfactant solution at about ten times the critical micellar concentration. The kinetics shows a prolonged "induction phase" where there is no visible film growth for five to ten hours and then a fairly rapid growth of film to thicknesses of a few microns. Figure 4 shows typical X-ray data in the induction period and to the point where observable Bragg peaks arise from the growing layer structure.

In the "induction phase" (a, b) the reflectivity is clearly modified from that of water or a surfactant solution of the same strength as that of the cetyl trimethyl ammonium bromide used for the preparation. The bump in the reflectivity shows that there is a "surface excess" and treatment of the data indicates that this "embryo film" is about 27Å thick. Its growth up to the point where a visible film can be seen has been followed Brown, A.S et al. (1998). The data contain a strongly falling ( $Q_z$ <sup>-4</sup>) Fresnel component which may be removed by multiplying by Q<sup>4</sup> to show the Kiessig fringe - the "form factor" - of this surface layer. At longer times of reaction (b,c) a clear Bragg peak emerges form the highly film structure at the interface.

Using neutron reflectivity and choosing deuterated and non deuterated surfactant against heavy water and "air contrast matched water" the contrast between the surfactant part and the silicate part of the film layer structure can be systematically varied (Brown *et al.*1998). In combination with the X-ray reflectivity results a complete picture of the development in one dimension, emerges. Figure 4 shows the Fresnel corrected reflectivity functions for several contrasts (left hand side) and the appropriately transformed real space densities at and below the growing surface at various times.



Figure 4. X-ray reflectivity from the air - water interface during the induction period showing the development of the reflectivity profile over time into diffraction peaks. a) 530 minutes, b) 608 minutes, and c) 687 minutes. The quality of the fits is shown.

#### Computing

With the availability of relatively fast mini computers, neutron scattering and data collection at the Institute Laue-Langevin (ILL), Grenoble in the 1970s was transformed. Real experiments began to be done rather than data measurements which were subsequently analysed days or weeks later. Two central computers were replaced by an individual computer at each instrument – the same thing happened at the Brookhaven high flux reactor and is now standard practice everywhere.

Subsequent improvements in computing speed and other computer performance, (graphics, peripherals etc), have clearly transformed neutron scattering experiments and have matched the growth in data production and data gathering power of the new generation of instruments. For example, at the ISIS spallation neutron source in the United Kingdom maps of the scattering function S(Q,w) are quickly available from the large detector arrays and acts at constant Q can be readily made. All of this permits the experimenter to enter the loop and do an experiment much as in conventional laboratory manner. Serendipity thus enters the process.

Attention is now focusing, and will focus more in the next few years, on how to best process and present the large data sets which can be taken in a matter of minutes or hours at modern sources. The computing power now available at the best laboratories, now permits data evaluation, on line, through the presence of models and fast simulation routines at the instrument. This can be taken much further and the data gathering power increased by progressive improvements in the modelling of instrumental effects such as multiple scattering and simulation the structure and dynamics of the system being studied.



Figure 5. Fresnel corrected reflectivity profiles for both X-rays (black), deuterated surfactant on air contrast matched water (orange), and protonated surfactant on  $D_2O$  (blue) with the corresponding real space profiles for a growing silicate film at the air - water interface.

An example of what might be expected is the simulation and constrained refinement of the Xray and neutron reflectivity profiles described above. As shown in the silicate film example it will be necessary not only to deal with the large data sets in real time, but also to use the constraints available form simultaneous refinement of data taken with different contrasts.

At the moment this is tedious. The person doing the refinement (although equipped with fast algorithms for finding the global minimum for a given set of parameters, is faced after each alteration with a list of parameters which must be manually changed. It is possible to imagine an essentially analogue version of this process given a fast enough computing capacity at the data reduction point. Parameter variation within certain limits could be done with "slide bars" for each parameter and the effect of the changes – via the fitting algorithms – observed

directly on the screen as a quality of fit to the reciprocal space data and an accompanying real space entity profile.

We thus foresee not only a great improvement in the data collection method but also a much greater user friendliness in the experimenter's interaction with the results.

#### **Conclusion**

As I write this lecture to celebrate the 40<sup>th</sup> Anniversary of a great Australian Institute I hear with great regret that the continuation application, through AINSE, to the Australian Research Council for funding of access to the advanced neutron facilities at the ISIS spallation neutron source in Britain has been denied. This should go on the record as our access to this Facility has been the result of a long and deliberate process to start cutting edge science using cold neutrons for the Australian chemistry and physics communities.

How grave is this problem?- AINSE has argued for cold neutron source in HIFAR since 1981: Australian structural work in the above fields has lagged Europe and USA in polymer, materials and surface science ; none of what I will describe in this lecture could have been done at HIFAR.

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#### **Australian Science - Past, Present and Future**

Max Brennan AO

I have been asked to talk to you about "Australian Science - Past, Present and Future" focusing on the period commencing in 1958, the year AINSE was established. Coincidentally, I completed my PhD degree and commenced my professional career in the same year, so I have a rather personal interest in the same period (but with a more limited future time span!).

#### *1958*

By 1958, CSIRO was well established and dominated the science research scene. It's research ranged across all types - from pure basic, through strategic, to applied and some experimental development. It was organised into divisions that were driven very much from the bottom up by the researchers, whose promotion was largely determined by their publications record. The 1964-65 Annual Report (one that I could readily lay my hands on) reported expenditure on research across nine areas. Agricultural research dominated with 39.0%, to which one can add another 15.9% for fisheries, processing of agricultural products and wool textile research. Industrial research and research "of industrial interest" amounted to 21.1%. Radiophysics, meteorological and upper atmosphere research (lumped together under the heading "general physical research") amounted to 6.2%. The total expenditure was 15.4 million pounds.

The AAEC Research Establishment was officially opened in 1958, with a research program strongly focused on the nuclear fuel cycle - driven by the assumption (as it has turned out, resoundingly incorrect) that Australia's coal resources were uncomfortably small.

University research was dominated by the ANU (to become the Institute of Advanced Studies after the amalgamation with the local CAE). Very little research was undertaken in the State universities although, by that time, the impact of Harry Messel's arrival in Sydney had produced Australia's first multi-professorial physics department with a strong research program. One measure of Harry's impact on Australian science is the step function jump in total funding for university research, clearly discernible in a plot of funding over time; and there are several other signs of his impact. Alongside the low level of university research activity at that time was the small number of PhD students. As an example, I was among the first few PhD graduates from the School of Physics at Sydney.

#### 1958 - 1988

The thirty years to 1988 saw several major changes in the structure and nature of Australian scientific research. The most striking change was the huge growth in research in the State universities and a steady growth in PhD enrolments. By the end of this period, the IAS no longer dominated the scene. This was particularly the case for postgraduate training, with the "sandstone" universities in the State capital cities having substantial postgraduate programs; also, by the late eighties there were pockets of strength in research and postgraduate training in the younger universities that were established in the sixties. The research was driven, like the earlier growth in CSIRO, by the researchers themselves; strong international links developed; interaction with industry was still relatively weak. The growth was fuelled by government funding - through the universities' operating grants and, increasingly, through the Australian Research Grants Committee, the main forerunner organisation of the Australian Research Council. Despite the increase in funding, and the high quality of much of the research, there were relatively few large groups that could claim a high level of international visibility.

The second major change during this period was the move by CSIRO, driven by successive governments, to move away from basic research towards research more explicitly and closely linked to the needs of industry. If I may jump ahead a little to illustrate this point: CSIRO's 1996-97 Annual Report is a vastly different document from the 1964-65 Annual Report. "CSIRO's research is (now) planned and resourced on a Sectoral basis". The names tell the story - agribusiness; environment and natural resources; manufacturing; information technology, infrastructure and services; minerals and energy. Within these five broad headings there are a total of 22 sectors. In addition, in a matrix approach to managing the organisation, there are four divisions - the "business units" of the organisation, that are "largely organised by scientific discipline". Research expenditure for the divisions is tucked away in an appendix. The text has a small table showing expenditure for "selected SEO subdivisions" (mineral resources, information & communications, manufacturing and the like). Total expenditure is now \$695 million (compared to 15.4 million pounds in 1964-65).

These two changes resulted in the university sector becoming almost the sole performer of pure basic research. This change, coupled with the growth in postgraduate training (so that only a small minority of students now go overseas for their training), has strengthened the university sector's role as the foundation for Australia's R&D system. This vital foundation role has many aspects. Three are particularly significant. First, pure basic research - the advancement of knowledge for its own sake; the advancement of our understanding of ourselves and the universe in which we live - contributes to the quality of our culture, connects us to the rest of the world (where 98% of the research is done), and (occasionally) leads directly to useful applications. Secondly, universities are ideally suited to undertake applied research aimed at the solution of problems in public interest areas such as health and the environment, and to the development of new and improved products and processes in industry. The reason why universities are so suited to this bridging role of applied research bridging between pure basic research and experimental development - is that it is most effective when coupled directly with a strong basic research and research training activity and, of course, with industry or other users of research. Thirdly, there is the universities' unique research training role.

Towards the end of this period, in 1987, the Australian Atomic Energy Commission (AAEC) was reviewed and, as one of the outcomes from the review, became the Australian Nuclear Science and Technology Organisation (ANSTO). This was more than a simple rebadging: the focus shifted away from nuclear energy (although, notably, work on SYNROC gas continued) to the use of nuclear techniques in a wide variety of applications. The environment became a particularly strong focus of attention; an all green cover for an early annual report provided graphic evidence of this change of emphasis. The change to a more "friendly" face for the organisation was also reflected in the rounded styling of the logo "a" (the design of which was commissioned and approved by the outgoing Commission).

#### 1988 - 1998

The pace of change has accelerated in the last ten years. The changes that have occurred include:

• the creation of John Dawkins' Unified National System of higher education; (It's worth pausing for a moment to make a few comments on this revolution in higher education. There were two agendas that drove the change. First, the Federal Government of the day wished to increase access to higher education without significant cost to the taxpayer. Secondly, the Colleges of Advanced Education and the Institutes of Technology were pushing for "a level playing field" - specifically, to get access to research funding through

their operating grants and to be allowed to offer higher degrees by research. Some of the major outcomes of this revolution are:

- ♦ the desired "massification" of higher education;
- ◊ the abandonment of accreditation procedures without the introduction of adequate quality assurance measures;
- ◊ a decrease in the number of institutions from about 70 to 36 (much easier for the bureaucrats to handle);
- the reclassification of institutions, so that they are now all called "university" (there are positive aspects to this); and
- $\diamond$  an enormous increase in the demand for research funds.)

To return to the list of changes that have occurred in the last ten years:

- greatly increased links between the research performers universities and the government research organisations and industry;
- a major, new government program the Cooperative Research Centres which has transformed the scope and nature of the links between all sectors, and which has provided a new environment for postgraduate training;
- enormous budget pressures on universities with a resulting fundamental change in the nature and management of the university, and some loss in staff morale;
- a static funding situation for basic research Australia is being priced out of the market in several research fields;
- a drop in the international "visibility" of Australian science (which, if it continues, will weaken our ability to keep in touch with the other 98%); and
- even less understanding than before, in government and the bureaucracy, of the vital role that university research plays in the national R&D system (witness the extraordinary and uninformed proposal by DETYA to demolish the ARC's programs).

It's very important to note that, while all of this structural change has been going on, the researchers have got on with the job as best they could. There are still many quite outstanding researchers who are leaders or among the best in their fields. For the universities, that can been seen in the outcomes of the peer review process for ARC supported research in the State universities and from the 1995 review of the IAS. I am sure that the internal review processes for the government research organisations reveal similar high levels of achievement.

#### The future

The plasma physicists in the audience will know, or know of, Boris Kadomtsev. I recall a remark he made at a meeting. "Making predictions is difficult, particularly if you are trying to predict the future." That's certainly true for Australian science!

As for changes in the fields of research, that's very much in the hands (indeed, the minds) of our young researchers: they will create their own future. Perhaps my own field, physics, will move from being a stretcher case to the morgue; but I doubt that. Surely, there will be an industry led recovery for physics some time in the next century when Australian industry catches up with the rest of the industrial world. High performance computing will undoubtedly open up new sub-fields of research, probing complex structures and systems that are currently beyond our reach. Multidisciplinary research will become the norm rather than the exception (perhaps physics *per se will* disappear).

As for structures and funding, I'm the perennial optimist. I don't believe the forces of darkness will destroy the ARC (particularly, if you all go home and write a letter to the Minister, Dr Kemp); and we will be forced to follow the lead of other OECD countries such as the US, UK, Japan and South Korea, which have substantially increased their funding of basic research.

And what of the government research organisations? I predict something of a swing back to basic research - not a big change, but significant. Why? Because they will find that to focus almost entirely on the immediate needs of industry and public interest issues threatens the long term viability of the organisation.

### AINSE – The First Thirty Years 1958-1988

Mr E A (Bill) Palmer

Former Executive Officer AINSE 1959-1988

The original constitution adopted on 4 December 1958 to establish AINSE, and thereafter known as "The Rules", remained unchanged for the first thirty years of the Institute's existence. In this period the number of member universities increased from nine to nineteen, the Australian Atomic Energy Commission (AAEC) became the Australian Nuclear Science and Technology Organisation (ANSTO), and the wording of the relevant Commonwealth legislation changed from "uranium and atomic energy" to "nuclear science and technology".

Foundation Members of AINSE	Standing representative originally appointed to AINSE Council by the ten foundation members	
ADE	Prof D O Jordan – Chemistry	
ANU	Prof E W Titterton – Nuclear Physics	
MEL	Prof H W Worner – Metallurgy	
NSW	Prof R H Myers – Metallurgy	
QLD	Prof H C Webster – Physics	
SYD	Prof H Messel – Physics	
TAS	Dr G H Cheesman – Chemistry	
UNE	Prof J M Sommerville – Physics	
UWA	Prof D J Allen-Williams – Mechanical Engineering	
AAEC	Prof Sir Leslie Martin Deputy Chairman AAEC	
AAEC	Mr A D McKnight Executive Commissioner AAEC	
AAEC	Dr C N Watson-Munro Chief Scientist AAEC	
AAEC	Prof J P Baxter Chairman AAEC	

This agreement on 4/12/58 by representatives of each of the nine universities then existing in Australia and four representatives of the AAEC, to establish the Institute, followed discussions initiated in 1957 by Professor J P Baxter then Chairman of the AAEC and Vice Chancellor of the University of New South Wales. The Atomic Energy Act (1953) authorised the AAEC to co-operate with Australian Universities to assist research and training in fields related to uranium and atomic energy, reflecting the hopes and expectations then widely held concerning the production of cheap and abundant energy for civil purposes from nuclear sources. Two significant events of this time were the building of HIFAR at Lucas Heights (criticality initially achieved on 26 January 1958) and the 1958 Geneva Conference on the Peaceful Uses of Atomic Energy. There was also much interest in radiation effects, radioisotopes and controlled thermonuclear fusion.

Adoption of the "Rules" created AINSE as an independent body governed by the AINSE Council, with funds provided by annual subscriptions from the member organisations and an annual "Contribution for Research and Training" from the Commonwealth Government. The income for 1960, effectively the first full year of normal operation, was about \$140,000.

Established rules and funding enabled AINSE to operate effectively as an unincorporated association for thirty years before changes in states laws permitted adoption of a corporate form.

The universities accepted a responsibility to assist in preparing Australia to benefit from the peaceful applications of nuclear science and engineering. This was shown in the Council's early decisions to provide AINSE assistance over a wide range of disciplines either relevant to the nuclear field or requiring the use of research facilities at Lucas Heights. As a result of having access, through AINSE, to the HIFAR and MOATA research reactors, no Australian academic institution found it necessary to incur the high costs and other problems involved in operating a nuclear reactor on its own campus. The wisdom and foresight of the persons responsible for this situation in Australia are manifest when the experience of several other countries is examined.

AINSE PRESIDENTS 1958 – 1988					
1958 & 1959	Prof D O Jordan	ADE – P & I Chemistry			
1960	Prof J M Sommerville	UNE – Physics			
1961 & 1962	Prof D O Jordan	ADE – P & I Chemistry			
1963 & 1964	Prof H W Worner	MEL – Metallurgy			
1965 & 1966	Prof R Street	MON – Physics			
1967 & 1968	Prof C N Watson-Munro	SYD – Physics			
1969 & 1970	Prof D E Caro	MEL – Physics			
1971 & 1972	Prof M H Brennan	FLI – Physics			
1973 & 1974	Sir E W Titterton	ANU – Nuclear Physics			
1975 & 1976	Prof P A Parsons	LAT – Genetics			
1977 & 1978	Prof E O Hall	NCT – Metallurgy			
1979 & 1980	Prof R W Parsons	QLD – Physics			
1981 & 1982	Prof S C Haydon	UNE – Physics			
1983 & 1984	Prof I J Polmear	MON – Materials Engineering			
1985 & 1986	Prof K N R Taylor	NSW – Physics			
1987 & 1988	Prof B M Spicer	MEL – Physics			

Critical actions of the early years include Council decisions to set up specialist committees in fields such as nuclear physics, plasma physics, radiation chemistry, radiation biology, neutron diffraction, aspects of engineering relevant to nuclear technology and nuclear techniques of analysis. Fortunately the necessary expertise was available from within the universities and the AAEC, and the Council received from these committees advice on the development of the Institutes' operations. Critical roles were also played by the Executive Committee, and each of the fifteen distinguished university representatives who held office as AINSE President in the first thirty years.

Organisations which became AINSE members 1958 - 1989		Standing representative originally appointed to AINSE Council	
1961	Monash University	Prof R Street – Physics	
1965	University of Newcastle	Prof E O Hall – Metallurgy	
1966	Flinders University	Prof M H Brennan - Physics	
1966	La Trobe University	Prof P A Parsons - Genetics	
1966	Macquarie University	Prof P Mason – Physics	
1970	James Cook University	Prof J F Ward – Physics	
1975	University of Wollongong	Prof P Fisher – Physics	
1975	Griffith University	Prof R L Segall – Physical Sciences	
1982	CSIRO	Dr P G Alfredson – Energy Technology	
1985	Murdoch University	Prof B Mainsbridge - Physics	
1988	University of Technology Sydney	Prof R J Breakspere - Chemistry	

Most universities appointed a representative on the Council for an extended period, providing opportunity for a Councillor to hold elective office and serve on the Executive Committee for up to eight years. As the four Councillors appointed by the AAEC usually served for several years also, this continuity was important in maintaining the understanding and the good relations which were critical factors in AINSE operations for the first 30 years.

This was also assisted by the requirement in the "Rules" that the Council must meet at least four times each year. From about 1963 two Council meetings each year took place at Lucas Heights and two were held at universities, enabling representatives of both the AAEC and the universities to retain a familiarity with the relevant research activities and facilities throughout Australia. Through the decades AINSE benefited from the active co-operation of the AAEC Commissioners and members of the AAEC staff.

From the early 1960's the Council initiated several series of AINSE Conferences, one series in each of the relevant fields, usually bi-annual. These conferences gathered together most of the people in Australia active in each research area, and frequently involved distinguished scientists visiting Australia. Most conference series have continued to the present and have encouraged the reporting of work in progress, thus providing opportunities for many research students to present papers. Research results arising from projects receiving AINSE support were always made available without restriction and gave rise to publications by the investigators in a wide variety of journals.

AINSE grants were originally awarded in 1960 and have continued as the major means by which the Institute has assisted research undertaken by the member universities. These grants provided modest but useful funds for the procurement of research equipment and met the cost of travel and accommodation for visits and attachments of university staff and research students to Lucas Heights. The projects assisted by these grants (up to one hundred per year by the third decade) frequently involved active collaboration with scientists at Lucas Heights. AINSE also assisted by arranging university access to certain materials such as heavy water and to the specialised facilities of AAEC laboratories and workshops.

Another early action was the decision to award each year a small number of AINSE Studentships fully funded by AINSE. These post graduate awards attracted able PhD candidates whose projects required them to spend more than one quarter of their working time at Lucas Heights. For more than twenty five years up to 1988, two or three AINSE Studentships were awarded each year, many of the holders subsequently progressing to distinguished research careers. Average tenure was about three and half years, so there were often up to ten AINSE Students active at any one time.

