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PHYSICAL, CHEMICAL, AND STRUCTURAL EVOLUTION OF ZEOLITE-CONTAINING WASTE FORMS PRODUCED FROM METAKAOLINITE AND CALCINED HLW

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RESEARCH OBJECTIVE

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 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. Dr. M. W. Grutzeck at the Pennsylvania State University is investigating the production of a zeolite waste form using non-radioactive simulants. Dr. C. M. Jantzen and J. M. Pareizs at the Savannah River Technology Center will use the results from simulant work as a starting point for producing a zeolite waste form from an actual Savannah River Site radioactive waste stream.

RESEARCH PROGRESS

This past year, work at the Savannah River Technology Center has been focussed on locating and characterizing a small quantity of Savannah River Site waste suitable for zeolitization experiments. The general criteria for the waste was that 1) it had to be sodium hydroxide based, and 2) the activity had to be low enough so the zeolitization process could be carried out in a radio hood (as opposed to a glove box or shielded cell).

Approximately two liters of diluted and decontaminated supernate from SRS Tank 44F that met the above criteria were obtained. The waste was left from SRTC researchers involved in ^{137}Cs removal experiments. The researcher diluted the waste, as received from the SRS Tank Farm, and passed it through an ion exchange column. The ion exchange resin selectively removed only cesium. The supernate was then filtered to remove any solids. The composition of the diluted and decontaminated Tank 44F supernate is given in Table 1.

Because nitrates and nitrites may be detrimental to the zeolitization process, a portion of the diluted and decontaminated supernate will be calcined to remove these anions. A reductant such as sugar may be used during the calcination. The reductant will allow nitrate removal at relatively low temperatures (~500°C), and minimize the loss of any volatile components. The resulting calcine composition is given in Table 2.

Table 1. Composition of Tank 44F Diluted and Decontaminated Supernate

Na^+	5.1 M
K^+	0.06 M
Cs^+	1.4 µCi/mL
OH^-	4.5 M
NO_3^-	0.5 M
NO_2^-	0.5 M
SO_4^{2-}	0.0013 M
AlO_2^-	0.2 M
PO_4^{3-}	0.0031 M
Cl^-	0.0093 M
F^-	0.00034 M
Cr	4.5 mg/mL

Table 2. Diluted and Decontaminated Tank 44F Supernate
Calculated Calcine Composition

	Wt%
Na_2O	91.8
K_2O	1.65
Cs_2O	0.0752
Na_2SO_4	0.110
Al_2O_3	5.94
P_2O_5	0.129
NaCl	0.315
NaF	0.00826
Cr_2O_3	0.00383

Note: One liter of supernate yields approximately 150 grams of calcine.

PLANNED ACTIVITIES

Radioactive supernate (Table 1) and calcine (Table 2) will be used as reactants in the zeolitization process. Processing times, temperatures, and additional reactants will be chosen based on the non-radioactive simulant work at Penn State. Some key goals of the work planned for the coming year are:

- Investigating the correlation between simulants and real radioactive waste in the zeolitization process
- Determining how detrimental nitrates and nitrites are to the zeolitization process
- Characterizing the radioactive zeolite waste form using X-ray diffraction and scanning electron microscopy.