PROGRESS REPORT FOR ALNGRA10130

APPLICATION OF GEOPOLYMER BINDERS IN NUCLEAR WASTE IMMobilIZATION

INVESTIGATOR(S)

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SCIENTIFIC OBJECTIVES

This study will aim at establishing:

a) The chemical and micro-structural phase development governing inter-relationships between immobilisation potential (chemical) and physical properties (strength/durability/leachability) of geopolymer systems

b) Effect of amorphous -> crystalline transformation, if occurs, on the immobilisation efficiency and the strength of the immobilised binder

c) Initial mix formulation(s) that gives products with optimum strengths, optimum immobilisation properties and, most importantly, long term stability.

d) Some aspects of the mechanism of immobilisation – whether waste cations/anions will be chemically bound in the matrix replacing alkali, silicon and aluminium ions

DATA, PROGRESS REPORT and RESEARCH OUTCOMES

Compressive Strength

Fig 1. Compressive strength development of with changing elemental ratio
• In general, increasing Si/Al ratios and increasing Si/Na compositions increase the strength of control mixes.
• The best strength results for the controls are with mix 1 & mix 6 – compositions high in Si and low in Al & Na.
• Addition of Cs gives samples with either same or slightly higher strengths than the respective controls. The best compositions for Cs-only mixes are also mix 1 & mix 6.
• Sr behaves differently. Only mix 1 shows comparable (or slightly lower) strengths with the controls. All other Sr-only combinations showed lower strengths than the controls – especially with Mix 6, there is a considerable drop in strength when Sr is added.
• Mix 1 is also the only good composition for combined Cs & Sr mixes.

Compositions high in Si and low in Al & Na produce high strength samples with Cs. Only compositions with high in Si and low in Al is suitable for Sr or Cs+Sr combined samples.

Phase Development – XRD

Fig 2: XRD patterns of mix formulations with changing Si/Al ratio a) Control b) Cs only c) Sr only d) Cs + Sr

• Controls - Mixes with high Si and low Al contents (mix 1) show amorphous patterns (high strength mixes). Mixes with low Si and high Al contents (mix 3) tend to produce more crystalline phases (low strength mixes).
• Cs only - All mixes display amorphous pattern.
• Sr only - Composition high in Si (mix 1) shows an amorphous pattern. But the matrix crystallinity increases with decreasing Si content (mix 3).
• Cs + Sr - Similar to the changes observed with Sr only samples.

High strength results observed for all combinations with mix 1 (Si/Al = 1.9) can be attributed to the amorphous nature of the matrix.
Compositions with high Si and low Na (mix 6, Si/Na = 2.14) favors amorphous structure, while increasing Na content increases the matrix crystallinity. Mix 7 (Si/Na = 1.07) is highly crystalline.

- **Controls** - Compositions with high Si and low Na (mix 6, Si/Na = 2.14) favors amorphous structure, while increasing Na content increases the matrix crystallinity. Mix 7 (Si/Na = 1.07) is highly crystalline.
- **Cs only** - All mixes (including mix 7) display amorphous patterns.
- **Sr only** - All mix compositions seem to be much more crystalline than the controls.
- **Cs + Sr** - Similar to the changes observed with Sr only samples.

Cs seems to favor amorphous nature of the matrix. Sr tend to favor crystalline nature of the matrix. High strength results observed with mix 6 (Si/Na = 2.14) control and Cs samples can be attributed to the amorphous nature of the paste matrix. Samples with crystalline matrices produced low strengths.
Microstructure – Scanning Electron Microscopy (SEM)

Addition of Cs maintains the amorphous nature of the matrix while the addition of Sr enhances the crystalline nature of the matrix.

PCT-B Leach Test

- Changing Si/Al ratio (Fig 5 a) - Decreasing Si content (or increasing Al content) increases leaching of Cs. Mix 1 with high initial Si content gives the lowest Cs concentration (less than 5g/L) in the leachate. However, all three mix compositions (mix 1, 2 & 3) give very low concentrations of Sr (less than 2 g/L) irrespective of the initial Si content present in these samples.
- Changing Si/Na ratio (Fig 5 b) - Changing Si/Na also has an effect on the leaching behaviour of Cs and Sr. Low initial Na content (mix 6 with Si/Na = 2.6) produces leachate solutions with low Cs concentrations. The concentration of Sr is very low in all three mix combinations, irrespective of the initial Na content.

Leaching of Cs is low in mix 1 and mix 6 combinations. With all mix combinations Sr concentration in the leachates are low, irrespective of the initial elemental composition. Low concentrations of Sr in leachates can be due to two reasons.

a. initial mix compositions contained low amount (0.43% or 0.85%) of Sr.

b. Possible precipitation of Sr(OH)₂ in high alkali media used in the mixes.
Fig 5 (a) . Al, Cs, Si and Sr ion concentrations of leachates with Changing Si/Al ratios

Fig 5 (a) . Al, Cs, Si and Sr ion concentrations of leachates with Changing Si/Na ratios
Research outcomes:

1. Cs and Sr, when added as nitrates, act differently in geopolymer formulations.
2. Mix formulations with (high) Si/Al ratio – 1.9 and Si/Na ratio 2.14 give good strength with Cs. Only Si/Al – 1.9 is suitable for Sr or Cs+Sr combinations.
3. Mix formulations with high strengths give less than 5g/L Cs concentrations in leachates. Sr concentrations in the leachates are very low, irrespective of the initial compositions. This could arise from possible precipitation of Sr(OH)₂ in highly alkaline environments.
4. CsNO₃ addition favors amorphous geopolymer formation. Sr(NO₃)₂ addition seems to favor crystalline rather than geopolymer formation. This suggests direct effects of Cs, Sr on the rate/mechanism of geopolymerisation, rather than nitrate.

PUBLICATIONS / REPORTS arising as a result of your work.

Chand H., De Silva P., Vance L, Sagoe-Crenstil K., “The role of Oxide content of geopolymers in Cs and Sr immobilisation” PacRim 9 “The 9th International meeting of Pacific Rim Ceramic Societies, Cairns, July 2011

PhD STUDENTS

Name of the Honours Student - Hasmukh Chand
Thesis Title – “Application of geopolymer binders in Cs and Sr immobilization”
Date of conferment – April 2011