Similarly, from the early 1960's at least two AINSE Research Fellowships were awarded each year in response to nominations submitted by the member organisations. These post doctoral awards were also fully funded by AINSE including costs of travel to and from Australia for successful overseas candidates. Competition was considerable and the awards provided valuable opportunities for persons at an early stage of an independent research career. AINSE Research Fellows worked in a wide variety of disciplines, usually involving co-operation between university based research groups and scientists at Lucas Heights. Duration of tenure would average about two and a half years resulting in about five Fellowships being held at any one time. Many ex-AINSE Fellows now occupy senior appointments in Australia and in other countries, the links thus established having a continuing value.

Also important were the Councils' decisions to provide AINSE scientific and technical staff at Lucas Heights to assist university visitors in using either AAEC or AINSE research facilities, to collaborate in the development of research equipment and undertake co-operative research projects. The AINSE Neutron Diffraction Group commenced operations in 1965 and eventually consisted of four staff members. AINSE funded the provision and operation of a number of neutron diffractometers on HIFAR, starting with a single crystal instrument in 1963 and concluding with the allocation of \$400,000 in 1988 towards the construction of AUSANS (the Australian Small Angle Neutron Spectrometer). There was a close association with members of the AAEC staff in neutron scattering activities and valuable support from the Commission in making available the neutron beams from HIFAR. Facilities evolved from single counter, manually operated instruments in the early 1960's to multi-counter arrays and computer controlled devices by the late 1980's. Helium cryostats were used from the early 1970's but efforts from 1974 towards persuading the AAEC to install a "cold-source" in HIFAR were not successful. The universities were major users of the neutron beam facilities at Lucas Heights (both AAEC and AINSE), with PhD students maintaining a continuous presence over the decades.

The AINSE Accelerator Group commenced operation in 1972 and was maintained at Lucas Heights to 1988 with two staff members. It's activities were in support of university groups using the 3MV proton accelerator (PIXE, PIGME, etc) and the 1.3MV electron accelerator (for radiation chemists, who also used AINSE's 600KeV Febetron). Demand for the use of accelerator based nuclear techniques of analysis grew strongly through the 1970's and 1980's with AINSE staff members taking an active role in research and assisting university groups from archaeology, anthropology and other disciplines. Such developments may be seen as reflecting the increasing interest in Australia in the "non-power" applications of nuclear science and technology, following the abandonment by the Australian Government in 1972 of plans for building a 500 MW(e) nuclear power reactor at Jervis Bay NSW.

All these AINSE activities involved visits and attachments of university staff and students to Lucas Heights, in considerable numbers, and sometimes for extended periods. Before 1965, visitors from outside the Sydney region were accommodated in hotels in the Sutherland Shire,

(one such hotel becoming known as the "AINSE Rex"), but thereafter most such visitors stayed at Stevens Hall built for the AAEC at Lucas Heights.

The AINSE building, outside the security fence of the AAEC Research establishment at Lucas Heights was completed in early 1960, about the same time as HIFAR was finally brought up to its full 10MW power level. The AINSE Building became the base for the AINSE office, and for AINSE meetings, visitors, lectures and conferences. Arrangements for all these and many other activities were made by the AINSE administrative staff, eventually three in number who assisted the Executive Officer. Some of the female members of the AINSE staff were with the Institute for many years, contributing greatly to its activities and AINSE's identity. Over the years the AINSE lunch room from about 1pm each day was a gathering place for university visitors from many disciplines, to interact with AINSE and AAEC staff members.

Maintaining an income each year adequate to finance the Institute's activities was a challenge from the commencement of activities in 1959, and particularly during the periods of high monetary inflation which occurred in each of the first three decades. Repeated representations to sources of funds, and an eventual agreement by the member organisations to certain forms of indexation enabled the level of operations to be sustained, and occasionally increased. Periods of high interest rates assisted in the building up of useful reserves, but throughout the first thirty years, total expenditure exceeded the total of funds provided by or through the member organisations.

Each of the first thirty years had its own highlights, some in the nature of objectives achieved, and some in the form of minor crises. These highlights included occasional visits by some AINSE staff members to overseas laboratories and international conferences, leading to valuable associations with institutions and individuals active in AINSE's fields of interest. AINSE was also able to assist in arrangements for visits within Australia by distinguished scientists from overseas, particularly those participating in AINSE conferences.

Forty years after the establishment of the Institute on 4 December 1958, many of the original concepts, and many of the ideas which evolved in the first thirty years still underlay AINSE's present activities. Through those three decades the input from each of the many hundreds of persons involved made life within the Institute a very interesting experience, and made it possible for AINSE to make a useful contribution to research and training in nuclear science and engineering and to the applications of nuclear technologies. New ideas have been introduced in the ten years since 1988, and AINSE will continue to build on its original foundations.

Some longer-serving members of AINSE council and/or committees 1958 - 88 (not including AINSE presidents or councillors initially appointed)		Some longer serving AINSE staff members for periods between 1959 and 1988	
Mr K F Alder	AAEC	Mr B Palmer	Executive Officer
Mr F L Bett	AAEC	Mrs J Kearns	Administration
Dr R J Bird	AAEC	Mrs B Smith	Administration
Prof H Bloom	TAS	Mr G Fisher	Administration
Prof S T Butler	AAEC	Mr D Wheeler	Neutron Diffraction Gp
A/Prof R Cooper	MEL	Mr P Lloyd	Neutron Diffraction
Dr C Dalton	AAEC	Dr F Moore	Neutron Diffraction
Mr D R Davey	AAEC	Mr R Ebdon	Neutron Diffraction
Dr M E Elcombe	AAEC	Mr C Bock	Neutron Diffraction
Mr G W K Ford	AAEC	Dr L Davis	Neutron Diffraction
Dr J N Gregory	AAEC	Mr S Kannard	Accelerator Group p
Prof S Hamberger	ANU	Mrs J Watson	Administration
Dr C J Hardy	AAEC	Dr D Cohen	Accelerator Group
Dr C J Hicks	MON	Mr A Katsaros	Accelerator Group
Dr C J Howard	AAEC	Mrs J Smith	Administration
Prof I Jones	FLI	Mrs J Brownjohn	Administration
A/Prof G Laurence	ADE	Mrs J Breakspere	Administration
Dr G L Miles	AAEC		
Dr J K Parry	AAEC		
Dr T M Sabine	AAEC		
Mr D F Sangster	AAEC & CSIRO		
Dr R Smith	AAEC		
Dr J L Symonds	AAEC		
Mr A D Thomas	AAEC		
Prof J J Thompson	NSW		
Mr M C Timbs	AAEC		
Dr D G Walker	AAEC		
Dr G M Watson	AAEC		
Dr A Wilson	AAEC		

#### AINSE - The Years Between 1988 and 1998

Roger B Gammon AINSE (1988 - 1998) Australian Institute of Nuclear Science & Engineering

#### Abstract

This paper lists some of main events in the history of AINSE between the years 1988 and 1998. It was a time of great change for AINSE. At the beginning of this period the Government implemented major structural reforms of the higher education system and new mechanisms for research infrastructure support followed a year later. This culminated in 1993 with a major restructuring of AINSE which had continued with little change since its inception in 1958. This was followed in 1994 with a comprehensive review and reorganisation of ANSTO, which also had a significant effect on AINSE. These events will be described and will include some of AINSE's considerable achievements and successes, which saw the organisation, in partnership with ANSTO, in a strong position at the end of this ten-year period.

#### Foreword

"There is no security on this earth: there is only opportunity" Douglas MacArthur, His rendezvous with history 1955

The above quote was recently reproduced in the *November 1998 issue of Australian Institute* of Company Directors - Forging the Future, Director Perspectives for the 21st Century. It seemed particularly appropriate as I looked back over the 1988 - 1998 period. After 30 years of comparative stability, change was inevitable. At first, I had every intention to keep things much as they were after such an illustrious record of success and achievement, but the forces for change were already waiting in the wings. One cannot ignore the threats to AINSE's security that arose but the positive side was that the opportunities presented by change were also grasped. AINSE today is an organisation in tune with the present social, economic and political environment. Threats to its security will no doubt continue to arise but so will opportunities.

#### 1988.

The President was Prof Brian Spicer (University of Melbourne). I joined as Scientific Secretary on 28 June and took over from Bill Palmer on 1st August. At that time there were 18 member universities in partnership with ANSTO and CSIRO. There were 5 specialist committees: Nuclear Physics, Radiation, Neutron Scattering, Plasma Physics and Engineering. There were 10 full-time AINSE staff members - a CEO, 3 scientists, 3 technicians and 3 administrative. AINSE was funded by subscriptions from its member universities matched by an equal contribution from ANSTO. The CSIRO also made a small contribution and there was a small amount earned from interest on AINSE's financial reserves. The largest component of income was in the form of a "Research and Training" grant provided by ANSTO. AINSE owned and maintained research equipment worth over \$2 million. Plans for the construction of a small angle neutron diffractometer on HIFAR were being advanced with the assistance of Dr Lindsay Davies and his team. My first pleasant task was to offer ANSTO funds of \$400,000 over a period of 4 years to purchase equity in this facility. It was expected the AUSANS instrument would be completed by the year 1992. It seemed a good start.

**1989.** "The period ... has been a most challenging time for AINSE as major changes in Commonwealth Government policy on funding of research, in particular, have had their impact on AINSE's member organisations. AINSE has responded to this challenge by taking positive steps to increase the efficiency and effectiveness of its operations." Scientific Secretary's Report to Council for the period 1st July 1988 to 31st December 1989.

The President was Prof Ross Miller, University of Adelaide. The composition of the Executive Committee was changed. Major activities included a review of benefits received by member universities over the preceding 10 years and a re-adjustment of membership subscriptions. This was largely in response to major changes in Commonwealth Government policy on university funding (the ARC "claw-back") and the application of the "user pays" principle to the payment of AINSE membership subscriptions. An investigation of incorporation issues was commenced. A new Council Room was constructed in the south wing of the AINSE Building. A Special Colloquium on "Cold Fusion - Fact or Fiction" was held and AINSE offered \$90,000 in support of the team working on cold fusion at ANSTO. We produced a promotional video and the first issue of a newsletter. In response to costcutting measures in ANSTO, services formerly provided by ANSTO, such as printing and graphic design, were contracted out. We reviewed AINSE grant procedures and the AINSE staff superannuation scheme. Many administrative procedures were computerised. AINSE was accepted as the representative organisation of radiation researchers in Australia and admitted as a member organisation of the International Association for Radiation Research. The Executive Director of ANSTO's appointment to the International Science and Technology Advisory Committee was supported with a view to representing AINSE's interests in access to major overseas science facilities. It was decided to gradually phase out AINSE studentships and use the funds for "supplements" to holders of Australian Postgraduate Research Awards to allow more students to participate. In this period there were 12 PhD students from 10 universities; and 6 Research Fellowships held with 5 universities. Three new universities were admitted to membership of AINSE - UTS, Curtin and RMIT.

# **1990.** "... AINSE has entered into perhaps its most critical period since its inception 32 years ago. ... AINSE faces the stimulating prospects of a new and expanded future provided it can successfully find its place in the changed economic and social environment in which it now finds itself." Scientific Secretary's Report to Council for 1990.

Prof Ross Miller was President for a second year. Government policy was concerned with the necessity to redress a serious economic situation facing Australia. Amongst remedial measures were major changes in Government funding mechanisms for research. Changes were aimed at encouraging industry to undertake more indigenous research and development, government research organisations to take on a commercial role, and universities to focus research efforts onto areas of short to medium term importance to the achievement of national goals. A new Fellowship scheme was set up (for implementation in 1991) under which high priority areas of research were identified and the Fellows were appointed to AINSE. AINSE was incorporated under the NSW Associations Incorporation Act 1984. There was also pressure exerted on AINSE to restructure. In July at the University of Newcastle, the Executive Director of Ansto, Dr David Cook, presented a number of options to the AINSE Council. Amongst them it was proposed that "AINSE could be absorbed within ANSTO at no cost". This paper had major implications for AINSE in so far as ANSTO proposed to play a much more pro-active role in the affairs of AINSE than had ever been the case in the past. The universities responded by agreeing, amongst other things, that AINSE's research activities should be expanded and that there should be less support given for direct grants to university-based plasma physics research. A strategic planning process was implemented. A new staff superannuation scheme was introduced. The AINSE Febetron was relocated at the

University of Melbourne. Submissions were prepared to fund university membership subscriptions from ARC Infrastructure Mechanism "A" and "C" Funds. A joint application with ANSTO was prepared to seek funding for the Australian Small-Angle Neutron Scattering Instrument under the ARC Major Equipment (Mechanism "B") program. The Government established a number of Co-operative Research Centres and many of AINSE's principles were incorporated into the new program.

**1991.** "Despite ... considerable achievements, the future of AINSE is uncertain. This is primarily because its principle ''shareholder'', ANSTO, wants to change the existing organisational structure, and universities are under increasing pressure from competitive demands for research funds". Scientific Secretary/Executive Officer's Report to Council for 1991.

The President was Prof Peter Fisher, University of Wollongong. To offset the "claw-back" and reductions in real terms to the "Research and Training" grant, AINSE sought external funding from the ARC for equipment for use by a number of universities. AINSE was successful in attracting approximately \$0.75 million in external grants and for the first time exceeded the funding provided by ANSTO. Every member university was visited to rally support for the payment of subscriptions from centralised funds. Four more universities applied for membership. AINSE's Mission, Objectives and Strategies were approved and published as a pamphlet. "The Vital Link" brochure was published. A potential set of performance indicators for AINSE was developed with assistance of the Centre for Technology and Social Change. In February, Council approved the title of AINSE Honorary Fellow for distinguished and dedicated service to AINSE. Further to ANSTO's restructuring proposals, Council agreed that AINSE should play a role in promoting ANSTO/University collaboration and set up new Specialist Committees to support ANSTO's widening research interests. The Cyclotron building at the Royal Prince Alfred Hospital in Sydney was completed and member universities showed great interest in getting access to the medical diagnostic PET facility. AINSE provided funding for the development of a small animal imaging system. A Fujitsu VP2200 supercomputer at Lucas Heights presented new opportunities for AINSE researchers. ANSTO was appointed the managing agency for Australian collaboration with overseas "big science" facilities. The erection of a security fence around HIFAR was commenced. Various organisational and structural changes were proposed during the year including one from ANSTO that Council members must be at the level of Pro-Vice Chancellor or equivalent. A motion was passed at the end of the year with the support of the university members that Council would not proceed further at that time with formal restructuring of the AINSE Council.

## **1992.** ".... External pressures ... threaten the viability, and even the existence of AINSE, but there may also be significant opportunities for AINSE." Foreword from the President, AINSE Annual Report for 1992.

The President was Prof Jim O'Donnell, University of Queensland. The year was dominated by negotiations between ANSTO and the university members of AINSE on the major restructuring of AINSE. The objective was to achieve more effective interaction with ANSTO by an integrated scientific and administrative operation with closer links between university and ANSTO researchers. To achieve this, the universities agreed that all AINSE scientific, technical and clerical staff, with the exception of the Executive Officer and a secretary, should either transfer to ANSTO or accept voluntary redundancy. AINSE's scientific equipment assets (with an historical cost value of \$2,454,000) would be transferred to ANSTO whilst financial reserves of \$1,195,000 would be held in trust for the university members of AINSE. ANSTO agreed to match future university subscriptions on a 2:1 basis and absorb much of AINSE's administrative costs. The Government decided to defer the decision to replace HIFAR. At the end of 1992 AINSE staff consisted of: an EO, 3 scientists, 3 technicians, 3 administrative plus 2 professional officers (funded by ARC) and 4 Post Doc Fellows - making a total of 16. There were 25 member universities, the CSIRO (which withdrew at the end of the year) and ANSTO.

# **1993.** "The present restructuring represents the greatest change that has occurred in the thirty five year history of AINSE. .... I remain very optimistic about the future of AINSE and feel confident that the partnership with ANSTO will continue in a spirit of productive cooperation." President's Foreword, AINSE Annual Report for 1993.

The President was Prof Len Lindoy, James Cook University. It was mainly a year of implementation of the MoU. Council met only twice (once to just approve the amended Rules). Throughout the year, mechanisms and procedures were developed which aimed to transfer much of the administration and management of AINSE's operations to AINSE whilst retaining overall control with AINSE through its EO. There were concerns that, although advances in technology facilitated better communication between universities, the AINSE networking of scientists and engineers would be lost. It was a difficult year for ANSTO with the Research Reactor Review finding against the near-term replacement of HIFAR; with the proposal to merge ANSTO with CSIRO causing uncertainty; and with the appointment of a new Board of Directors in December. In previous years the bulk of AINSE's funds were spent on developing instruments and paying salaries of scientists and technicians. Specialist Committees allocated time on instruments at no additional cost. Under the new arrangements, AINSE's funds were used to cover charges for use of the facilities. This provided an incentive to ANSTO staff to collaborate and provide support. A new and important facility for AINSE users, the Tandem AMS, received funding from the ARC to support their use of it in collaboration with ANSTO. The inaugural AINSE Gold Medal was awarded for excellence in research supported by AINSE during the preceding 5 years.

## **1994.** ".... looking forward to a nice smooth run after the many significant changes that had been made in the previous few years. .... I suppose I should have expected that change would be the order of the day." President's Foreword for AINSE Annual Report 1994.

The President was Prof Brian Stone, University of Western Australia. Dr Cook resigned and in view of the changes taking place within ANSTO, there was concern about obtaining guaranteed access to all the facilities currently used by university researchers. The new Chair of ANSTO, Ralph Ward-Ambler, attended Council and outlined the outcomes of the ANSTO Mission Review. The major objective would be first and foremost to support the Government's nuclear policies, but ANSTO would continue to maintain a high quality nuclear science base and enable academic institutions and other science organisations to perform research by providing access to unique facilities and expertise. Councillors were assured that AINSE was an important and welcome partner with ANSTO. Activities were to be focussed around a small number of well-defined key research areas. Some facilities would shut down. AINSE established a Task Force to consider the Review recommendations. On the initial closure or transfer from ANSTO list was the MEVVA ion beam implanter, 1.3 MeV electron beam accelerator, GATRI and Gamma Pond, MOATA, Neutron Activation Analysis, plasma implantation, high energy physics, the Biomedicine and Health Program, and Australian Radioisotopes. It was established that AINSE would have guaranteed access to 50% of the neutron beam time on HIFAR. All remaining facilities would be made available in the national interest - subject to ANSTO's commercial requirements being met. The first international institution, the University of Auckland, joined AINSE but CSIRO withdrew under the new arrangements with ANSTO.

**1995.** ".... my first year as President .... has been one in which AINSE has progressed markedly. ... the Executive Director of ANSTO and myself committed ourselves to develop AINSE into an organisation of significance by strengthening the partnership between the universities and ANSTO." President's Foreword, AINSE Annual Report for 1995.

The President was Prof Bob Breakspere, Central Queensland University, and the Executive Director of ANSTO was Prof Helen Garnett. A major issue was the decision of the AV-CC to exclude AINSE Grants from being counted in the Competitive Research Grants Index - which essentially determined the amount of government research infrastructure funds to each university. There were mutually acceptable outcomes to facilities affected by ANSTO Mission Review. Membership was increased to 31 universities. The National Plasma Fusion Research Facility received \$8.7 million under the Major National Research Facilities Program. AINSE was to play an important role in facilitating the use of this facility by researchers from member universities.

# **1996.** "... my second year as President ... has been one of both expansion and consolidation for AINSE. ... substantial progress has again been made ... to develop AINSE into an organisation of international significance." President's Foreword, Annual Report for 1996.

The President for a second year was Prof Bob Breakspere. There was a further increase of membership to 34 universities. Strategic planning for the new AINSE was considerably progressed and Council unanimously approved AINSE's objectives on the 6th December. An AINSE Web page was set up and \$500,000 was put aside in a Long Term Projects Reserve. "AINSE now truly represented researchers and institutions that are concerned with access, replacement and development of major nuclear science and associated facilities within Australasia. This was just part of AINSE's contribution to Australia's nuclear science expertise for the 21st century."

