Physical, Chemical, and Structural Evolution of Zeolite-Containing Waste Forms Produced From Metakaolinite and Calcined HLW (EMSP Project ID 65366)

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Background

Natural and synthetic zeolites are extremely versatile materials. They can adsorb a variety of liquids and gasses, and also take part in cation exchange reactions. Zeolites have the ability to sequester ions in lattice positions or within their networks of channels and voids. The zeolites can host alkali, alkaline earth and a variety of higher valance cations. As such they may be a viable alternative for immobilization of low activity waste (LAW) salts and calcines.

The process for synthesizing zeolites is well documented for pure starting materials. A reactive aluminosilicate is reacted with an alkaline hydroxide at low temperature (<300ºC) to form a zeolite. Processing time and temperature and specific reactants (waste) determine the type of zeolite formed. Zeolites are easy to make, and can be synthesized from a wide variety of natural and man made materials. However, relatively little is known about the process if one of the starting materials is a poorly characterized complex mixture of oxides (waste) containing nearly every element in the periodic table.

The purpose of this work is to develop a clearer understanding of the advantages and limitations of producing a zeolite waste form from radioactive waste.

General Zeolization Reaction

Reactive Aluminosilicate + NaOH $\xrightarrow{80\text{-}200^{\circ}C}$ Zeolite

Zeolization Process

- Denitrate waste
- Mix denitrated waste with an aluminosilicate (clay such as metakaolinite)
- Add water to make a paste
- Heat or autoclave (80-200º)

Composition of Actual SRS Tank 44 Decontaminated Supernate to be Converted to a Zeolite Waste Form

Na^+	5.1 M	Cl^-	9.27E-03 M
K^+	0.06 M	F^-	3.38E-04 M
AlO_2^-	0.2 M	PO_4^{3-}	3.12E-03 M
OH^-	4.5 M	SO_4^{2-}	1.33E-03 M
NO_3^-	0.5 M	$^{37}Cs^+$	1.4 µCi/mL
NO_2^-	0.5 M		

Advantages of Zeolite Waste Form

- Zeolites readily form at low temperatures
- High pH wastes well suited to zeolization
- Preliminary tests show that zeolite is a chemically durable waste form

Roles of Principal Investigators

Penn State University

- Scoping studies using simulated waste and a variety of aluminosilicate starting materials
- Develop experimental procedures to be used on actual radioactive waste

Savannah River Technology Center

Use data from Penn State to make radioactive zeolites using real Low Activity Waste (decontaminated High Level Waste)

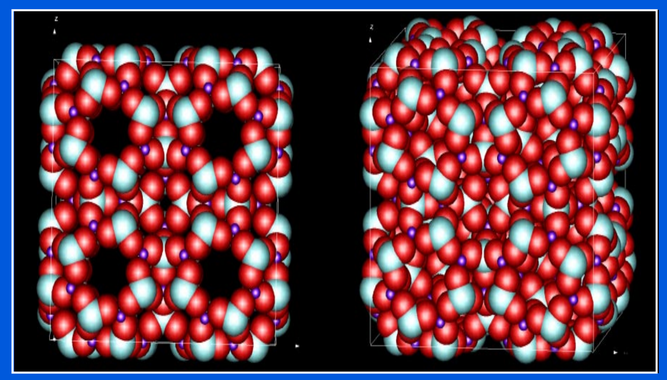
Open Issues

- Radiation stability of hydrated phases
- Large Scale Processing
- Process Control

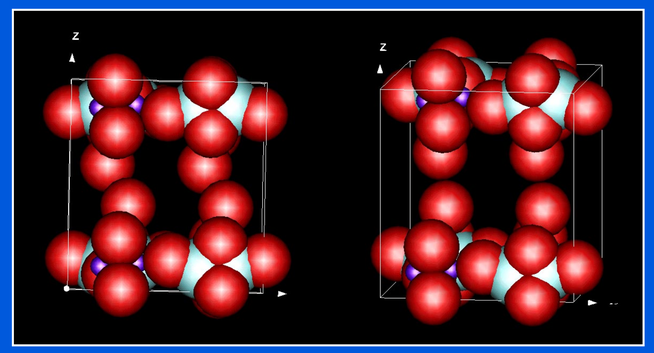
Some Reactants Used to Successfully Fabricate Zeolites

Reactive Aluminosilicate	Sodium Hydroxide
Fiberglass	NaOH Solution (unplanned)
Perflo (perlite) and Diatomaceous Earth	SRS M-Area Waste (Zeolite formation was in-tank and unplanned)
Metakaolinite	Simulated SRS Waste
Fly Ash	Simulated SRS Waste, INEEL Sodium Bearing Waste

Typical Zeolites Formed During Zeolization of Waste



Zeolite Type A



Zeolite Type P1