# **1997.** "AINSE continues as a successful organisation, exploiting the privileges provided by ANSTO for the benefit of university researchers. ANSTO derives no less benefit from the involvement on site of the large pool of researchers and graduate students from around Australia, along with gaining the ability to demonstrate efficient and widely based utilisation of central resources." President's Foreword, AINSE Annual Report for 1997.

The President was Prof Trevor Ophel, Australian National University. This year marked the end of the first five years Memorandum of Understanding between AINSE and ANSTO. Overall, it was felt that the new arrangements operated well. A new MoU of similar duration was signed. Council adopted performance indicators for AINSE's objectives. In May, ANSTO proposed that AINSE should take over responsibility for facilitating access to the Advanced Neutron and Muon Beam facilities (ISIS) in the UK. The 1.3 MV electron accelerator was decommissioned and re-established at the Australian Radiation Laboratories in Melbourne. A major refurbishment of the AINSE Theatre and headquarters building, including an expansion of the Council Room, was completed. The first "AINSE Winter School at ANSTO" was held where top undergraduate students from all member universities (each funded by an AINSE scholarship) spent time at ANSTO working on major facilities with international experts in their field to encourage students to go on to postgraduate studies.

### **1998.** *"AINSE (has) emerged ... as a stronger organisation with a clearer view of its future and a more secure relationship with ANSTO." Executive Officer's Report to Council, 1997.*

I will conclude on the subject of opportunities. These sometimes take a few years to materialise. On the 31st March 1998 I retired from AINSE but not before the first 2-D small angle scattering pattern from AUSANS was obtained! We were highly successful in attracting

research infrastructure funds for the shared use of all member organisations. Since 1992, external grants totalling \$3,954,000 had been won. AINSE was also part of the consortium that was awarded \$8.7 million in 1996 under the Major National Research Facilities program for the National Plasma Fusion Facility. A RIEF Grant of \$300,000 has been awarded for access to ISIS and opens up opportunities for future AINSE use of major overseas facilities. Since 1988 the number of projects supported by AINSE Grants and Postgraduate Research Awards has increased from 101 to 234 and now include a much wider range of disciplines. AINSE conferences are now held regularly in association with other major national and international conferences, and specialised workshops and seminars are held on emerging techniques at ANSTO. The number of member universities has risen from 17 to a peak of 36 in 1997. The combined representations of all these scientists have supported ANSTO in the successful quest for a replacement reactor for HIFAR. This is surely another big opportunity for AINSE.
# Nuclear Science and a Better Environment – an Oxymoron ?

Gerald Laurence The University of Adelaide and Aduchem Pty Ltd

No – or at least nuclear science and a better understanding of the environment is not an oxymoron. It is only through that understanding that a better environment is possible. AINSE has provided the means for many research projects to use nuclear techniques in studying the environment but nuclear science is a means not an end in environmental studies.

### The Past

The original technique driven studies derived from nuclear and physical sciences (1930 - 1960) had given way to issues studies by the 1980s and new techniques are now rapidly exploited for issues research.

The use of Pb-210 in soil studies in the late 1930s depended on the new Geiger-Muller counters. Tracer studies expanded with the provision of reactor radioisotopes in the 1950s and improved dating and detection techniques (C-14, AMS) extended nuclear techniques to erosion, palaeoclimate, and quaternary studies in general by the 1990s.

Australia was initially slow to use these techniques for environmental studies. An AAEC sponsored Symposium in 1958 contained no papers dealing with environmental science, and the list of broadly defined nuclear science research in Australia indicates that no biological, geographical, environmental, or quaternary research in Australia was using these methods.

In 1960 an AINSE Conference was organised to discuss radiocarbon dating and AINSE appointed a post-doctoral fellow in 1961 to provide a link between the universities and AAEC in the development of C-14 dating in Australia. Dating remains a major area of AINSE activity and AINSE and ANSTO support for special dating techniques makes a major contribution to Quaternary and environmental research.

AINSE supported projects naturally reflect current scientific issues and the availability of techniques at ANSTO. These trends and the importance of AINSE to Australian environmental science in particular areas are shown by the environmental projects supported over the years. My list is representative, not exhaustive, and reflects an increasing interest in dating in environmental studies.

### Contributions To A Better Environment "By-Product" Contributions

Unintended nuclear contributions to environmental studies include the "bomb pulse" injection of anthropogenic C-14, H-3 and Cs-137 into the atmosphere in the early sixties because of bomb testing; the injection of Kr-85 from fuel reprocessing plants, and the research into the effects of uranium mining operations. A number of long term environmental studies are associated with uranium mining. One effect of the Roxby Downs operation has been the establishment by WMC of the environmental monitoring system required by the state government. This has provided a large amount of information on the eco-system of an area which had previously been visited by biologists very infrequently – the WMC environmental operation as similar to establishing a permanent Antarctic base.

#### **EXAMPLES OF AINSE PROJECTS OVER 40 YEARS**

1961 Fruit fly control by radiation sterilisation
1969 Biomass productivity of phytoplankton in Tasmanian lakes using O-18
1976 Erosion in the Condamine basin using Pb-210 & Cs-137 dating
1977 Isotopic ratios in groundwater
1981 Cross-placental transfer of uranium in marsupials
1983 Mobility of thorium in mill tailings
1985 Role of iron oxides in heavy metal retention in soils
1985 Biomineralisation studies
1986 Heavy metal accumulation in mangroves
1992 Be-10 as a sedimentation rate indicator
1993 Study of groundwater recharge
1993 Sources of particulates in urban haze in Sydney basin
1994 Coastal climate change 20000 BP to present

### Main Applications

The contributions of nuclear science to environmental studies fall in three primary areas: tracer studies, elemental analysis, and temporal information (mainly dating).

The use of stable and radionuclides as tracers for studies of dynamic process in the environment – coastal sediment movement, water transport (both surface and sub-surface), soil-water exchange, atmospheric and oceanic circulation.

The determination of a large number of elements simultaneously, automatically and without chemical separations at low detection limits is frequently demanded in large-scale environmental research projects and nuclear techniques of analysis such as NAA, PIXIE and PIGME are often the method of choice in areas such as air-borne pollution and biomineralisation.

Comparison of analytical techniques in particulate air pollution					
Technique	Multi-element	Sensitivity	Accuracy	Sample simplicity	
ICPES	+	+	+	+/-	
AAS	-	+/-	+	+/-	
XRF	+	-	+	+	
PIXIE	+	+/-	+/-	+	
INAA	+	+	+	+	

Temporal information may be provided by radionuclide decay or by the slow establishment of exchange equilibrium such as that between atmospheric carbon-14 and the hydro- and biospheres. The range from the Rn-222 half-life of 8.2 days to the U-238 half-life of 4.5 x  $10^9$  years covers applications as diverse as air-flow measurements in caves to geology. AMS determination of nuclides at high sensitivities with low detection limits is commonly (but not exclusively) used for dating purposes. About 70% of the AMS measurements made on ANTARES cover climate, geomorphology, paleoenvironment and hydrology.

# Lithosphere

Nuclear science contributes to studies of sedimentation and erosion, mining impacts and mine remediation, fertiliser and pesticide application, utilisation, and control.

The importance of dating in studies of deposition processes (lake infill, soil erosion) or where deposition has preserved samples of material from past climate regimes or human activity cannot be underestimated. Sedimentation rates are not uniform naturally and are changed by human activity, so that temporal information must be derived from independent measurements. For recent periods, the Pb-210 method which depends on the deposition of radon daughters in soils and sediments can cover a period of about 100 years, and can be supplemented by Cs-137 measurements since the 1960 bomb tests.

#### Applications of radionuclides to lithosphere studies

Dating rocks K-40, Ar-39, Rb-87, Nd-143, Sm-147, Hf-174, Lu-176, Re-187, U & Th Dating carbonate deposits C-14, Th-230, U-234 Dating lacustrine sediments C-14, Cs-137, Pb-210, Th-230, U-234 Surface exposure dating Be-10, C-14, Al-26, Cl-36 Soil erosion Be-10, Cs-137, Pb-210 Earthquake monitoring Ra-222 Paleoseismicity and volcanic eruptions Be-10, , Al-26, Cl-36

An AINSE supported study of soil erosion in New England has important implications for soil management in Australia. It showed that the fall in deposition in recent years was not due to improved soil management practices so much as to a greatly reduced reservoir of transportable material due to past management practices. These had caused the relatively rapid removal of all potentially transportable material. In such circumstances the introduction of soil management schemes may not address the serious issue, which is that of reestablishing topsoil. It may be believed that standard conservation methods have produced an improvement, when it was not the rate of soil removal which was the limiting factor but the availability of transportable material. In this study, the normal fall in Pb-210 with the depth and age of the deposits was followed by a relatively constant Pb-210 level before 1916, which was interpreted as a shift at about this time from a regime controlled by transport to one controlled by availability.

This study illustrates the important point that in much environmental research a wide variety of information, historic, political, chemical, and biological as well as nuclear dating methods must be used.

# Hydrosphere

Tritium is widely used as a tracer in groundwater studies. The use of bomb-test tritium is declining along with the tritium activities but increased detection sensitivities through the use of AMS have partly compensated. The importance of groundwater supplies in Australia is obvious but AINSE has funded few studies.

The most widely used nuclear techniques in environmental control is the use of neutron moisture meters for soil moisture determination in irrigated agriculture and horticulture. This contribution to better irrigation and horticultural practices cannot be underestimated.

An example of the many indirect applications of nuclear and radiation science to research into natural water chemistry and biology is our own current measurements of the free radical superoxide, in natural water systems. These are the first direct, dynamic measurements of a free radical in "the wild" and the chemiluminescent technique was developed using gamma radiation to produce known concentrations of superoxide radicals to calibrate our apparatus. We have measured superoxide concentrations for the first time in natural water and found appreciable (>10 nanomolar) concentrations in a fresh water dam and an estuary. The concentrations are sunlight dependent and the data are so new that implications of such appreciable concentrations of superoxide in nature can only be speculative.



#### Applications of radionuclides to continental hydrosphere studies

Replenishment of groundwater sources H-3, Cl-36 Dispersion studies of surface water H-3 Aeration studies Kr-85 Interaction between ground and surface water H-3, C-14, Ra-222 Groundwater dating H-3, C-14, Cl-36, Ar-39, Kr-81, Kr-85 Rock-water interactions, Ra-226, Ra-228, U-234, U-238 Sedimentation rates Cs-137, Pb-210 Radioactive waste disposal Cl-36, I-129

#### Applications of radionuclides to oceanic hydrosphere studies

Circulation and mixing H-3, C-14, Kr-85 Age of water masses H-3, C-14, Ar-39, Kr-85 Transfer of anthropogenic CO<sub>2</sub> to ocean C-14 Dating of ocean sediments C-14, K-40 History of sea-level variations C-14, Th-230, U-234

# Biosphere

The importance of applications to animal and plant ecology, water use, energy economics, and the influence of pollutants on plants and animals, is illustrated by the measurements of the lifetimes of tommy ruffs (deep sea perch) and the influence on stocks and the sustainable extraction rate.

The use of radiotracers in measuring the uptake and biological and metabolic pathways of metals by plants and animals is important in Australia not only for science but also because of the importance of native species to indigenous lifestyles – where native species may become reservoirs of metals such as uranium, radium or plutonium, a detailed understanding of the uptake rates, bio-equilibrium and compartment flows of the metals is needed to manage the use of the animals, and may also contribute to bio-monitoring for the remediation of contaminated mine sites and the reoccupation of the Maralinga test sites. The East Alligator river in NT has been the site of many such studies by ANSTO staff from the Environment Division often in collaboration with universities and important systems have included mussels, tortoises and crocodiles.

Two examples of AINSE supported use of nuclear dating techniques, quite different in kind, illustrate the applications to biosphere problems.

The effects of changes in UV-B flux on phytoplankton have been difficult to establish because in many studies the baseline data and "normal" fluctuation history are unknown and apparent UV-B induced changes may be masked. At one geographically well defined site in Antarctica (Vestfold Hills) where the sediments are free from biologically induced mixing (which can introduce errors of 100 years) the diatom distributions in sediments over a 600 year record show changes in species abundance greater than those since the 1970s when UV-B fluxes increased.

Another study pin-points the origins of a dynoflagellate species in SE Australian waters.

Some years ago outbreaks of a dynoflagellate that can produce toxic effects in shellfish were observed in Tasmanian waters. These are potentially hazardous to local shellfish populations and the origin of the species was important for long-term control. Were they always present in SE waters and increased in population perhaps because of pollution, or were they a recent introduction? The plankton produce a robust cyst which can be identified in sediments, and dating the sediments by Pb-210 methods has established that they were introduced, probably in ships' ballast water.

### Atmosphere and climate

Air pollution analysis, pollutant source identification, atmospheric circulation and exchange processes important for climate, such as  $CO_2$  with the oceans are contemporary areas of nuclear science contributions but the study of climate change in the period out to 100,000 year BP is critically dependent on dating techniques and makes use of a range of nuclides

AINSE supported work has ranged from systems like the perched lakes on Frazer Island, to studies of pollen in sediment and the use of PIXE for the study of the types and origins of particulate matter in urban haze in Sydney and Queensland.

# Applications of radionuclides to atmospheric studies

Dispersion transport and mixing processes on local, regional and global scales H-3, C-14, Kr-85, Ra-222 Transport of water vapour H-3 Stratosphere-troposphere exchange H-3, C-14, Be-7, Be-10, Kr-85 Carbon cycle and sources/sinks of CO<sub>2</sub> and CH<sub>4</sub> H-3, C-14 Atmospheric deposition Be-7, Be-10, Cl-36, Sr-90, Cs-137

Nuclides used in climate studies					
Nuclide	Materials studied	Climate information			
Stable nuclides H-2, O-18, C-13 Anthropogenic radionuclides H-3, C-14, Kr-85, Sr-90, Cs-137 Natural radionuclides: C-14, Ar-39, K-40, Pb-210, Ra-222, Th-230, Pr-231 U-234, U-238	Ocean and groundwater marine and cave sediments and carbonates ice cores and precipitation	Dynamics of oceanic and atmospheric circulation, ocean and air temperature, ice sheet volume, dating of sediments and cave deposits of ocean and ground water, and sedimentation rates			

### The Future

While Medawar properly declared that if he knew the research that would be done in ten years time he would be doing it now, it is easy to see that new applications of nuclear science in environmental studies will again be driven by new and improved technologies.

Some effects can be foreseen without specifying the technologies.

Improved sensitivities and detection limits, such as the way AMS improved on C-14 counting techniques, and an extension of dating techniques because of this will lead to an ability to answer more complex questions and this will involve:

- an increasing interest in "micro" rather than "macro" studies especially for localised biosystems;
- improvements in remediation techniques through the use of nuclear science derived information on plant and animal tolerances and soil science;
- a greater understanding of paleo-climates and the use of this pre-historic data to better model long-term climate changes.

# Getting Warmer? Can Models Predict The Future Of The Earth's Climate?

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### Introduction

Both of the questions posed in the title of this paper were answered in the affirmative by the Intergovernmental Panel on Climate Change (IPCC) Second Scientific Assessment (Houghton et al., 1996). However, the answers contained in the IPCC 1996 report provide only indications of the impacts likely to arise from and responses demanded by climate change. Scientists must now tease out additional questions and determine answers to them in order to provide policy advice for future climate-related mitigation and adaptation activities. Specifically, whilst it is known that numerical models can predict future climates, their accuracy, regional specificity and policy usefulness remain open to challenge and certainly require further refinement and improvement.

# **Global Climate Modelling**

Following from a meeting in Villach, Austria in 1984, the world's climate scientists became increasingly vocal about the potential consequences of greenhouse gas induced global climate change (eg. Taplin, 1996). Scientific research on this topic came to be organised, reviewed and presented by the International Panel on Climate Change (IPCC) in a series of "Assessment Reports" (eg. Houghton et al., 1990, 1992 and 1996). The message from the IPCC Second Scientific Assessment was clear: "the balance of evidence suggests a discernible human influence on global climate" (Houghton et al., 1996, p4).

While the basic physics which underpin the predictions of greenhouse-induced climate change have been understood for over 100 years (eg. Jones and Henderson-Sellers, 1990), the science of global climate modelling is still young (McGuffie and Henderson-Sellers, 1997) and open to challenge (eg. Seitz and Singer, 1996 cf. Hansen et al., 1998 and Wentz and Schabel, 1998).

A climate model is an attempt to simulate the many processes that produce climate. It can be considered as being comprised of a series of equations representing physical, chemical and biological principles. Any model must be a simplification of the real world. In the case of the climate system processes are not fully understood, but they are known to interact with each other producing feedbacks so that any solution of the climate model's equations must be an approximation. The approximations made to the laws governing climatic processes can be approached in several ways leading to a range of different global-scale climate models.

A full general circulation (or global change) model (GCM) is estimated to take about 25-30 person-years to code and the code requires continual updating as new ideas are implemented and as advances in computer science are accommodated. Most modellers who currently perform experiments with the most complex of models modify only particular components of the models. Computational constraints lead to additional problems. For example the coarse resolution of global models cannot represent small-scale atmospheric motions (termed sub-gridscale) such as thundercloud formation. Fine grid models can be used for weather prediction because the integration time is short. Climate models, however, all rely on some form of parameterisation of sub-gridscale processes.

The performance of climate models can only be tested against the past or present climate. Usually when a model is developed an initial objective is to ascertain how well its results compare with the present climate and then to examine the sensitivity of the model to a known forcing such as the solar irradiance distribution changes during a glacial epoch. Although past climates are by no means well known, this comparison provides a very useful step in establishing the validity of the modelling approach. After such tests, the model may be used to gain insight into possible future climates.

The important components to be considered in constructing or understanding a model of the climate system include:

*Radiation* - the way in which the input and absorption of solar radiation and the emission of infrared radiation are handled.

*Dynamics* - the movement of energy around the globe by winds and ocean currents specifically from low to high latitudes) and vertical movements (eg. small scale turbulence, convection and deep-water formation).

*Surface processes* - inclusion of the effects of sea- and land-ice, snow, vegetation and the resultant change in albedo, emissivity and surface-atmosphere energy and moisture interchanges.

*Chemistry* - the chemical composition of the atmosphere and the interactions with other components (eg. carbon exchanges between ocean, land and atmosphere).

*Resolution in both time and space* - the timestep of the model and the horizontal and vertical scales resolved.

The relative importance of these processes and the basis for parameterisations employed in their incorporation give rise to a number of different types of climate models.

Global circulation models (GCMs) incorporate the three-dimensional nature of the atmosphere and the ocean. In these models an attempt is made to represent most climatic processes. They solve fundamental equations including:

Conservation of energy (the first law of thermodynamics);

Conservation of momentum (Newton's second law of motion);

Conservation of mass (the continuity equation); and the

*Ideal gas law* (an approximation to the equation of state of the atmosphere).

The dynamics of the ocean are governed by the amount of radiation which is available at the surface and by the wind stresses imposed by the atmosphere. The flow of ocean currents is also constrained by the positions and shapes of the continents. Ocean GCMs calculate the temporal evolution of oceanic variables (velocity, temperature and salinity) on a three-dimensional grid of points spanning the global ocean domain. The formation of oceanic deep water is closely coupled to the formation and growth of sea-ice so that ocean dynamics demands effective inclusion of sea-ice dynamics and thermodynamics. As well as acting as a thermal 'fly-wheel' for the climate system, the ocean also plays a central role in the carbon cycle, absorbing approximately half of the carbon which is released into the atmosphere every year.

The interactive nature of the plant life of the planet has only recently been included in climate models. The first approach has been to delineate geographic boundaries of biomes vegetation groups characterised by similar species) by using simple predictors available from the GCM such as temperature, precipitation and possibly sunshine (photosynthetically active radiation).

Currently, attempts are being made to evaluate these methods using palaeo-reconstructions of vegetation cover during past epochs. These interactive biosphere models are still in their infancy but may provide useful predictions of future responses of the biosphere including the issue of possible future  $CO_2$  fertilisation of the biosphere.

The climate system is very complex possessing infinite degrees of freedom. Any attempt to model such a multi-faceted system must neglect or represent incompletely many aspects. This process, termed parameterisation, can take many forms. The simplest form is the null parameterisation where a process, or a group of processes, is ignored. The second level, climatological specification, is a form of parameterisation which has been widely used in most types of model. In the 1970s, it was not uncommon to specify oceanic temperatures (with a seasonal variation) and in some of these models the clouds were also specified. In climate sensitivity experiments, it is important to recognise all such prescriptions because feedback features of the climate system have been suppressed. The third type of parameterisation is only slightly less hazardous. Here processes are parameterised by relating them with reference to present-day observations: the constants or functions describing the relationship between variables are `tuned' to obtain agreement.

The most advanced parameterisations have a theoretical justification. For instance, in some two-dimensional zonally averaged dynamical models the fluxes of heat and momentum are parameterised via baroclinic theory (in which the eddy fluxes are related to the latitudinal temperature gradient). The parameterisation of radiative transfer in clear skies is another example. All that needs to be known is the vertical variation of temperature and humidity. Unfortunately, these parameterisations can lead to problems of uneven weighting because another process of equal importance cannot be adequately treated. In the case of heat and momentum transport by eddies, the contribution to these fluxes from stationary waves forced primarily by the orography and the land/ocean thermal contrast cannot be so easily considered. In radiation schemes, the parameterisation of cloudy sky processes is not as advanced as for clear skies.

The interactions between processes in any model of the climate are crucially important. In climate modelling the relative importance of processes and the way that different processes interlink is a strong function of the time-scale being modelled. Whether a system is likely to be sensitive to the parameterisation used for a particular process often depends upon the response time of that feature as compared with other `interactive' features. It is unhelpful to invoke a highly complex, or exceedingly simplistic, parameterisation if it has been constructed for a time-scale different from that of the other processes and linkages in the model.

The time-scale of response is crucially important to all aspects of climate modelling. This time-scale, variously referred to as the equilibration time, the response time or the adjustment time, is a measure of the time the subsystem takes to re-equilibrate following a perturbation. A short equilibration time-scale indicates that the subsystem responds very quickly to disturbances and can therefore be viewed as being quasi-instantaneously equilibrated with an adjacent subsystem which possesses a much longer equilibration time. The response time is generally assessed in terms of the thermal response time. The longest equilibration times in the climate system are those for the deep ocean, the glaciers and ice sheets  $(10^{10}-10^{12} \text{ seconds})$ , while the remaining elements of the climate system have equilibration times nearer  $10^5-10^7$  seconds.

Climate modellers are most confident about the radiative heating by greenhouse gases and cooling by industrial, and other, aerosols (eg. Carson, 1998). We have considerable confidence about our ability to simulate the large scale circulation of the atmosphere and

oceans but as yet have little prediction skill for near-surface continental climates, extreme events or knowledge of the sensitivity to socio-economic forcing functions which drive climate change (cf. Gates et al., 1998).

# The Earth's Future Climate

The most widely known application of climate model predictions currently is the evaluation of the impacts of greenhouse warming. Evaluating future climate in this context is prompting the evolution of climate models to take into account the ramifications of human health; food supply; population policies; national economies; international trade and relations; policy formulation and attendant political processes; national sovereignties; human rights; and international, inter-ethnic and inter-generational equity. In considering the estimated damages due to current emissions of greenhouse gases, the arguments for action are now extending beyond `no regrets' measures: those whose benefits, such as reduced energy costs and reduced emissions of conventional pollutants, equal or exceed their cost. Decisions to be taken in the near future will necessarily have to be taken under great uncertainty. However, these decisions may be very sensitive to the level at which atmospheric concentrations are ultimately stabilised and to the environmental effects on ecosystems: the net productivity of the oceans, the response of trees and forests to carbon dioxide fertilisation and climate change, and methane production by thawing tundra. It is clear that evaluation of this large suite of possible responses to the threat of future climatic projections must incorporate a myriad of issues outside the scope of current climate models but, perhaps, encompassed by future integrated assessment models.

The building blocks for the Kyoto Climate Protocol were established at the UN Conference on Environment and Development held in Rio de Janeiro in 1992 (eg. Taplin, 1996). At this conference, the nations of the world agreed (and later ratified) the "Framework Convention on Climate Change" which establishes an overarching set of guiding principles about human activities that disturb the gaseous composition of the atmosphere. The most significant principle of this "Framework Convention" is Article 2 which states that the ultimate objective of any action should be to "stabilise the concentrations of greenhouse gases in the atmosphere" in way(s) which avoid "dangerous interference with the climate system" but permit "sustainable development". In other words, Article 2 calls for actions which solve the greenhouse problem but at the same time allow the world's economies to develop.

This very challenging dual goal has prompted political activity in a number of countries. In the run-up to the Kyoto meeting, the US Senate voted 95 to zero against specific commitments to greenhouse gas reduction targets unless these included all developing as well as developed nations. Despite this, the Kyoto agreement does commit the developed nations together to reduce their combined greenhouse gas emissions by "at least 5%" from 1990 levels by between 2008 and 2012. Australia struggled throughout 1997 with the concept of committing to greenhouse gas emission reductions below 1990 levels. Economic studies were commissioned and the Federal Government finally declared that the costs to the nation of such reductions were too great both in terms of immediate job losses and for economic development. Despite careful and significant challenges to the economic arguments being employed from within Australia and overseas (eg. Jackson, 1998), the negotiating position adopted was to refuse to reduce emissions: a strategy that was ultimately successful in Kyoto. Australia, Norway and Iceland are the only three (of 39) developed nations allowed to increase greenhouse gas emissions.

It is not anticipated that the Clinton administration will bring their target (7% below 1990 levels) to the Senate for ratification until at least 1999 in the hope that some developing

nations will by then have joined the agreement. The target decrease is compounded by a population increase  $\sim 1\%$  per year and economic growth which are currently increasing American greenhouse gas emissions by about 8% per year. Certainly the USA's target seems unachievable without carbon taxes and other economic tools. The next climate conference in Buenos Aires at the end of 1998 will have to agree the mechanisms for emissions trading among nations.

In one sense, the consequences of the Kyoto Protocol are negligible. Climate models can barely distinguish between a fully implemented "Kyoto atmosphere" and one generated by ignoring (or failing to implement) the new Protocol. Predictions indicate that the very small reductions in ambient greenhouse gas concentrations are likely to diminish predicted temperature rises by only 0.2° to 0.3 °C by 2100. The question for climate scientists now is what is their future contribution (eg. Kerr, 1997; Wigley, 1998).

As yet no formalism exists for validation of coupled climate models but evaluation and confirmation can and must be attempted by examination of the results of fully coupled simulations; model component intercomparisons; and sensitivity studies of interactions between numerical model components (Gates et al., 1996). Modelling and observation communities must jointly strive for improved accuracy (determined by more careful validation against high quality data); enhanced regional to local specificity (gained by model improvements and enhanced validation); and by increasing the skill with which we can detect climate change (observationally driven enhanced by model-based sampling strategies and scenarios).

This paper reviews these challenges using specific examples: better accuracy in terms of continental surface climate prediction (eg. Qu et al., 1998); enhanced specificity in terms of tropical cyclone predictions (eg. Henderson-Sellers et al., 1998); and improved detection in terms of increased understanding of the global carbon cycle (eg. Schimel et al., 1996).

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# **Diverse Applications of Radiation Chemistry**

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Since the early days of radiotherapy, when reliable dosimeters were needed, radiation chemistry has played a crucial role in an increasingly wide range of areas not all directly related to nuclear energy production. Sensitisation/protection in radiation therapy, corrosion chemistry in reactor cooling systems (water and gas), stability of chemical reagents used in spent fuel elements processing, degradation of radioactive waste, food irradiation, sterilisation of medical products, chemical processing, radiation induced electrical breakdown in ceramic insulators, environmental remediation , atmospheric chemistry and the behaviour of electrical discharges. This wide range of application is built on a solid foundation of fundamental work establishing the nature and number of chemical species formed by ionising radiation as it traverses matter. Adding to this, the advent of short pulse, high current electron accelerators enabled radiation chemists to determine precisely the RATE at which these and subsequent processes can occur.

### Energy Losses By Ionising Radiation

Particle Physics	How much energy does the particle / photon lose?
Radiation Chemistry	Where does the energy go within target molecules?
	How long does energy transfer take?
	What are the initial products?
	How do they subsequently react?

Theoretical calculation using Spencer-Fano theory, plus an AINSE studentship, and experimental verification by pulsed e - beam kinetics (ANSTO Febetron), resulted in the first demonstration of the temporal evolution of the fundamental effects of irradiation.

Monitoring the optical emission from excited electronic states of a simple system, eg. neon, revealed excellent agreement between theory and experiment.

Creation Times for Species Gases (P= 1 Torr) Condensed phases of density 1 mg. L<sup>-1</sup>

Ions	inner core	$\sim 5 \times 10^{-9} s$	
	valence shell	$\sim 10 - 20 \ge 10^{-9} $ s	
Secondary Electrons	$1 \text{ MeV} \longrightarrow \text{therma}$	ll ; up to $10^{-4}$ s	
Electronic Excited States	10ns to ~ 100ns		

Irradiated gases contain the same brew of excited species, viz, ions, secondary electrons and neutral excited states as an electrical discharge or plasma.

 $M \xrightarrow{} M \xrightarrow{} M^+ + e^- + M^*$ 

Pulsed electron beam and fast response monitoring techniques have enabled radiation chemists to study and report on areas in gaseous electronics such as

The dynamics of:

Ion-electron and ion-ion recombination

Light production via recombination

Energy loss by energetic electrons

Electron capture kinetics eg.  $e^- + SF_6 \longrightarrow SF_6^-$ 

These studies are important in design and modelling:

high voltage transformers / switching

plasma Discharges- surface etching.

intractible waste destruction via plasma torches.

atmospheric electrical phenomena.

e beam clean up of flue gases.

Radiation chemistry techniques can generate and study these species at ambient pressures whereas discharge physics systems are limited to relatively, (less than 50torr), low pressures. This was especially illustrated by the demonstration of both two and three body processes in ion electron recombination.

#### Radiation Chemistry Of Water

The importance of water as the major component of cells together with the role of water as a coolant in nuclear reactors focussed the attention of chemists for about thirty years. The full mechanism for the action of X rays,  $\gamma$ -rays, electrons, protons,  $\alpha$ -particles is now precisely known. The fundamental species generated in water by radiation can now be utilised with qualitative and quantitative control. Oxidising or reducing conditions can be generated as well as a library of free radicals useable in chemical synthesis.

Following primary ionisation and excitation,

 $H_2O \longrightarrow H_2O^+ + e^- + H_2O^*$ 

Secondary reactions, within  $10^{-12}$ s, establish the essential chemistry,

 $H_2O^+ + H_2O \longrightarrow \bullet OH + H_3O^+$ acidity  $H_2O^* \longrightarrow \bullet OH + H$ 

The •OH radical is a powerful OXIDISER  $e_{aq}^{-}$  and H are REDUCING agents

 $e^-$  +  $nH_2O$   $\longrightarrow$   $e^-_{aa}$ 

#### Quantitative Yields

Precise dosimetry and accurate chemical analysis have produced internationally accepted standard yields from these well defined chemical processes.

Yields are expressed as G values, G, number of molecules produced per 100eV of absorbed radiation energy. Usually 100eV G values are in the range 1-10

In  $\gamma$  , X or  $\beta$  irradiated water, pH 7

$O_2) = 0.75$

The above G values and an established mechanism for the radiolysis of water enables the precise prediction of G ( $Fe^{3+}$ ) = 15.6 for the Fricke Dosimeter which is based on  $Fe^{2+} \rightarrow Fe^{3+}$ . The ability to generate solvated electrons, and hydroxyl radicals in water enables the widest range of selected oxidation and reduction processes to be studied. Inorganic coordination chemistry is able to probe electron transfer processes in transition metal complexes. These molecules are considered as candidates for solar energy conversion systems as well as controlled drug release agents. Radiation chemistry can quantitatively produce and study the redox forms of these compounds whereas photolysis cannot.

Oxidation reactions of the type

$Fe^{2+}$	+	•OH	$\longrightarrow$	$Fe^{3+}$	+	OH
ferrous iron				ferric iron		
Cl	+	•OH	$\longrightarrow$	$\frac{1}{2}$ Cl <sub>2</sub>	+	OH-

Reducing reactions are especially facilitated

$Ag^+$	+	$e_{aq}^-$	$\longrightarrow$	Ag <sup>0</sup> silver atoms, colloids
$\mathrm{Cd}^{2+}$	+	e_aq	$\longrightarrow$	Cd <sup>+</sup> hyper-reduced cadmium
$\mathrm{CCl}_4$	+	$e_{aq}^{-}$	$\longrightarrow$	•CCl <sub>3</sub> + Cl <sup>-</sup>

subsequently

•  $CCl_3 \longrightarrow carbonate + 3Cl^-$ 

 $MnO_2 + e_{aq}^- \longrightarrow Mn^{2+}$  reductive dissolution of manganese dioxide (very insoluble)

 $O_2 + e_{aq}^- \longrightarrow \bullet O_2^$ superoxide

Superoxide is a generator for biologically important free radicals.

The above is a brief list of the types of reactions that can be initiated in a controlled and quantitative way by using radiation chemistry techniques.

The principal control of the quality of an irradiated solution to be oxidising or reducing is quite straightforward.

In a water saturated with nitrous oxide gas, N<sub>2</sub>O, the reaction

 $e_{aq}$  +  $N_2O$  +  $H_3O^+$   $\longrightarrow$   $\bullet OH$  +  $N_2$ 

converts all the reducing species ( $e_{aq}^{-}$  and H atoms) to •OH radicals i.e. an exclusively oxidising medium.

Alternatively addition of a small trace of methanol to water effectively scavenges all the H atoms and •OH radicals and the solution is left as a reducing medium i.e. only  $e_{aa}^{-}$ .

These reactions can now be used to tailor the subsequent chemistry in the solution.

# Electron Transfer

Transport of charge in complex molecular systems is currently a very important area of study. Organic "metals", conducting polymers, light harvesting systems etc rely on electron transfer to achieve charge separation and relaxation. The mechanism of this process through various chemical bond type systems is of crucial importance. Pulse radiolysis techniques as well as laser photolysis has been used to determine the lifetime of charge separation and transport. Skilful molecular engineering can generate donor acceptor compounds of the following type.

The solvated electron in water, alcohols and the highly mobile electron in alkane solvents can be used to probe electron transfer, capture and relaxation in a range of dielectric domains.

The use of scintillation counting techniques in radionuclide assay was significantly enhanced by the radiation chemical studies of the yields of luminescence from a new range of organic scintillators. The fundamental processes of electron scavenging by the scintillator followed by recombination with solvent cations were shown to result in radioluminescence.

# Radiosensitisation-Protection In Radiotherapy

Complex structures similar to that above can be synthesised such that they specifically bind in the minor groove of DNA. These molecules can act as traps for species generated by the radiolysis of the ambient water. These trapped free radicals adsorbed onto the DNA chain can react directly with nearby DNA components leading to DNA damage. Alternatively the adsorbed ligands can react with free radicals created directly on the DNA thus resulting in *repair*. Charge transfer down the DNA chain is crucial to the action of these protective agents. Currently there is heated debate on the assertion by USA photochemists that " DNA is conducting wire". Consequently the whole topic of electron transfer is one of intense importance and activity. The collaboration between organic chemists, radiation and photochemists and molecular biologists is crucial. AINSE projects such as those involving Melbourne University, Peter MacCallum Cancer Institute and the University of Auckland are typical of the interdisciplinary approach needed for this work.

### **Solids**

The electrical and mechanical stability of materials in high radiation fields stimulated the attention of radiation chemists to the study of defects in solids. The coupled use of radiation and ESR enabled the identity of defect structures to be probed. This research led to the development of the sensitive Thermoluminescent **D**osimeters, TLDs and a technique for dating of archaeological pottery artefacts. More recently the ESR detection of free radicals has been used to determine the extent of irradiation in food preservation. The electrical

breakdown of insulators has also been a cross discipline study by radiation chemists and solid state physicists. Recently a workshop in the USA examined the use of ultra high purity sapphire ( $Al_2O_3$ ) as the electrical insulator and first wall material in a high temperature fusion reactor; measurement of the radiation induced conductivity by pulse radiolysis was a significant factor in the decision by electrical engineers to use sapphire for this purpose. One of the processes leading to mechanical and electrical breakdown in solids is the creation of displacement or F centres. Here the irradiating particle transfers momentum to the nucleus of a lattice ion or atom in a crystal. The resultant vacancy can be filled with free electrons liberated by the conventional radiolysis action of the radiation. These trapped electrons are the source of radioluminescence, colour centres, and thermally activated conductivity. Suitably "doped" F- centre lasers are produced by irradiation.

The observation of "threshold" effects in the production of F centres in ceramic oxides followed from pulse radiolysis experiments using the ANSTO Febetron 706 electron pulser. This effect enabled the energetics of displacement of oxygen ions in high lattice energy crystals to be unambiguously determined for the first time. In a series of related refractory oxides eg. spinels and alumina, the displacement energy was found to be in the range 50-58 eV. This technique has much to offer the field of solid state physics.

### **Polymers**

One of the major success stories in radiation research in Australia has been the application of radiation chemistry to polymers and polymerisation. The research groups in Sydney and Brisbane have both demonstrated excellence to AINSE Gold Medal standards in their contributions to polymer initiation, characterisation, degradation and materials science using radiation techniques. A more detailed presentation of this area is the subject of a separate presentation to this conference.

# Future Prospects

Radiation chemistry has explored the fundamental chemical physics of the action of radiation. It can now introduce precisely known amounts of ions free radicals and electronic excited states into gases liquids and solids. Further, the development of time resolved analytical techniques has made pulse radiolysis a powerful tool with which to measure the rates at which the entities derived from radiation action react with their surroundings. Reaction mechanisms can be proposed and modelled with confidence from the large data banks<sup>2</sup> of radiolysis measurements. The following is a list of some of the areas currently under active study using radiation chemistry methods.

- Flue gas clean up of  $NO_x$  and  $SO_x$  by low energy electron beam treatment.
- Rate coefficient measurement for reactions in gaseous electronics. Crucial processes in describing and modelling electrical discharge phenomena such as lighting, welding, gas lasers, atmospheric electrical storms, surface etching etc.
- Chemistry of the storage of radioactive waste from the nuclear fuel cycle eg. the "burping" storage tanks at Hannford USA
- Pulsed irradiation techniques enable the direct observation of the reaction of species important in atmospheric and combustion chemistry.
- Radiation chemistry in medicine is very active with fundamental studies of the mechanism of DNA strand breakage and the development of radiation sensitisers and protectors for therapeutic purposes.

- Radiation induced degradation of pesticides, and intractable materials eg. halocarbons, PCBs etc
- Electron transfer processes in organo-metallic arrays leading to new concepts in producing bio-switches and conducting films.
- nano sized colloidal particles for use in solar cells, electrochromic devices, non-linear optical switches, conducting and fluorescent coatings.
- curing of paints, resins and surface coatings.
- production and property modification to produce novel polymers.

Radiation Chemists and chemistry have spread into a very wide range of applications. It is due to the early careful fundamental work that this field offers a precise qualitative and quantitative base to work from. Even a cursory view of the titles of papers at AINSE Radiation Chemistry conferences will reveal that not just chemistry but applied physics, biology and medicine are areas that are now actively involving radiation chemistry and chemists.

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# Uses Of Neutron Scattering In Supramolecular Chemistry

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#### New Macrocyclic reagents for Metal Ion Recognition

Metal-ion recognition by organic substrates is a topic of fundamental importance and has significant potential application in such areas as environmental chemistry (selective removal of toxic metal pollutants), hydrometallurgy (separation of metal ions by solvent extraction of leach liquors) and in a range of analytical applications. The author's group has been concerned with the investigation of metal recognition involving macrocyclic ligand systems; that is, cyclic organic systems incorporating a central cavity that acts as the binding site for the metal ion.<sup>1</sup> Representative macrocyclic ligands used in these studies are illustrated by (1) and (2): each of these contain mixed oxygen-nitrogen donor atom sets and hence fall intermediate between the classical 'crown' poly ether macrocycles such as 18-crown-6 (3) and the all-nitrogen donor systems such as the tetraaza derivative cyclam (4).<sup>2</sup>



Strategies for obtaining metal ion recognition have been developed by us.<sup>1</sup> Recognition is usually associated with achieving a good degree of steric and electronic complementarity between the metal and its binding site. In a typical study a macrocyclic ligand is designed which is anticipated might show selectivity for a particular metal of interest. The macrocycle is then synthesised and an investigation of its binding behaviour towards this metal ion relative to other ions, both in solution and the solid state, is undertaken. Based on the results of this study, likely factors influencing any observed metal ion recognition of the required type are then assessed and a decision taken as to what structural modification might be carried out on the parent ring to 'tune up' such behaviour. Either the required ligand modification is first assessed by means of molecular modelling programs which may range from a molecular mechanics investigation through to Density Functional Theory (DFT) computations. Using

the above strategies, it is generally possible to modify a ligand type progressively such that enhanced metal ion discrimination is the outcome. In our studies so far, a number of such tuning cycles have usually taken place before the optimal ligand discrimination behaviour has been achieved.

Macrocyclic ligands have proved ideal for use in such studies. Apart from the usual parameters influencing the metal-ion specificity of open-chain ligands (namely, variation of 'backbone' structure, degree of substitution, donor atom type etc.), macrocycles can be further 'tuned' by adjusting the macrocyclic hole size. The potential for using macrocyclic hole size to control the thermodynamic stabilities of the resulting metal complexes thus makes this category of reagent especially suitable for studies of the above type that, for example, involve the recognition and selective binding of a metal ion of given ionic radius in the presence of others with different radii.

Although of fundamental importance for the design of new discriminating reagents, it needs to be noted that there are a number of difficulties in defining macrocyclic hole size. However, in general these need not concern us here. Nevertheless, in the present context it is relevant to outline one study we have undertaken that impinges on hole size when the cyclic ligands incorporate  $OCH_2CH_2O$  groups as part of their macrocyclic rings [as do (1) and (2)].

The neutron diffraction structure of (2). For a considerable period it had been recognised from X-ray and other data that the C-C bonds between ether oxygen donors in crown polyether macrocyclic ligand systems appear shorter than is typical of C-C bonds in simple aliphatic carbon chains.<sup>4</sup> Whether this apparent shortening reflects abnormal thermal motion or is a real effect has been the subject of much discussion.<sup>5</sup> Low-temperature X-ray investigations in which thermal motions are suppressed suggest that, at least in small part, the effect is real.<sup>6</sup> In our studies involving the mixed donor system (2), it was of interest to investigate this aspect with respect to the  $-OCH_2CH_2O$ - linkage in this ligand. It was considered that further information about the nature of such bond geometries would assist in more closely modelling the behaviour of this and related di- and polyether systems. Accordingly, the neutron diffraction study of (2) was carried out<sup>7</sup> (see below) and the results compared with the corresponding X-ray structure of this compound.<sup>8</sup> This comparison was motivated by the realisation that the single crystal neutron technique will give rise to a structure based on the positions of the respective atom nuclei (whereas the X-ray technique yields atom positions based on their respective electron clouds).



Overall, the bond distances and angles in the X-ray and neutron structures were found to be in reasonable agreement. However, the C-C bond in the  $-OCH_2CH_2O$ - string was found to have a value of 1.515(2) Å in the neutron structure which is of intermediate length between the value of 1.494(2) Å found in the X-ray structure and the average (X-ray) C-C length of 1.524(15) Å for straight-chain alkane derivatives listed in the Cambridge Structural Data

Base. The evidence thus suggests that the 'short' bond observed by X-ray diffraction in this and in many other polyether ligands is largely an artefact, with the 'actual' component of any shortening being relatively minor.

### Ligand Assembly and Metal Ion complexation

A major thrust in recent chemical research has been the development of supramolecular chemistry - broadly the chemistry of large *multicomponent* molecular assemblies in which the component structural units are held together by a variety of weaker (non-covalent) interactions that include hydrogen bonding, dipole stacking,  $\pi$ -stacking, van der Waals forces and favourable hydrophobic interactions. Much of the activity in the area has been motivated by the known behaviour of biological molecules (such as enzymes). A feature of much of this recent work has been its focus on molecular design for achieving complementarity between single molecule 'hosts' and 'guests'.<sup>9</sup> We have investigated one such area involving the formation of host-guest adducts between organic molecules which are themselves potential metal-ion ligands. In particular, we have been interested in the situation where the ligand components of a metal coordination sphere 'assemble' spontaneously in the absence of the complexing metal ion. Thus, in the presence of the metal ion, the corresponding metal complex will exist in solution in equilibrium with its corresponding ligand assembly. Our studies have concentrated on ligand assemblies involving a number of amine-containing macrocyclic hosts and aliphatic or aromatic carboxylic acid guests. The results from this work have clearly demonstrated that ligand assembly contributes to metal complex formation (and stability) under appropriate conditions. Thus, if the ligands are present in solution as an assembly of the required stoichiometry (rather than being mutually dispersed throughout the bulk solution) this is expected to result in a favourable contribution towards metal-ion complexation since an element of coordination shell preorganisation is present. That is, the entropy term for complexation by the assembled ligands should be more favourable (on average) than for the situation in which the equilibrium lies towards the corresponding dissociated ligand species. Less 'loss of disorder' on complex formation will occur in the former case.

*Host Guest Adduct Formation between Cyclam and 4-tert-butylbenzoic Acid.* In a number of cases it has proved possible to isolate solid ligand adducts of the type discussed above. One such assembly, the host-guest complex between 4-tert-butylbenzoic acid and cyclam (4), has proved ideal for investigating the nature of the carboxylic acid-amine interaction that occurs in this assembled system.<sup>10</sup> Both nmr and calorimetric titration studies indicate the formation of a 2:1 (acid:cyclam) adduct in chloroform. An X-ray structure of the solid adduct confirmed that two carboxylic acid groups bind directly to the cyclam by means of a network of six hydrogen bonds even though the crystalline adduct was found to contain, in all, four hydrogen-bonded carboxylic acid groups (with only two of these interacting directly with the macrocycle).

The two 4-tert-butylbenzoic acid groups that bind to the cyclam are associated with a network of hydrogen bonds that involves all four NH sites on the macrocycle. Each of these carboxylate groups are bound to the macrocycle via three hydrogen bonds: one strong interaction (2.78Å), one moderately strong interaction (3.05Å) and one weak interaction (3.58Å). Each carboxylic group effectively spans three N-donor atoms on one side of the donor plane of the cyclam. The latter maintains a favourable trans-III configuration in the adduct. Overall, the arrangement of the components remains quite close to the structure required to act as a coordination shell for an octahedral metal ion. For a divalent metal, all that is required is the loss of two protons from the assembly to balance the metal's di-positive charge as it enters the macrocyclic cavity. Indeed, the X-ray structure of the corresponding

nickel(II) complex,<sup>11</sup> prepared from nickel(II) benzoate and cyclam, shows an almost identical arrangement of the ligands in its coordination sphere as that found in the solid metal-free adduct.

*The neutron diffraction structure of the adduct between 4-tert-butylbenzoic acid and cyclam.* The neutron diffraction study of the above adduct was undertaken in order to locate accurately the hydrogen atom positions in the crucial H-bonded network binding the attached carboxylic groups to the cyclam.<sup>10</sup> Such information was not obtainable from the X-ray investigation. It was anticipated that further knowledge of the nature of the hydrogen-bonded network present would enable a fuller understanding of the system and, in particular, provide a background against which the results of planned semi-empirical computational studies could be evaluated.

The neutron data show that each of the strongest hydrogen bonds have an N-H bond length of 1.033(14) Å, which differs in length only marginally from the N-H bond in the weakest hydrogen bonds present. The similarity of these bond lengths can only occur if there has been an effective transfer of a carboxylic acid proton to an amine group on formation of each of the strongest hydrogen bonds. Namely, the result confirms that that the interaction is essentially between a NH<sup>+</sup> group and a -COO<sup>-</sup> group and can hence be considered to be analogous to the formation of a 'host-guest zwitterion'. The thermal ellipsoid for the transferred proton was found to the almost symmetrical in each case and hence gave no evidence that this proton has significant occupancy in other regions between the respective carboxylate and the amine groups. Clearly, the observed strength of the 2:1 adduct observed in solution reflects to a large degree the presence of charge separation arising from the proton transfer discussed above.



Comparison of the Calculated (AM1) Hydrogen Bond Data with the Neutron Structure of the 1:4 Adduct

	Bond 1 Exp./Calc.	Bond 2 Exp./Calc.	Bond 3 Exp./Calc.
N-H (Å)	1.03 / 1.04	0.99 / 1.04	1.00 / 1.00
HO (Å)	1.75 / 1.90	2.06 / 2.05	2.58 / 2.34
NO (Å)	2.78 / 2.89	2.81 /2.82	3.09 / 2.96
N-HO(°	) 172.2 / 159.3	130.9 / 130.2	112.0/119.2

Semi-empirical MO calculations using PM3 and AM1 (within the MOPAC 6.0 package) were successful in modelling the structural features evident from the neutron structure determination reasonably well (see above). In particular, the proton transfer behaviour is predicted by the calculations. The charge distribution within the host-guest assembly was calculated (using AM1) to correspond to near full charge separation; the values obtained were  $1.89^+:0.93^-:0.93^-$ , respectively.

### A Trilayered Supramolecular Array

Co-crystallisation of a 2:1 mixture of benzoic acid and ethane-1,2-diamine from methanol yielded large transparent crystals which proved suitable for study by neutron diffraction.<sup>12</sup>

The structure of the resulting 2:1 supramolecular array once again was shown to contains hydrogen bonded networks in which the acid protons have transferred on to the amine nitrogens (see below); as mentioned earlier, the resulting charge-separated hydrogen bond interactions can be very strong.



The structure incorporates two-dimensional planar layers that are composed of parallel rows of 2:1 acid/amine motifs stacked side-by-side. Each such row is offset with respect to the next. The stacking of these planar layers leads to the adjacent layers being bound together by edge-to-face (T-oriented) aromatic interactions which result in a herringbone arrangement. As a consequence, a pair of benzoate sheets effectively sandwich a sheet of ethane-1,2-diamine molecules lying parallel to be *ab* plane. The layers yield an overall ...*bab'bab'*... pattern where *b* and *b'* represent non-identical benzoate buyers and *a* is the amine layer. Hydrogen bonds also occur between adjacent benzoate participates in three strong hydrogen bonds. Charge separation results in the amine layer being formerly positively charged, with the adjacent benzoate face bearing a formal negative charge.



Interestingly, hydrogen bond geometry optimisations carried out on isolated 2:1 adducts by DFT at the SVWN5/6-31G\* or B3LYP/6-31G\* levels of theory did not predict proton transfer as observed in the neutron structure of the crystalline product. This result was interpreted as suggesting that the charge-separated hydrogen bonds are stabilised by the presence of the extensive hydrogen bond network present in the solid lattice (but which was not specified in the structure employed for the calculations).

Finally, this study has acted as a prototype for the synthesis of a range of other related carboxylic acid/amine arrays - a class of charge-separated assemblies of potential interest as new opto-electronic materials.

#### Acknowledgment

The author thanks his students and co-workers for their contributions to the work discussed in this paper. Their names appear in the list of references.

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<sup>[10]</sup> 

# How γ Radiolysis Gives Unique Information in Emulsion Polymerisations

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#### Introduction

Polymers literally form the stuff of our everyday lives. They are very much the material of the second half of the twentieth century, and doubtless of the next century as well. Both improving the ways of making present polymers in industry, and devising new polymeric materials, require better knowledge of the mechanism of polymer formation. One of the most important means of making polymer involves initiation by free radicals. A subset of this, especially important in industry, is emulsion polymerisation, whereby the polymerisation process is dispersed in water [1]. The process is used for example to make many paints, adhesives and rubber for tyres, as well as high-value-added products such as polymer colloids for medical diagnostic kits. The monomers are often of the formula  $CH_2=CHX$ , where X is a phenyl group for styrene monomer,  $CH=CH_2$  for butadiene (where these two monomers are used in artificial rubber), OCOCH<sub>3</sub> for vinyl acetate (a common adhesive),  $CO_2C_4H_9$  for butyl acrylate (a common paint ingredient), and so on. The process has many advantages: it is environmentally benign (the solvent is water), it is relatively easy to go to high conversion, the final product is a latex, which can be pumped around even though it is 100% polymer, which also facilitates temperature control during manufacture.



Figure 1. An emulsion polymerisation, showing the formation of a latex whose particles contain many individual polymer chains.

The process is summarised in Figure 1. It can be seen that not only does it involve freeradical polymerization, but also colloid chemistry, as well as the complexities of phasetransfer events, such as radicals entering and exiting from particles. The system is a complex one, because so many kinetic events occur simultaneously. Gamma radiolysis as an initiation medium for free-radical polymerisation can give unique mechanistic information. First,  $\gamma$  rays can penetrate opaque media such as a latex (which other radiative initiation techniques such as UV cannot), and second,  $\gamma$  can also be switched off essentially instantly. This enables *radical loss mechanisms* to be investigated independent of other kinetic events. These loss events are radical exit from the particle (which can occur following transfer to a monomer or other small species), and termination of two radicals within a particle, summarised in Figure 2. It also gives a steady radical flux at any temperature, which is particularly convenient for looking at reactions at low temperatures, thereby giving the means of "tuning out" complications that can occur at elevated temperatures. The final trick is to use seeded emulsion polymerisation, whereby a pre-formed and well characterised latex is used as the polymerisation medium, and conditions are chosen so that no new particles are formed. This enables the complexity of particle formation to be avoided, and once seeded results yield information about particle growth. Studies can then be carried out in systems where particle formation is taking place concurrently with particle formation, when enough information on growth is available to deconvolute the mechanisms of particle formation from those of growth (eg., [2]). Because the important information from the  $\gamma$  system comes from relaxation (i.e., after removal from the source), the complexities of the radical initiation process involving gamma radiation [3] usually do not need to be considered.



Figure 2: exit and termination

Methods have been deduced by the Sydney group [4-6] to show how the rate coefficients for radical loss (exit and termination) can be obtained from such relaxation experiments, along with comparison of theories for these quantities. The starting point is accurate measurement of the polymerisation rate, particularly in "relaxation" mode (i.e., after removal from the radiation source. This is relatively simple through automated dilatometry, where the small shrinkage that accompanies polymerisation (polymer being slightly denser than monomer) is monitored through the decrease in height in a capillary attached to the small thermostatted reactor; the entire process has been automated in the TRAKA device designed by Mr David Sangster.

#### **Results for exit**

The kinetics of exit can be studied in so-called "zero-one" systems, where the seed latex is chosen to be sufficiently small that termination is so fast as not to be rate-determining (i.e., the particles can contain only zero or one growing radical [7]). The basic kinetics are then typically [8,9] given by:

$$\frac{\mathrm{d}\overline{n}}{\mathrm{d}t} = \rho_{\mathrm{spont}} - 2k\,\overline{n}^{2} \quad (1)$$

where  $\overline{n}$  is the average number of radicals per particle,  $\rho_{\text{spont}}$  is the rate of spontaneous generation of radicals (i.e., the radical flux after removal form the radiation source), and *k* is the rate coefficient for radical loss. Some results are shown in Figure 3 [4], from which the value of *k* is readily obtained through eq 1. Theory (eg., [10]) suggests that *k* should be given by:



Figure. 3. Conversion and resulting values for  $\overline{n}$  as functions of time for a  $\gamma$  relaxation seeded emulsion polymerisation, after removal from the source.

$$k = \frac{3D_{\rm w}k_{\rm tr}C_{\rm w}}{r_{\rm s}^2k_{\rm p}^1 C_{\rm p}} \tag{2}$$

where  $D_{\rm W}$  is the diffusion coefficient of the exiting radical in water,  $k_{\rm tr}$  and  $k_{\rm p}^{\rm 1}$  are the rate coefficients for transfer and propagation of a monomeric radical,  $r_s$  is the swollen radius of the particle, and  $C_{\rm w}$  and  $C_{\rm p}$  are the concentrations of monomer in the water and particle phases. The fit of eq 2 to a large body of data typified by the results of Figure. 2 [9] is shown in figure. 4. It is seen that the fit is adequate although imperfect, suggesting the acceptable applicability of the exit mechanism of figure. 2. However, the situation is quite different if, unlike the data in figure. 4, the particles are *electrosterically* stabilised (a situation commonly used in industry), when it is found [11] that the model of eq 2 is inapplicable: an area of further, and indeed urgent, work. Recent results [12] have also suggested that this relaxation technique becomes greatly complicated for vinyl esters, where it may be that long-lived nonpropagating radicals are formed in the  $\gamma$  source.



#### Fig. 4. Points: measured coefficients for different radii; line: the fit to eq 2 stabilised by surfactant (SDS).

#### Some results for termination

Termination rate coefficients can be obtained from relaxation studies using a latex that is sufficiently large that termination within a particle is the only radical-loss event. Some  $\gamma$ relaxation data [13] are shown in figure. 5. Data are then fitted to:

$$\frac{\mathrm{d}\bar{n}}{\mathrm{d}t} = \rho_{\mathrm{spont}} - 2\frac{\langle k_{\mathrm{t}} \rangle}{N_{\mathrm{A}}V_{\mathrm{s}}}\bar{n}^{2} \qquad (3)$$

where  $\langle k_t \rangle$  is the termination rate coefficient averaged over the chain lengths of all terminating chains,  $N_A$  is the Avogadro constant, and  $V_s$  is the swollen particle volume. The  $\gamma$ relaxation technique is especially convenient, as with multiple removals and insertions during a single run, data can be simply obtained for many values of the weight-fraction of polymer,  $w_{\rm p}$ . It is noted that the termination rate coefficient depends on the length of both terminating chains [14-17], and thus  $\langle k_t \rangle$  is dependent on radical flux; this dependence is significant but relatively small. In addition to the  $\gamma$  relaxation method, means have recently been developed [18] that for the first time enable  $\langle k_t \rangle$  to be obtained by another completely independent

means: from the molecular weight distribution. Comparison of the  $\langle k_t \rangle$  values can verify the results from both techniques.



Figure 5. Relaxation data for a terminationdominated system (styrene, large latex particles).

The results can also be compared to the extensive theory for this process which is now available [15-17,19], and which in principle enables  $\langle k_t \rangle$  to be predicted solely from quantities such as the dependence of monomer diffusion coefficient on  $w_p$  (which in principle, but not in practice, can be predicted from free-volume theory [20,21], although free-volume theory can be used for interpolation and extrapolation) and the spin-factor *p* [17].

Some results [18] are shown in figure 6, where it can be seen that the two means of measuring  $\langle k_t \rangle$  are in accord. This is a spectacular result because it is the first time that this rate coefficient has been obtained by two independent means ( $\gamma$  and molecular weight distributions), fulfilling an important goal of an IUPAC Working Party on polymerisation rate coefficients [22]. Moreover, the *a priori* theory is able to give acceptable accord with experiment with relatively minor parameter adjustment. This supports the correctness of the theoretical model; however, it suggests that the model cannot be used for highly accurate *a priori* prediction, although it provides good approximate estimates.



Figure 6. Points: experimental data for  $\langle k_t \rangle$  from  $\gamma$  relaxation (**O**), and from molecular weight distribution data at low ( $\Delta$ ) and high ( $\Box$ ) initiator concentration. Broken line: completely *a priori* prediction; full line, prediction from doubling both  $D_{\text{mon}}$  and the spin factor *p*.

#### Spontaneous polymerisation

Because the  $\gamma$  relaxation technique yields  $\rho_{\text{spont}}$ , it therefore provides quantitative data on spontaneous polymerisation. It has been established previously that this is small but significant in styrene [4,7]. New  $\gamma$  data [13] show that this effect is a major determinant of rates and molecular weight distributions in the seeded emulsion polymerisation of chlorobutadiene (Neoprene), which has potential implications for this important industrial product. Because the  $\gamma$  data provide a "Rosetta Stone" for this monomer, one can then obtain many kinetic parameters such as the first reliable values of the rate coefficient for transfer to monomer, elucidation of the contributions from droplet polymerisation, and so on [13]. The fact that the  $\gamma$  initiation facility provides a radical flux that is independent of temperature was crucial in this, the first elucidation of the mechanism for the formation of Neoprene by emulsion polymerisation.

#### **Conclusions**

The  $\gamma$  radiolysis facility at Lucas Heights is unique in the world in its ability to provide quantitative and qualitative mechanistic information about emulsion polymerisation and other free-radical polymerisation processes. This is important for basic science, and in addition, because of the industrial importance of the process, has the potential to give Australian industry as well as science a leading edge. It is also an incentive for attracting overseas R&D funds to this country.

Fruitful collaboration over many years with Professor Don Napper and David Sangster are gratefully acknowledged, as is the support of AINSE and the ARC. Some of the work discussed here has also been supported by AC Hatricks (now Nuplex Australia), and by DuPont; constructive interactions with Drs John Congalidis and John Richards of the latter company are also gratefully acknowledged.

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# **Fusion plasma physics research on the H-1 National Facility**

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#### Abstract

Fusion reactions like those that power the stars have the potential of providing bulk electricity generation with reduced emissions and low radioactive hazard, but pose many challenges in physics and technology. The H-1 Heliac Major National Research Facility now being developed offers Australian scientists and engineers an opportunity to participate in the collaborative international fusion research program. Work on H-1 contributes not only to the realisation of fusion power, but offers the stimulus and opportunity for advanced training and the development of spin-off technology.

### 1. Introduction

Nuclear fusion, in which light elements in an ionised plasma combine to form heavier elements, is the ultimate source of energy in the universe, as it powers stars. Research to develop a terrestrial fusion reactor has been pursued since the 1950s in laboratories all over the world, including Australia. Fusion is attractive as a means to generate bulk electricity with low greenhouse gas emissions and low radioactive waste hazards, as its fuel cycle is based on hydrogen isotopes found in water, and fusion confinement devices have intrinsically lower stored fuel in the reacting core.

Conditions for fusion temperatures of 10 keV (100 million °C), densities of  $10^{14}$  particles/cm<sup>3</sup>, and energy confinement times of the order of 1 second are difficult to achieve. Nevertheless, recent experiments on large toroidal magnetic fusion devices in the US, Europe, and Japan have demonstrated plasma conditions like those in a reactor. But much further work is needed to develop toroidal magnetic confinement schemes that are attractive for commercial reactor applications, and fusion power reactors could become available sometime toward the middle of the 21<sup>st</sup> century. This is the time scale over which for climate effects and electricity supply problems are expected to become serious.

Australia has participated in fusion research with small, fundamental experiments in universities for many years, and Australian scientists have long worked on large fusion experiments all over the world.

In the 1995 Major National Research Facility funding round, ANU and the Australian Fusion Research Group (AFRG) submitted a proposal and won \$8.7M to develop the H-1 toroidal heliac experiment at the ANU Research School of Physical Sciences & Engineering, National Plasma Fusion Research Facility. In April 1997 the contract between DIST and the Host organisation (ANU) was signed, and work on capital improvements to the facility began.

The AFRG is an organisation of six university research groups from around Australia acting under the umbrella of the Australian Institute for Nuclear Science and Engineering (AINSE). The Group was formed in late 1994 with the specific aim of consolidating fusion research in Australia on the large Heliac device at the ANU. The AFRG coordinates national collaboration on the Heliac. It is a recognised AINSE facility for the purpose of AINSE Postgraduate Research Awards. At present the AFRG has participating members from the Australian National University, Central Queensland University, Flinders University, University of Canberra, University of New England and University of Sydney.

# 2. The H-1NF Device

The H-1NF Heliac (shown with the vacuum tank removed in figure. 1) is a medium sized device from the Stellarator family of toroidal confinement devices. These devices use external conductors to produce the helically-twisted toroidal field needed to confine plasma particles. They are closely related to widely studied Tokamak, in which part of the helical field is produced by an induced plasma current, but have the advantage of steady-state operation without current-driven plasma instabilities that can disrupt the plasma column.

There are many specific configurations which use variations on these helical fields. These include a number of *heliotron* and *torsatron* systems developed in Japan (and sometimes referred to together as *helical systems*), *advanced stellarators* developed in Germany, and helical-axis systems called *heliacs* which have been studied in Australia, Spain, the US, and Japan.

The H-1NF device<sup>1</sup> has been operational at low powers for some three years and has already produced some notable papers.<sup>2,3</sup> The MNRF funding is being used to upgrade the machine systems to higher power levels to allow access to higher plasma temperatures and densities enabling research into the stability and confinement of fusion relevant plasmas.

The Heliac design has a helical toroidal axis, motivated by theoretical studies indicating that such plasmas will have good stability properties at high values of plasma pressure. The strong breaking of axisymmetry, combined with a highly non-circular plasma cross section, means that designing a heliac requires a theoretical effort combining the development of new analytical tools and advanced computational methods. The H–1NF device parameters are shown in Table 1.

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Fig 1 H-1NF with the vacuum vessel removed

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Major Radius:	R = 1.0  m
Avg. Minor radius:	<a> = 0.2  m</a>
Toroidal Field:	$B_{\rm T} < 1.0 ~{\rm T}$
	(< 0.2 T DC)
RF Heating:	4-26 MHz
	500 kW
Microwave Heating:	28 GHz, 200 kW
Vacuum Vessel:	Diameter = $4 \text{ m}$
	Height = $4 \text{ m}$
Gas Feed:	Ar, H, He and Ne
	< 300 Torr-l/s



Fig. 2. The H-1NF helical-axis

The resulting plasma has a bean-shaped cross section and a helical axis with 3 periods about the major axis as shown in figure. 2. Although this geometry seems rather complicated and

difficult to model; most of the coils can be circular, which greatly simplifies construction. There is a central conducting ring coil, and simple circular toroidal field coils arranged, offset, around the ring to generate the plasma shape shown in figure 2. An additional helical winding is wrapped around the central ring coil. The major constructional difficulty in this geometry is the threading of the central current conductor through all the toroidal coils and the accurate positioning of the components.

The main advantage of this geometry is that it offers good physics properties with relative ease in construction. It is flexible<sup>4</sup> in that changing the relative currents in the various coil sets can vary the magnetic geometry. From a physics point of view, the geometry has an inherent "magnetic well" that has a stabilising influence on the plasma. Theoretical studies of a linear Heliac model have shown that high normalised plasma pressures  $\beta = p/(B^2/2\mu_0) \approx$  30% can be stably confined. Values of  $\beta \ge 5\%$  are needed for an attractive fusion reactor, and the pressure limit in a toroidal heliac is expected to be set by pressure-driven "ballooning" instabilities which cause the plasma energy to leak across the magnetic field to the wall.

The H-1NF device was designed using state-of-the-art, three-dimensional, computer design tools, and the magnets and supporting structure were constructed and assembled with component location tolerances of  $\pm 1$  mm using the facilities available in the Research School of Physical Sciences and Engineering at the Australian National University.

# 3. Research Program

The facility objectives are fourfold:

- to provide a high temperature plasma National Facility of international standing on a scale appropriate to Australia's research budget.
- to provide a focus for national and international collaborative research, to make significant contributions to the global fusion research effort and to increase Australia's presence in the field of plasma fusion power into the next century.
- to gain detailed understanding of the basic plasma physics of hot plasma confined in the helical axis Stellarator configuration.
- to develop advanced plasma and fusion measurement systems, integrating real-time processing and multi-dimensional visualisation of data.

The first two of these objectives emphasise national and international collaboration. Such collaboration is already well under way in the form of the AFRG nationally and in the formal agreement between H-1NF and the Japanese National Institute for Fusion Science (NIFS), which operates the very large LHD fusion experiment.

Connection to larger research programs abroad is important because H-1NF itself is designed for fundamental physics experiments and not for "parameter pushing" to temperatures and densities where fusion power is actually produced. H-1NF will be used to gain understanding of the fundamental physics of plasma (particle and energy) transport and confinement in the Heliac geometry as well as a test bed for the development of advanced diagnostics for which Australian plasma physicists are justifiably renowned. Collaboration with large programs outside Australia gives Australian researchers opportunities to apply their ideas and equipment on large machines with reactor-grade plasma parameters. The facility has three regimes of operation that depend broadly on the plasma heating used. Scheduling of experimental work in these different regimes depends therefore on the installation program of the different heating systems:

- high-temperature plasma heated by Electron Cyclotron Heating (ECH). Only fixed frequencies are available (28 GHz) which restricts operation to high field (0.5 to 1.0T) and hence moderate plasma pressure.
- high-pressure plasma heated by high power RF in the MHz range giving moderate temperatures and high densities and thus higher beta.
- low-temperature plasmas in the edge of the discharge, an important region where probes can be used. Experiments carried out in this regime link well to studies of the plasma processing of materials that are carried out in Australia.
- these different operating regimes will support investigations in the following research areas:
- finite pressure equilibrium and stability
- transport in high temperature plasmas (~500 eV)
- plasma heating and formation
- instabilities and turbulence
- edge plasma physics
- advanced diagnostic development.

# 4. International Collaboration

The most developed international collaboration for H-1NF is with the Japanese fusion research program. The National Institute for Fusion Science (NIFS) in Toki operates the medium-sized (R = 0.9 m), the CHS (Compact Helical System) experiment, and is constructing a large (R = 4 m) experiment, the LHD (Large Helical System), which is the largest magnetic fusion experiment in the world. Kyoto University operates the Heliotron-E experiment (R = 2.2 m) and is designing a new device that is related to H-1NF by virtue of having a strongly helical magnetic axis.

NIFS and Kyoto University have joined together to collaborate with the Australian fusion program by loaning a 28 GHz gyrotron for use in electron-cyclotron heating experiments on H-1NF. This system, which is worth about A\$1 M, has been installed and tested at the ANU, and awaits the upgrade of the H-1NF magnetic field system to be used in plasma experiments. Japanese researchers will also contribute to the planning and analysis of heating experiments on H-1NF.

NIFS and Australian fusion researchers are also collaborating on diagnostics for LHD and H-1NF, low frequency plasma heating, equilibrium, stability, and transport theory, and 3-D computation. Australian researchers also collaborate on the theory and design of new stellarator configurations and plasma diagnostic development with scientists at the Princeton Plasma Physics Laboratory in the US.

### 5. Recent experimental results

During the low-temperature phase of operation before the magnetic field and heating power, the temperature and energy content of the H-1NF plasma are low enough that small metal probes can be inserted into the plasma. By measuring the current-voltage characteristics of these probes, the plasma density, temperature and electric field can be determined.

Experiments on H-1NF in this regime have already interesting plasma confinement revealed phenomena. For discharges in which the magnetic field exceeds a critical value that depends on the pressure and the magnetic configuration, the density suddenly increases by a factor  $\sim 2$ , the profiles of density and electric field change (figure. 6), and the energy of the ions increases. The outward transport of particles due to plasma turbulence decreases. This is evidence of a transition to an improved mode of plasma confinement; one model for this transition involves reduction of turbulence and transport due to shear in the plasma drift induced by the radial electric field. Such transitions are of critical interest in magnetic fusion research because improvements in confinement directly affect the overall size (and therefore cost) of a magnetic fusion reactor that produces electric power. Typically, the transitions to improved confinement occur in large devices with megawatts of heating power.<sup>5</sup> In H-1NF, qualitatively similar regimes can be attained at low powers ~50 kW and low temperatures, which permits detailed measurements with relatively simple diagnostics.

### 6. Spin-off developments.

The many technical problems that must be solved in doing fusion experiments provide a stimulus to develop new techniques and instruments that can be applied in other fields. Historically, the most important spin-offs have been in the areas of computation and the plasma processing of semiconductors.

During 1997-98, two spin-off activities have developed as a result of research on H-1NF. A new electro-optically modulated solid-state spectrometer (MOSS) was developed for the measurement of line emissions from excited species in plasma



Figure 3. Radial profiles of plasma density  $(n_e)$  and radial electric field  $(E_r)$  in an H-1NF plasma. Profiles taken before (dotted) and after (solid) transition to improved confinement.



Figure 4: Prototype plasma antenna constructed with fluorescent tube.
discharges. This device is much smaller, more rugged, and less expensive than the spectrometers typically used for these measurements, and has potential commercial applications in semiconductor materials processing, plasma chemistry, and related fields. An industrial version of the MOSS is being developed and marketed by Australian Scientific Instruments Pty Ltd.

Members of the H-1NF team worked with the Defence Science and Technology Organisation (DSTO) in Salisbury to develop schemes for using plasma tubes (figure. 4) as antennas in the HF through UHF frequency ranges. These antennas have the advantage of low radar cross section when not in operation, and may make possible the development of novel directional arrays for communications and radar use. ANUTECH Pty., Ltd and the Rapid Engineering Development Centre (REDcentre) are exploring possibilities for commercial development of this concept.

# 7. Conclusions.

The Australian fusion program is centred around the H-1NF heliac, an innovative and flexible experimental facility located at the ANU. Promising experimental results are being obtained in low-power operation, work to increase the heating power and magnetic field is under way, and a network of research collaborations involving Australian and overseas scientists is being developed. Work on H-1 has already produced several inventions whose commercial exploitation is being pursued.

Further information concerning the H-1 National Facility and the AFRG collaborative research can be found on the World-Wide Web at:

http://rsphysse.anu.edu.au/prl/

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# The Rotamak -Contributions from ANSTO and Flinders University

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# 1. Introduction

For the past 30 years, the tokamak has been the frontrunner in the quest for controlled nuclear fusion power. As originally conceived, the tokamak has two fundamental drawbacks. The first is that it is an inherently pulsed device since the toroidal plasma current is produced by induction and cannot be maintained indefinitely. The pulsed nature of the machine leads to severe mechanical and thermal stresses. For the past 15 years or so, a number of research groups in the world have concentrated on producing current drive schemes which are non-inductive in origin and which are capable of driving continuous plasma current in a toroidal device. These efforts have been largely successful and, amongst them, include the pioneering Australian work which has used a rotating magnetic field (RMF) to generate the required continuous toroidal plasma current.

The second drawback in the tokamak approach concerns the appreciably sized 'hole in the doughnut' which is a feature of the conventional tokamak designs. It leads to a wasteful use of magnetic energy and a subsequent uncompetitive price of electricity. The 'compact torus' approach to fusion aims to reduce the size of the central hole to a minimum thus restoring economic competitiveness.

The investigation of plasma/magnetic field configurations of the compact torus variety is of great current interest in the field of fusion research. Two configurations of this genre are the field reversed configuration (FRC) which does not have an externally applied toroidal magnetic field and the spherical tokamak (ST) which possesses such a field. Both of these compact torus concepts, as studied outside Australia, suffer from the very serious disadvantage that they are still inherently pulsed devices since the toroidal plasma current is produced by inductive techniques and cannot be maintained indefinitely.

# 2. The Rotamak project and its genesis

The Rotamak is a compact torus configuration having the unique and distinctive feature that the toroidal plasma current is driven in a steady-state, non-inductive fashion by means of the application of a rotating magnetic field. In its basic form, the Rotamak is operated as an FRC. However, by means of a simple modification, a steady toroidal magnetic field can be added to the basic Rotamak apparatus and the configuration then becomes that of an ST.

The Rotamak project started as follows. Between 1976-79, I had an excellent PhD student called Waheed Hugrass (subsequently my Post Doc; staff member at Los Alamos; and, for the past 10 years, academic staff member at the Department of Physics, University of New England, Armidale, NSW, Australia). He prepared a Ph.D thesis entitled "Production of plasma currents using transverse rotating magnetic fields". In it he profoundly extended the line of research started by Harry Blevin and Peter Thonemann at Harwell/Culham in the early 1960s, both theoretically and experimentally. At the end of Hugrass' Ph.D work, the Rotamak had not yet been conceived but work had been done which demonstrated the usefulness of rotating magnetic field (RMF) current rive.

In August 1978, I attended the IAEA Conference on Plasma Physics and Controlled Nuclear Fusion Research at Innsbruck. One morning before leaving for the conference, I scanned the abstracts of the morning's talks and noticed that Harold Furth was giving a paper on, what was then, a new configuration....the Spheromak. It occurred to me that I knew of a technique to sustain the compact torus configuration, not only the toroidal current, but even the toroidal field. The application of two RMFs should do the trick, one to drive toroidal current and one to drive poloidal current! And, at that moment, the Rotamak was born.

During the Innsbruck conference, Charles Watson-Munro let it be known that there was a new source of research grants for energy research coming into being in Australia (the NERDDC grants) and, as soon as I got back, an application to NERDDC was made (with M. G. R. Phillips and R. G. Storer) for monies to fund research into the "feasibility of establishing a small aspect ratio tokamak (spheromak) field and current configuration by means of rotating magnetic fields, to study the equilibrium and stability of this configuration and to measure the main plasma parameters"; the date of the application was October 5, 1978. We were successful and Rotamak research was under way at Flinders; the project officially started on March 1, 1979. I coined the name 'Rotamak' during the course of 1979. The Rotamak project has been continuously pursued at Flinders University to the present day, a period of nearly 20 years.

In the late 1970s, the AAEC decided that it wished to establish a fusion research program and, to this end, it placed a number of its staff at Flinders, Sydney and ANU with the aim of having them re-trained in the field of fusion physics research. By July 1981, the then Director of the AAEC Research Establishment, Prof. Stuart Butler, had formulated a fusion research program which had been approved by both the Commission and the then Minister for National Development and Energy, Senator Carrick. The program was expected to remain a major component of the Commission's overall program for many years to come. It was also decided that a significant part of the AAEC fusion research program be based on the Rotamak concept; Stuart Butler was an enthusiastic supporter of this approach. The seconded AAEC staff returned to Lucas Heights in 1982 and, for the next 5 years, Rotamak research flourished there in a synergistic relationship with the Flinders group which was both collaborative and competitive.

# 3. The main results from the Rotamak project

It is an invidious task to list what may be regarded as the highlights of an extended and generally successful project. My own listing would include the following (I do not distinguish between the contributions of the Flinders and AAEC groups):

#### 3.1 Theory

- (a) The extension of Hugrass' original RMF current drive theory in an infinitely long plasma column to include the effects of
  - an applied steady azimuthal field on RMF current drive
  - finite geometry, specifically the development of RMF current drive theory in a spherical plasma
  - ion motion.
  - (b) The investigation of the bi-directional toroidal field which, under certain circumstances, can spontaneously appear during RMF current drive in a spherical plasma.
  - (c) The elucidation of the power and momentum relations in RMF current drive.

- (d) The recognition of the existence of nonunique steady state solutions to the RMF current drive equations and the development of a powerful analogy with the induction motor.
- (e) The development of power balance models for spherical Rotamak plasmas.

# 3.2 Experiment

Here I wish to distinguish between the experimental investigations of the reversed field configuration manifestation of the Rotamak, the so-called Rotamak-FRC, and those concerned with the spherical tokamak version, the Rotamak-ST.

#### 3.2.1 Rotamak-FRC

(a) The application of an RMF to a fill of neutral gas both creates a plasma and drives appreciable plasma current. A fully quantitative understanding of RMF current drive in a cylindrical plasma column has been achieved.

(b) Compact torus configurations of the FRC variety (no applied toroidal magnetic field) can be generated in a straightforward manner in a simple apparatus by means of the application of an RMF. They are highly reproducible, stable and of long duration, their lifetimes being governed solely by the duration of the RF pulses which are used to generate the applied RMF.

(c) It is possible to drive more toroidal plasma current, whilst simultaneously preserving the FRC, by increasing the amount of RF power transferred to the plasma. In the most successful Rotamak-FRC experiments to date, a toroidal current of 3600A has been driven with an input of 180kW of RF power into the plasma (Nuclear Fusion **37**, 1505 (1997)).

#### 3.2.2 Rotamak-ST

(a) The application of a steady applied toroidal magnetic field to the basic Rotamak apparatus has significantly increased the amount of driven plasma current and improved the plasma parameters. In the latest experiments (Phys. Rev. Letters **81**, 2072 (1998)), currents in excess of 12,000A are driven and plasmas having  $n_e = 7x \ 10^{18} \text{ m}^{-3}$  and  $T_e = 35 \text{eV}$  are produced. We show that the addition of the toroidal field markedly improves the electron particle confinement time and, consequently, the electron energy confinement time.

(b) The magnetic field lines of a spherical tokamak have been directly reconstructed from experimental data for the first time.

Even though some preliminary work involving the addition of a steady toroidal magnetic field to the basic Rotamak apparatus was undertaken at Flinders in the early 1980s, the ANSTO group's paper entitled:

"Small aspect ratio tokamak configurations generated by rotating magnetic field current drive"

G. A. Collins, G. Durance, G. R. Hogg, J. Tendys and P. A. Watterson

Nuclear Fusion **28**, 255 (1988)

is the first report in the world of a systematic experimental investigation of the spherical tokamak configuration. Their paper arrived at the Nuclear Fusion editorial office one day before that of a rival group from Heidelberg! The ANSTO claim of priority is widely acknowledged throughout the fusion community. It is indeed ironic that the ANSTO group had actually been disbanded by the time that this paper appeared in print.

# 4. Difficulties

An account of the Rotamak project would not be complete without some mention of the difficulties which it has faced over the years.

Firstly, the project suffered in its early days from opinions expressed about it by Australian and foreign plasma physicists which were arrant nonsense and were founded on a basis of complete ignorance. A straightforward reading of the open literature would have convinced any open-minded critic that the theoretical underpinnings of the unconventional concept were indeed sound.

The initial funding by NERDDC was generous. However, for reasons which remain a mystery to me, a decision was taken in 1984 by NERDDC to no longer fund fusion research (I do not believe that the fusion community was consulted at this stage) and the funding of the Rotamak project was subsequently undertaken by the ARC at a significantly lower level. ARC support continued until 1995 when the project failed to gain funding. A change in the title of the 1996 ARC grant application which entailed removing the word 'Rotamak' and replacing it with the anodyne 'The interaction of radio-frequency power with plasma' resulted in a new 3-year grant which terminates at the end of 1998. It is appropriate at this time to gratefully acknowledge the funding that the Rotamak project has received from AINSE over its lifetime.

Probably the most significant blow to the Rotamak project occurred in 1987/88 when an incoming ANSTO Director decided, with unseeming haste, to disband the fusion research group at Lucas Heights at a time when it was performing with maximum impact and productivity. The reasons for this decision are unknown to me; I was not consulted at any stage even though, at the time, I carried the title of Senior Consultant (Fusion). There is no doubt that the success which the Rotamak project currently enjoys would have come at a much earlier time had the ANSTO group been allowed to continue their excellent work.

# 5. Outputs and the future

In a nutshell, the Rotamak project has produced nearly 100 journal and conference papers and given rise to 15 Ph.D theses. Most importantly, it has convincingly demonstrated that meaningful fusion research can be undertaken in an Australian university on a modest budget.

The Rotamak project now leaves the shores of Australia. It has been enthusiastically embraced in both the U.S. and Japan. Investigations of the Rotamak-FRC at higher RF power levels are currently under way at Alan Huffman's group at the University of Washington, Seattle and preparations for a Rotamak-ST experiment are being made by Dick Majesty's group (CDX-U) at Princeton University. Further theoretical investigations of the concept are currently being undertaken at the Institute of Advanced Energy, Kyoto University.

I wish to gratefully acknowledge the valuable support which the project and I have received from colleagues, friends and students over the past 20 years. Diolch o'r galon!

# Low Temperature Plasmas -Fundamentals to Commercial Applications

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#### Abstract

This paper presents a progress report on the collaboration between UNE and ANSTO on plasma nitriding. It is intended as a commemoration of the recent work of Professor S.C. Haydon, who died in October 1998. The paper begins by summarising the gaseous electronics of metastable neutral molecules in nitrogen discharges, and indicates the relevance of this to plasma nitriding. Recent nitriding studies by the ANSTO–UNE collaboration are described, followed by an account of the behaviour of prebreakdown nitrogen discharges with heated electrodes. It is too early to draw firm conclusions from this work, but it currently points to a view of plasma nitriding in which control of impurities is more prominent than hitherto.

#### Introduction

AINSE had invited Professor Syd Haydon to present a paper at this Conference, and he proposed the title above. Following his sudden death on 15th October 1998, AINSE approached me to present the paper in Syd's place. I cannot bring the breadth of experience to the topic that Syd had at his command, and so my paper does not address the early work mentioned in Syd's abstract. Rather, the paper summarises aspects of our work over the past decade or so, during which our interests moved from fundamental studies of the gaseous electronics of nitrogen to applications of the resulting knowledge to the field of plasma nitriding. This development was in no small measure mediated by AINSE. It began, I believe, with a discussion between Syd and Ron Hutchings at an AINSE Council meeting in about 1991. The ensuing collaborative work with ANSTO Advanced Materials has been, and continues to be, generously supported by AINSE.

#### Metastable states in prebreakdown discharges

Diagrams like figures 1-3 were almost ubiquitous in Syd Haydon's presentations during the last fifteen years of his life. They show results from the two types of experiment which came to be regarded as his signature. Figure 1(a) shows the classical Townsend experiment on a prebreakdown discharge: the current between the electrodes is monitored as the electrode separation *d* is altered under conditions of constant reduced electric field *E/N*. (*N* is the number density of the gas between the electrodes.) Electric field values are kept below the level needed to break the gas down, so maintenance of the discharge requires an external mechanism for releasing electrons from the cathode surface; we do it photoelectrically by irradiating the cathode surface with uv light. The great virtue of this experiment is its simplicity: everything happens in an essentially cylindrical volume with constant electric field strength. This makes it possible to extract the physical quantities influencing the behaviour of the discharge with confidence.

Figure 1(b) shows an example of results from such an experiment: the discharge current grows faster than exponentially with electrode separation, eventually asymptoting at a separation called the breakdown or sparking distance  $d_s$ . The  $d_s$  values for nitrogen show extreme sensitivity to the presence of certain impurities, but negligible sensitivity to others.

This struck Syd very forcibly and was the dominant theme of his research in the last years of his life. In the case shown in figure 1(b), the difference between curves (1) and (2) is due entirely to the addition of 70 ppm of  $O_2$  to the gas sample; the 500 ppm of added Ar has essentially no effect. The further increase in  $d_s$  shown in curve (3) is due to isotopic substitution. Such sensitivity to the mass of the molecule indicates a significant role for vibrationally excited states in the discharge dynamics [3].



**Figure 1.** (a) Electrode configuration to measure spatial growth of current in a prebreakdown discharge. The apparatus features accurate measurement of gas pressure (~40–2700 Pa) and all-metal construction. The discharge is maintained photoelectrically by ultraviolet irradiation of the cathode. For further details, see refs. [1,2]. (b) Spatial growth of current in (1) pure <sup>14</sup>N<sub>2</sub>, (2) <sup>14</sup>N<sub>2</sub> with the addition of 501 ppm Ar and 70 ppm O<sub>2</sub>, and (3) <sup>15</sup>N<sub>2</sub> as supplied by the manufacturer [3].



**Figure 2.** (a) Signal-averaged data on the decay of current for a nitrogen discharge (E/N = 283 Td, d = 9.0 mm) after baseline subtraction. The scale on the ordinate is logarithmic above 2.0 and linear below; the time scale on the abscissa is 8.0 µs per channel. (b) Like (a), but at d = 11.0 mm and with the ordinate scale change occurring at 1.0. (c) Data of (b) after subtraction of the slowest exponential component (that due to the second state). The inset shows, on a linear scale, the region near lamp switch off after subtraction of two exponential components; the circled region indicates the fastest component [2].

That these excited states are also metastable is shown in figures 2 and 3. Figure 2 shows examples of the decay of discharge current in the period immediately after the uv light sustaining the discharge is switched off. The current decay occurs in two stages: an initial rapid decrease, lasting  $\sim 20 \ \mu s$ , followed by a slow exponential decay. The initial decrease is caused by the electric field sweeping the electrons and ions out of the discharge gap. The slow decay is due to the diffusion of neutral excited molecules through the background gas.

As these energetic particles reach the cathode, they may cause an electron to be released, thereby prolonging the discharge. Depending on experimental conditions, the characteristic decay time of the exponential decay can range from tens of milliseconds to more than 1 s. In the cases shown in figure 2, the current decay takes about 200 ms. Such long times clearly imply metastability of the particles involved.

Figure 2(a) shows a case in which slow current decay is closely exponential. In this situation, the behaviour is caused by the well known A  ${}^{3}\Sigma_{u}^{+}$  state of the nitrogen molecule. This is shown by the variation of the characteristic decay time  $\tau$  as the gas pressure and electrode separation *d* are varied. Diffusion theory predicts that  $\tau$  should vary as

$$\frac{1}{\tau} = \frac{\pi^2 D_{\rm m} N}{N d^2} + G \,,$$

where  $D_{\rm m}$  is the metastable-particle diffusion coefficients and G is the rate of metastable-state quenching in the discharge volume. According to transport theory, the product  $D_{\rm m}N$ , where N is the gas number density, should be constant as N is varied. Figure 2(a) shows these predictions are well obeyed in our system. The value of  $D_{\rm m}N$  obtained from the slope of the line agrees with other measurements for the A  ${}^{3}\Sigma_{\rm u}^{+}$  state [2]. The zero intercept indicates that the diffusing particle is metastable, in that there is negligible loss from radiative decay.



**Figure 3.** Diffusion plots for (a) the  $A^{3}\Sigma_{u}^{+}$  state of N<sub>2</sub>, (b) the second slowly diffusing state of N<sub>2</sub>. The pressures indicated are reduced to 0° C.

In nitrogen, a real surprise occurs when levels of impurities are very low. Figure 2(b) shows an example: the decay of current becomes much slower than expected from the behaviour of the A state, and takes on a multiexponential character. Figure 2(c) shows the remainder, which is mainly the A-state contribution, after the subtraction of the slowest exponential component. (For full details of the theory and analysis, see ref. [2].) Figure 3(c) shows that the characteristic decay time of the slowest component also obeys diffusion theory, providing very strong evidence that it is caused by a diffusing particle. The nature of the second species is canvassed at length in ref. [4], with the conclusion that it is probably a highly vibrationally excited nitrogen molecule in the electronic ground state. The data in figure 1(b) give additional support to this conclusion [3].

When it is active, the second slowly diffusing state can dominate the discharge, as in figure 2(b). In such a situation, it is no exaggeration to say that electrical breakdown in nitrogen is caused by the action of this state. By deliberately contaminating the discharge gas, we have found that the second state is strongly quenched by both  $O_2$  and CO. As little as 100 ppm of

either gas essentially destroys the action of the second state. On the other hand, over ten times this level of  $H_2$ ,  $CO_2$  or Ne has negligible effect.

# The connection with plasma nitriding – nitriding mechanisms

Nitriding is a widely used technique for hardening the surface of metals, particularly steel. In Australian industry, nitriding is performed chemically: the component to be treated is heated to  $\sim 600^{\circ}$  C in an ammonia atmosphere. Plasma nitriding offers several advantages over its chemical counterpart. It can give better control and reproducibility of the hardness gain. And, as we have shown [5–7], it can be performed at lower temperature than can chemical nitriding. This is important if one wishes to treat alloys that are degraded by high temperature, such as austenitic stainless steel.

Plasma nitriding has been studied for about two decades, so it may be surprising to learn that the mechanisms operating are still not well understood. There remains great controversy over the nature of the active species. The argument between those favouring ion bombardment and those advocating a role for neutral metastable species dates back to two papers from the 1970s [8,9] and is as vigorous as ever today. Intriguingly from our point of view, a series of papers from the Nancy–Orsay Group has pointed to high concentrations of highly vibrationally excited electronic ground-state molecules in dc nitriding plasmas [eg. 10]. It is very likely that the details of the nitriding mechanism vary with the type of plasma and the alloy being treated. As detailed in the next section, we have been studying a low-pressure diffuse rf plasma, and we find evidence that ion bombardment is not important when treating austenitic stainless steel [6], but does assist the processing of low-alloy steel [5].

Another poorly understood aspect of plasma nitriding concerns the role of added hydrogen. Adding hydrogen to the process gas is a common practice in plasma nitriding, and most studies show that this aids the process. Some process regimes call for over 70% hydrogen in the gas mix. Such high levels of hydrogen profoundly alter the dynamics of the discharge, quenching almost all candidates for the active species. Our study [11] therefore indicates that the main effect of high concentrations of hydrogen lies in action at the surface, but detailed mechanisms have yet to be pin-pointed.

# Nitriding studies by the ANSTO-UNE collaboration

The rationale for the collaboration is to bring together our experience in the gaseous electronics of nitrogen with the expertise of ANSTO Advanced Materials in the use of low-pressure rf plasmas for materials processing and in surface-diagnostic techniques. Following a priming grant from AINSE, the collaboration won an ARC large grant in 1994. This funded the construction of a nitriding reactor at UNE and contributed to the installation of a hot-wall furnace at ANSTO. Both facilities reached full operation this year.

In the meantime, we conducted nitriding experiments on several steel alloys using the Mark 1 PI<sup>3</sup> chamber at ANSTO. This chamber is unusual in that the ratio of ion density to excitedneutral density just above the sample varies as the gas pressure is varied, so allowing us to separate the effects of these two classes of species. We installed a large 'plasma-control' electrode to allow us to increase ion bombardment of the sample by biasing the plasma, and we also looked at the effect of biasing the workpiece to several hundred volts. Typical results are shown in figure 4. Here, we use the areas of appropriate peaks in X-ray diffraction spectra as an indicator of nitriding effectiveness. The peaks chosen arise from the nitrogen containing phase of AISI 316 stainless steel called expanded austenite ( $\gamma_N$ ), and the phases known as  $\epsilon$ (Fe<sub>2-3</sub>N) and  $\gamma'$  (Fe<sub>4</sub>N) of the low-alloy steels [5]. In the pressure range shown in figure 4(a), Langmuir-probe measurements and optical spectroscopy show that the ion density in the region of the sample being treated falls monotonically with increasing pressure, whereas the density of excited neutral molecules is a maximum at ~300 mPa [5]. In the biasing experiments (figure 4(b)), ion bombardment to the surface increases successively with sample bias and plasma bias, with little change to the density of excited neutral species. It is clear from figure 4(a) that nitriding effectiveness follows the trend in excited neutral-particle density rather than that in ion density. Figure 4(b) shows that increasing ion bombardment aids only in the formation of the  $\varepsilon$  phase in low-allow steel. It has no beneficial effect when treating austenitic stainless steel in a low-pressure rf plasma.



**Figure 4.** Variation of nitriding effectiveness, as indicated by peak areas in X-ray diffraction spectra, with (a) gas pressure and (b) bias conditions [5]. The peaks used arise from nitrogen-containing phases of the alloys treated. Data from each alloy are separately normalised. The lines are guides to the eye; in (b) they serve only to connect points from the same phase and material.

These conclusions are based on peak areas in X-ray diffraction spectra, but we find similar trends with other diagnostics, such as increase in surface hardness or roughness, size of nitrogen peaks in proton-backscattering spectra, or thickness of the treated layer as determined by scanning electron microscopy or secondary-ion mass spectrometry [5-7,11-13].

Recent results from the hot-wall reactor and the new reactor at UNE are very interesting in view of our results from prebreakdown discharges. Figure 5 shows nitrogen concentration as a function of depth for samples treated in three different reactors under conditions as closely matched at possible given the differences in reactor geometry. The increased nitriding effectiveness in the UHV chamber at UNE compared with the Mark 1 PI<sup>3</sup> chamber at ANSTO may be due to the higher level of process-gas purity. If so, this supports the contention that metastable species are important in nitriding. The further improvement shown by the hot-wall reactor is especially intriguing. During a treatment, the whole reactor is raised to the process temperature of 400°C. This produces significant outgassing from the chamber walls, probably leading to rather clean conditions. One might expect this to improve with further use.

The prebreakdown experiments described in the next section show that raising the temperature of the electrodes increases secondary ionisation in the discharge. So far, it is not clear whether this is due to an increased population of metastable states flowing from reduced quenching in the discharge volume, or whether it is reflects an increase in secondary-electron emission because of changes to the cathode surface. Either way, the prebreakdown experiments led us to predict that the hot-wall reactor would give better results simply because its walls are hot. It was gratifying to see this prediction borne out.



**Figure 5.** SIMS depth profiling for samples of AISI 316 stainless steel treated in three different reactors, with treatment conditions as closely similar as possible. The sputter yield of the NCs<sup>+</sup> cluster ion is shown as a function of depth below the surface of the samples [7,12]. 'UHV' refers to the new reactor at UNE.

#### Prebreakdown discharges with a heated cathode

In the last four years, we at UNE have explored in some detail prebreakdown discharges under conditions of relevance to nitriding. Initially, we looked at high values of E/N (up to 2 kTd) and at gas mixtures containing large percentages of H<sub>2</sub>. We also replaced our usual copper cathode with one made of AISI 316 stainless steel, and this turned out to be a confounding factor in the experiments. For over a year, our beloved second slowly diffusing state refused to show itself. Finally, Syd convinced the rest of us that the answer had to lie in impurities being released by the stainless-steel cathode. We decided that the only way to test this was to construct a heated electrode which we could take up to temperatures typical of nitriding.

Figure 6 shows some results from this work. These data were taken with a new stainless-steel surface, which was outgassed in vacuum at progressively higher temperature. After each outgassing episode, the cathode was returned to room temperature and the spatial and temporal growth in current was measured in 5.0 torr of N<sub>2</sub>. As figure 6(a) shows, the outgassing produced successive increases in secondary ionisation up to 350°C. The 400°C episode gave no further improvement. The temporal-decay data show that the second slowly diffusing state is present once the stainless-steel surface has been outgassed at 250°C or above, but it is not present with degassing at lower temperature. This represents rather a triumphant vindication of Syd's point of view.

The data in figure 6(a) were collected with the cathode cold. The real question, of course, is what happens with a cathode at 400°C, the temperature of our nitriding process. Figure 6(b) is a first examination of this point. These data were taken immediately after the experiments of figure 6(a). The most striking feature is the exceedingly small breakdown distance, by far the smallest that we have observed at this value of E/N. However, our excitement must be tempered with the realisation that the E/N value will be altered by the presence of the hot cathode. It is difficult to determine the temperature gradient, and hence the gradient in N, in the discharge gap under these conditions, so we are now contemplating the design of a hot-wall chamber for prebreakdown measurements.



**Figure 6.** (a) Spatial growth in ionisation in nitrogen at E/N = 283 Td following vacuum degassing of the stainless-steel cathode at the temperatures indicated. The curves for 350°C and 400°C are essentially coincident. (b) As in (a), but while the cathode is at the temperature indicated.

#### 6. Conclusions

Our phenomenological studies of plasma nitriding have demonstrated that a low-pressure rf plasma can harden the surface of austenitic stainless steel without loss of corrosion resistance. This is difficult to achieve with the more conventional technique using a dc discharge. The phenomenological studies also indicate that the presence neutral metastable molecules is important for efficient nitriding. Ion bombardment may be important for some materials, but is not generally so. About 10% of added hydrogen also assists the process, as does the use of a hot-wall furnace.

Both the fundamental studies of prebreakdown discharges suggest that control of gas composition is a significant factor for nitriding. By reducing quenching impurities, one allows secondary processes to increase the level of ionisation and hence the activity of the discharge. The extent to which the second slowly diffusing state of nitrogen plays a role is still not clear.

# **Acknowledgments**

A principle purpose of this paper is to highlight the seminal role played by Syd Haydon in our research collaboration with ANSTO. As one worked with Syd, it was remarkable to observe how often experiment showed his views and ideas to be essentially correct. However, it is also the case that this work is the product of a rather large research team, all of whom have made significant contributions. As usual, much of the hard slog was carried out by graduate students: Allan Ernest, Matthew Baldwin and Jayson Priest. Sunil Kumar spent two years with us as an ARC-funded Postdoctoral Fellow, bringing particular skills in surface diagnostics, especially electron microscopy. And finally, little of this would have been possible without our ANSTO collaborators: George Collins, Ken Short, John Tendys and Ron Hutchings in Advanced Materials, Kathryn Prince and Patrick Burke on the SIMS, and Nick Dytlewski with the proton-backscattering work.

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# **Radiolanthanides in Therapeutic Nuclear Oncology**

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Radiolanthanides such as holmium-166 (T1/2 26.8h), samarium-153 (T1/2 46.3h) and lutetium-177 (T1/2 6.7 days) all have beta emissions suitable for radiopharmaceutical therapy. They also emit gamma photons of energies which permit quantitative imaging on conventional gamma cameras which facilitates calculation of dosimetry in individual patients. The range of half-lives has the potential to match the irradiation of tumour cells to the residence time of the radiopharmaceutical thus minimising radiotoxicity to normal tissues. Relative ease and low cost of production of these radiolanthanides in reactors such as HIFAR at ANSTO render radiolanthanides a practical option for radiopharmaceutical treatment of cancer for patients in whom conventional therapy has failed.

Samarium-153 ethylenediaminetetramethylene phosphoric acid (EDTMP) has been developed in clinical trials over the past decade for pain palliation of disseminated skeletal metastases arising from prostate and breast cancer. Response rates of 87% have been reported with 25% of patients becoming pain-free. Prospective individual dosimetry is performed at Fremantle Hospital using a tracer activity of <sup>153</sup>Sm-EDTMP a new whole body imaging method has been validated against the urine sampling technique which we originally developed to limit the radiation exposure of red marrow to 2 Gy to avoid haematological toxicity from this bone-seeking radiopharmaceutical. Re-treatment in patients who relapsed following initial pain response to <sup>153</sup>Sm-EDTMP has also produced gratifying results.

Both samarium-153 and holmium-166 when chelated with bone seeking chelates (EDTMP and DOTMP respectively) and given in high dose may be used to ablate bone marrow, in combination with chemotherapy, to treat haematological malignancy. We have ablated marrow in experimental animals using a combination of <sup>153</sup>Sm-EDTMP and Melphalan, and with subsequent bone marrow transplantation treated murine myeloma with encouraging results. A clinical trial of samarium-153 -EDTMP bone marrow ablation is contemplated at Fremantle Hospital and early clinical data from trials of <sup>166</sup>Ho-DOTMP will be presented.

In experimental studies in pigs we have validated a method for estimating dosimetry in the liver following intra-hepatic arterial administration of <sup>166</sup>Ho-microspheres. A phase 1 clinical trial of this therapy is in progress at Fremantle Hospital in patients with liver cancer and preliminary results will be reported.

Radioimmunotherapy of cancer using various monoclonal antibodies targeted to specific tumour-cell antigens has been performed with samarium-153, holmium-166 and lutetium-177 labelled antibodies in human tumour xenografts in nude mice and in clinical trails. Development of a large animal model of human cancer is described which relates to improvements in the accuracy of prediction of dosimetry and preclinical evaluation of efficacy and toxicity of radiolanthanides in therapeutic nuclear oncology.

# Development of Fission Track Dating and Thermochronology in Australia

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The concept of fission track dating was developed in the very early 1960s by three physicists, Robert Fleischer, Buford Price and Robert Walker, then working at the General Electric Research Laboratories in Schenectady, New York. The basic principle for this new dating technique was elegant in its simplicity: spontaneous nuclear fission of <sup>238</sup>U produces radiation damage trails, or fission tracks, in natural uranium-bearing minerals and glasses, which steadily accumulate to leave a record of geological time. From their initial discovery of naturally-occurring fission tracks and the subsequent calculation of some rudimentary fission track ages it took nearly two decades of research to realise the full potential of the new dating method for routine application in a range of geological environments.

Initially fission tracks were observed in thin cleavage flakes of synthetic mica as diffraction contrast images under transmission electron microscopy, a procedure which was impractical for routine observation in most natural materials for two reasons. First, sample preparation procedures were at best cumbersome, and unsuitable at all for many natural materials. Second, the tracks gradually faded and disappeared while being observed under the electron beam. The second problem was solved by briefly etching the mica in hydrofluoric acid, which effectively "fixed" the tracks for EM observation, but also showed them to be highly chemically reactive. It was soon discovered that prolonging the etching treatment enlarged the fission tracks to optical dimensions so that they become visible by conventional optical microscopy. This discovery was the critical breakthrough from this early period and enabled fission tracks to be easily observed, and measured, in a wide range of natural minerals and glasses. By the mid 1960s it had been shown that not only micas, but a range of natural uranium-bearing minerals and natural glasses contained fission tracks from the spontaneous fission decay rate for <sup>238</sup>U, a geological age could be calculated from the number of fission tracks observed on a polished surface cut through the track-recording material.

To calculate a quantitative age in millions of years it is also necessary to know how much uranium is present in the material being dated. This is determined by irradiating the sample with thermal neutrons to produce a second set of fission tracks from induced fission of a small fraction of the <sup>235</sup>U present. Knowing the natural isotopic ratio of <sup>235</sup>U/<sup>238</sup>U as well as the <sup>238</sup>U spontaneous fission decay constant, a geological age can be calculated from the measured area-densities of 'spontaneous' and 'induced' tracks.

Fission track dating was introduced into Australia by Professor John Lovering during the mid sixties who, along with several others at the time, saw this as an important new opportunity for dating the enigmatic materials related to extra-terrestrial impacts, known as tektites. The requirement for thermal neutron irradiations meant that AINSE was involved in supporting this work from its inception. In the late 1960s a PhD student of Lovering's at the Research School of Earth Sciences at ANU, John Kleeman, discovered an important method of using induced nuclear particle tracks for mapping the distribution of uranium (along with Li and B)

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on a microscopic scale. The Lexan-print method used a polycarbonate track detector to reveal an image of neutron-induced <sup>235</sup>U fission tracks from a polished rock thin-section within a background 'print' of small alpha-particle pits from n- $\alpha$  reactions with Li and B in the rock. This led on to a range of studies of the geochemistry of uranium in a variety of terrestrial and extra-terrestrial materials, culminating in studies of moon rocks from the Apollo program following Lovering's move to the University of Melbourne in 1969.

In Melbourne during the early 1970s the emphasis returned in earnest to developing techniques of fission track dating for the routine analysis of common uranium-bearing minerals apatite, zircon and sphene, which occur widely in ordinary rocks. This work continued to be supported by AINSE and many basic analytical and calibration issues were systematically worked out at this time. Up until this time there was little standardisation in laboratory procedures and inter-calibration between different laboratories remained a major issue. Gradually a consensus emerged as to appropriate standardised procedures and calibration against an agreed set of age standards, which were then widely distributed. The Melbourne group together with a number of international collaborators played a leading role in the universal adoption of these more unified approaches. Another important technical innovation in the late 1970s in Melbourne was the development of an automated microscope stage-system, which dramatically increased the productivity of fission track dating by the now widely preferred 'external-detector' method. These stage-systems have now been adopted in virtually all fission track dating laboratories around the world.

It was realised quite early that fission tracks exhibit only limited thermal stability and were known to undergo fading, or annealing, when heated to even relatively low temperatures over geological time-scales. Initially this was seen as a fundamental limitation of the method for geological dating, but it was gradually appreciated that this property could be turned to advantage for studying the thermal history, rather than the age of formation, of rocks. Throughout the 1970s laboratory annealing studies on a wide range of materials in various laboratories showed the broad temperature-time dependence of fission track annealing and allowed a qualitative interpretation of fission track ages in different minerals in terms of cooling through a series of critical annealing temperatures. Apatite fission-track ages, for example, were interpreted as the time of cooling to approximately  $100\pm20^{\circ}$ C and sphene to approximately  $300\pm50^{\circ}$ C over millions of years. By the end of the 1970s it had been recognised that fission track dating would have a quite different role to other, more established geological dating methods, and that the potential existed for extracting significantly more information from the fission track record.

The development of more quantitative analytical techniques for the reconstruction of thermal histories was hampered by the limited precision of laboratory annealing studies at that time. At the time such studies were all based on measurements of track densities, ie the number of tracks per unit area, which generally lacked sufficient precision for confident extrapolation from laboratory to geological time scales. A new approach was needed. During the 1980s a series of studies were published by the Melbourne group based on a new kind of fission track parameter – the lengths of confined fission tracks. Confined tracks are those which are completely enclosed within the body of the mineral, being etched either from a crack, or from another, surface-intersecting track. Confined tracks reveal the full etchable range of the fission track and can be measured much more precisely than track densities, thus enhancing the quality of geological predictions based on laboratory annealing data. In addition, all fission tracks initially have closely similar lengths so that departures from the initial distribution of confined track lengths provide a record of the style and extent of past thermal annealing.

In geological annealing, each fission track behaves like a maximum-recording thermometer, shrinking to a length which is characteristic of the maximum temperature that it has experienced. As new tracks are continually added to the mineral by spontaneous fission decay, a distribution of track lengths results which provides a complete record of the thermal history experienced at temperatures below about 120°C. Above this temperature virtually all fission tracks are completely annealed over millions of years and no fission track age is recorded. The lengths of annealed tracks this can be compared with the lengths of fresh, unannealed tracks to give a precise estimate of the amount of annealing which has occurred over the time that track has been in existence.

A combination of these new approaches based on track length measurements with the earlier developments in fission track dating became known as 'fission track analysis'. During 1980s fission track analysis helped to establish the new field of *thermochronology*, the quantitative reconstruction of past variations in the temperature of geological environments. Apatite fission track analysis became a routine tool for studying the low-temperature regime characteristic of the upper 3-5 km of the Earth's crust where thermal gradients are typically around 20-30°C/km.

With these new techniques it became possible to predict the fission track parameters which would result from any given thermal history, and these could then be compared with actual observations to assess the likely thermal evolution of a rock sample, essentially a forward-modelling procedure. Based on these techniques a new phase of geological applications flourished, including ventures into fields, such as the study of sedimentary rocks, where there had previously been little role for radiometric dating studies. For example, it was recognised around 1980 that the temperature interval over which track annealing occurs in apatite was similar to that required for oil generation in sedimentary basins, and an important new direction for fission track analysis became apparent. This discovery ultimately led to the routine application of fission track analysis to oil exploration and the establishment of *Geotrack International Pty Ltd* by the Melbourne group to provide fission track analytical services to the international oil industry.

During the 1980s the Melbourne group became the largest working in this area anywhere in the world and established Australian research at the forefront of this field. The group moved again, in 1988, with Professor Gleadow to La Trobe University where the long association with AINSE has continued. In this new environment a new phase of research into the diverse geological applications of fission track thermochronology began. These have included: continued studies of sedimentary basin analysis, continental extension tectonics, the formation of rifted continental margins, the evolution of mountain belts, and the long-term stability of ancient shield areas of the crust.

Over the last six years the group has played a significant role in the Australian Geodynamics Cooperative Research Centre (AGCRC) under whose auspices the newest developments of fission track thermochronology have taken place. An important achievement has been the development, with Dr Kerry Gallagher of Imperial College, London, of a practical procedure for solving the inverse-modelling problem, ie reconstructing the thermal history directly from the observed fission track parameters. This is actually a 'pseudo-reversed' modelling procedure based on a Monte-Carlo simulation of many possible thermal histories and testing of these against the actual observations. A genetic algorithm is used to accelerate the convergence to an acceptable result. In this way it has become possible to automate the reconstruction of likely thermal histories for large arrays of samples covering a wide area of the crust and, for the first time, to investigate the variation in underlying geological processes on a regional scale. The results can be combined with other regional data sets, such as surface heat-flow and digital elevation models, to examine a new class of problems associated with the long-term evolution of the landscape. The results are interpolated and visualised using computer imaging techniques. Time-sequences can be displayed for various calculated parameters, such as the variation of sample temperature, the amount of surface denudation, and the palaeo-elevation of the earth's surface. Results for hundreds, and even thousands, of fission track samples can now be analysed together using this regional imaging approach, achieving a greater level of understanding than was possible with earlier studies of individual or small groups of samples.

A basic assumption, on which part of this regional imaging approach is based, is that the cooling experienced by rocks in the upper few kilometres of the crust is primarily due to their gradual approach to the surface by the denudation of the overlying rocks. This surface denudation is likely to be the dominant control on near-surface cooling in many geological environments, especially in tectonically-mature basement terranes characteristic of much of Australia. Quantification of the cooling history enables the rates, timing and amounts of surface denudation to be estimated and their regional variations to be elucidated. By 'back-stacking' the amount of removed material onto the present land surface and restoring this to gravitational equilibrium it is possible to reconstruct variations in the land surface at various times in the past. Other explanations for cooling, besides surface denudation, may be responsible for variations in temperature in certain environments. These include such processes as fluid flow and igneous activity, especially in sedimentary basins, active tectonic environments in young mountain belts and active igneous provinces. In these areas fission track thermochronology can also be used to reconstruct the thermal histories and study the processes in operation.

A major project now under way within the AGCRC is to use fission track thermochronology to develop a series of thermotectonic images of the whole continent of Australia. Measurements on nearly 3000 samples over much of the exposed basement rocks of Australia have now been produced and modelling work is proceeding towards an initial continental overview in the year 2000. At the same time work by other members of the group is also producing similarly large data sets for other continental regions, particularly in southern and east Africa, which are capable of the same kind of analysis. Many ore deposits in the continental crust are formed in close relationship to the land surface and there is a growing realisation that understanding near-surface, rather then deep-seated, processes will be important in fully understanding these. This recognition, and the insights coming from continental-scale imaging, are now providing further ideas and approaches to the study of the Earth's crust based on low-temperature thermochronology which will continue to build on the research opportunities provided in Australia by AINSE.

# PARTNERSHIPS IN NUCLEAR SCIENCE AND TECHNOLOGY CONTRIBUTING TO THE 'HIGH ROAD' TO THE FUTURE

Professor Helen Garnett Chief Executive Australian Nuclear Science and Technology Organisation,

The focus of this AINSE conference is a celebration of the many contributions that nuclear science and technology have made to the development of knowledge over the past forty years. While understanding and reviewing the past can help us along the road to the future, and in particular help us avoid mistakes of the past, such an historical review cannot determine what the future road will be.

What is possible is to scope possible scenarios taking account of a wide variety of inputs from many and varied sources; to assess the facts and the trends in many fields and then to couple this information with some 'dreaming' and 'what if' questioning. We are in a knowledge revolution. The available information is currently doubling every seven years. For socioeconomic development some of the knowledge, currently available and to be generated in the future, must be converted into value, into innovative products, services and solutions which will give rise to wealth creation.

What are the likely sources of wealth creation in the first twenty years of the next century? Economists, engineers and scientists seem to agree that the 'high road' for Australia is likely to involve the exploitation of a range of advanced and emerging technologies for which a reasonable scientific base has been developing over the past twenty years of this century. These include communication technologies, biotechnology, nanotechnology, biomedical engineering, novel resource processing technologies, environmental technologies, advanced transportation technologies and smart food technologies together with participation in large 'infrastructure' projects which will generate a quick start in Asia.

Technological developments require further scientific questions to be answered and a raft of enabling technologies. Underpinning many of the emerging technologies, considered likely to contribute to socioeconomic development, are both materials science and various processing technologies. There is a requirement to understand the structure and function of natural and man-made molecules and changes in the structure of materials during processing steps and/or functional performance in a range of environments. There is a need to better understand natural processes.

An analysis of the structure, function and performance of materials requires the use of the fundamental probes, neutrons, charged particles and X-rays. Understanding natural processes is facilitated by isotopes. The past has delivered a solid knowledge base for understanding the properties of these probes and their potential applications. Each has unique properties and access to all types of probe is required. Recent innovations, in materials development, have themselves resulted in the design of facilities to deliver these probes more efficiently and effectively and in the construction of better detectors to 'visualise' the outcome, thereby allowing a broader range of questions to be answered. The crystal ball cannot forecast specifically what creative ideas will emerge, innovations that will be considered nor what specific questions will require solution. However, trends show that the development cycle for new products is quickening. Access to solutions for materials and processing problems will be required in an ever-decreasing time span for any company wishing to maintain the

competitive edge. Nuclear science and technology can and will be an essential component of this innovation cycle, assisting Australian enterprises to be at the leading edge of competitive markets.

AINSE has enabled and fostered projects across scientific disciplines and partnering between institutions has improved. However the effective application of the appropriate nuclear probe/s to solving questions, in materials science and the processing industries, of direct relevance to advancing the emerging technologies and their innovative application to provide high value products and services, will, almost certainly, require different ways of working. It will be up to the scientific community to show that they can make a difference. Multidisciplinary teams, including not only scientists and engineers but consisting of all disciplines involved in a product life-cycle, or value chain, on a scale not common for current AINSE projects, will almost certainly be required; improved communication and genuine inter-institutional teaming for specific projects will be essential. ANSTO and others are attempting to address these issues. However if the benefits of nuclear science and technology are to be captured, embracing such new partnership models will need to become commonplace in the years ahead.

We will plan and commission facilities, we will foster the scientific capabilities but we must not just create knowledge, but also create the opportunities to assist in adding value and thereby make a difference.

We must dream about what might be, brainstorm as to what should be and then influence the external environment and create networks to make the 'high road' a reality. We must share our dreams.

# **APPENDIX – List of members and abbreviations**

ADE	Adelaide University
AKL	University of Auckland
ANSTO	Australian Nuclear & Scientific Technology Organisation
ANU	Australian National University
BAL	Ballarat University
CBR	University of Canberra
CQU	Central Queensland University
CSU	Charles Sturt University
CUR	Curtin University
DEA	Deakin University
ECU	Edith Cowan University
FLI	Flinders University
GRI	Griffith University
JAM	James Cook University
LAT	Latrobe University
MAC	Macquarie University
MEL	University of Melbourne
MON	Monash University
MUR	Murdoch University
NCT	Newcastle University
NSW	University of New South Wales
NTU	Northern Territory University
QLD	University of Queensland
QUT	Queensland University of Technology
RMI	Royal Melbourne Institute of Technology
SCU	Southern Cross University
SWI	Swanbourne University
SYD	University of Sydney
TAS	University of Tasmania
UNE	University of New England
USA	University of South Australia
USQ	University of South Queensland
UTS	University of Technology Sydney
UWA	University of Western Australia
UWS	University of Western Sydney
VIC	Victoria University
WOL	University of Wollongong

# Glossary

14 UD	pelletron accelerator at ANU	
4 H1 and 4 H2		
	neutron beam facilities on HIFAR	
6 HGR8,9 and 10, 4 H1 and 4 H2 neutron beam facilities on HIFAR		
AAS	atomic absorption spectrometry	
AFRG	Australian Fusion Research Group	
AMS	accelerator mass spectrometry	
CERN	European Centre for Nuclear Research in Grenoble France	
EN	6 MV tandem accelerator built by High Voltage Engineering Corporation	
ERD	elastic recoil detection	
FN - 9 MV	tandem accelerator built by High Voltage Engineering Corporation	
FRS	forward recoil scattering	
GATRI	<sup>60</sup> Co gamma facility suitable for research and development projects and small scale industrial irradiations	
HIFAR	High Flux Australian Reactor, 10 MW	
HRPD	High Resolution Powder Diffractometer	
IBA	ion beam analysis	
ICPES	inductively coupled plasma emission spectroscopy	
ILL	Institut Laue-Langevin	
INAA	instrumental neutron activation analysis	
IUPAC	International Union of Pure and Applied Chemistry	
LEIS	low energy ion scattering	
MEIS	medium energy ion scattering	
MEVVA	ion beam implanter, 1.3 MeV electron beam accelerator	
MOATA	100 kW research nuclear reactor	
NRA	nuclear reaction analysis	
PIGME	particle induced $\gamma$ -ray emission	
PIXE	particle induced X-ray emission	
RBS	Rutherford backscattering	
XRF	x ray fluorescence	