



# High-sodium waste streams stabilized with inorganic acid–base phosphate ceramics fabricated at room temperature<sup>☆</sup>

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Received 9 December 2013; received in revised form 7 March 2014; accepted 7 March 2014

Available online 19 March 2014

## Abstract

A method to stabilize liquid high-sodium waste streams with chemically bonded phosphate ceramics (CBPCs) is presented in this paper. Waste form samples based on CBPC were prepared by mixing a sodium-based waste simulant with calcined magnesium oxide (MgO) and Class C fly ash as filler. The microstructure was identified with X-ray diffraction and electron microscopy. Compressive strength was evaluated for the compositions in which a solid material was obtained. Of the 15 different simulated waste form compositions fabricated, only a few of them did not set into a ceramic material. Immersion tests were conducted over selected compositions. All phases were identified in the solid materials. Results showed that a maximum concentration of 60 wt% NaH<sub>2</sub>PO<sub>4</sub> was attained in the set product showing that CBPC is a promising material for encapsulation of high-sodium nuclear waste.

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**Keywords:** Ceramic matrix composites; Chemically bonded phosphate ceramics; High-sodium waste streams

## 1. Introduction

Typically, sodium wastes are the product of nuclear site operations such as decontamination activities, some of which use dilute sodium hydroxide to wash surfaces and solubilize residues. As a result, significant sodium-based salts are present in the liquid wastes. The high sodium content in these solutions makes them unsuitable for direct calcination as the sodium nitrate melts at low temperatures and does not produce

a granular, free-flowing calcination product. Sodium waste in the nuclear industry is closely associated with the development of liquid metal fast reactors [1], which are an advanced type of nuclear reactor where the primary coolant is liquid sodium [2]. These reactors have received considerable attention due to safety concerns regarding the possibility of an accident in the containment building leading to a sodium fire that could potentially release radioactivity to the environment [3].

Since these high-sodium salt wastes are very sensitive to elevated temperatures, an ideal solution is to stabilize them at room temperature in solid materials like ceramics. It is well known that ceramics usually require high temperature processing like sintering. These processes are not desirable because they involve high energy consumption and are not cost-effective for large-scale manufacturing. In addition, the use of coal as a fuel in the traditional processing also leads to an increase in emitted gases and particles that contribute to global warming. These conditions lessen the advantages of nuclear power energy production over conventional fossil fuels. Several solutions have been proposed to stabilize

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sodium-based nuclear waste streams involving high temperature methods [4], which can involve the release of radioactive and toxic waste.

Chemically bonded phosphate ceramics offer a solution to synthesize ceramics at room temperature. With this waste form, bonding takes place in a chemical reaction at low temperatures by acid–base reactions as opposed to the thermal diffusion or sintering at high temperatures ( $> 1000\text{ }^{\circ}\text{C}$ ) used for traditional ceramics and cements [1–11]. In traditional cement hydration products, van der Waals and hydrogen bonding dominate. In contrast, CBPCs form by acid–base reactions between an acid phosphate and a metal oxide (such as that of magnesium, calcium, or zinc) and involve a mixture of ionic, covalent, and van der Waals bonding, with the ionic and covalent bonding dominating.

On the other hand, sodium phosphates have been used as a host structure to stabilize nuclear waste streams, such as sodium zirconium phosphate [12] stabilizing the waste streams from a fast breeder reactor [13]. Other materials and technologies such as titanite ceramics [3] and glasses [14] have also been used for the immobilization of sodium-bearing nuclear waste. In general, the main disadvantage with respect to the CBPC method is the processing temperature, which not only increases the cost and complexity of the process, but also restricts the use when hazardous volatiles are produced.

In a waste reprocessing plant, low level liquid waste is concentrated, and subsequently, solidified using a low-temperature process such as cementation or bituminization. The remaining solvent waste is treated with an acid digestion process that includes phosphoric acid. The resulting waste is an effluent

that is concentrated with  $\text{NaH}_2\text{PO}_4$  (specifically sodium ions reacting with dihydrogen phosphate anion  $\text{H}_2\text{PO}_4^-$ ) that is difficult to solidify and contain using standard low temperature waste form technologies. This paper addresses the feasibility of using CBPCs for solidifying and containing concentrated  $\text{NaH}_2\text{PO}_4$  solutions.

For this study, we prepared simulated CBPC samples by mixing liquid simulants of high-sodium waste streams. These simulants were fabricated with different concentrations of anhydrous monobasic sodium phosphate ( $\text{NaH}_2\text{PO}_4$ ), water, calcined magnesium oxide ( $\text{MgO}$ ), and fly ash type C. A maximum concentration of 60% of  $\text{NaH}_2\text{PO}_4$  was consolidated into a solid ceramic material. This demonstration of encapsulating a high concentration of sodium-based liquid solution in a ceramic material fabricated at room temperature indicates that this method can be used at a large scale for stabilizing nuclear sodium-base waste streams.

## 2. Experimental

Sodium waste stream simulants with different sodium phosphate contents were fabricated by mixing anhydrous monobasic sodium phosphate ( $\text{NaH}_2\text{PO}_4$ ) from Fisher Scientific and water. Scanning electron microscope (SEM) images for the  $\text{NaH}_2\text{PO}_4$  are shown in Fig. 1a. Table 1 summarizes the different  $\text{NaH}_2\text{PO}_4$  loadings used in this research. For loading  $< 50\text{ wt}\%$   $\text{NaH}_2\text{PO}_4$ , the solution was mixed at room temperature ( $25\text{ }^{\circ}\text{C}$ ). For loadings  $\geq 50\text{ wt}\%$   $\text{NaH}_2\text{PO}_4$ , the solution was heated up to  $60\text{ }^{\circ}\text{C}$  while being continuously stirred with a magnetic stirrer to completely dissolve the

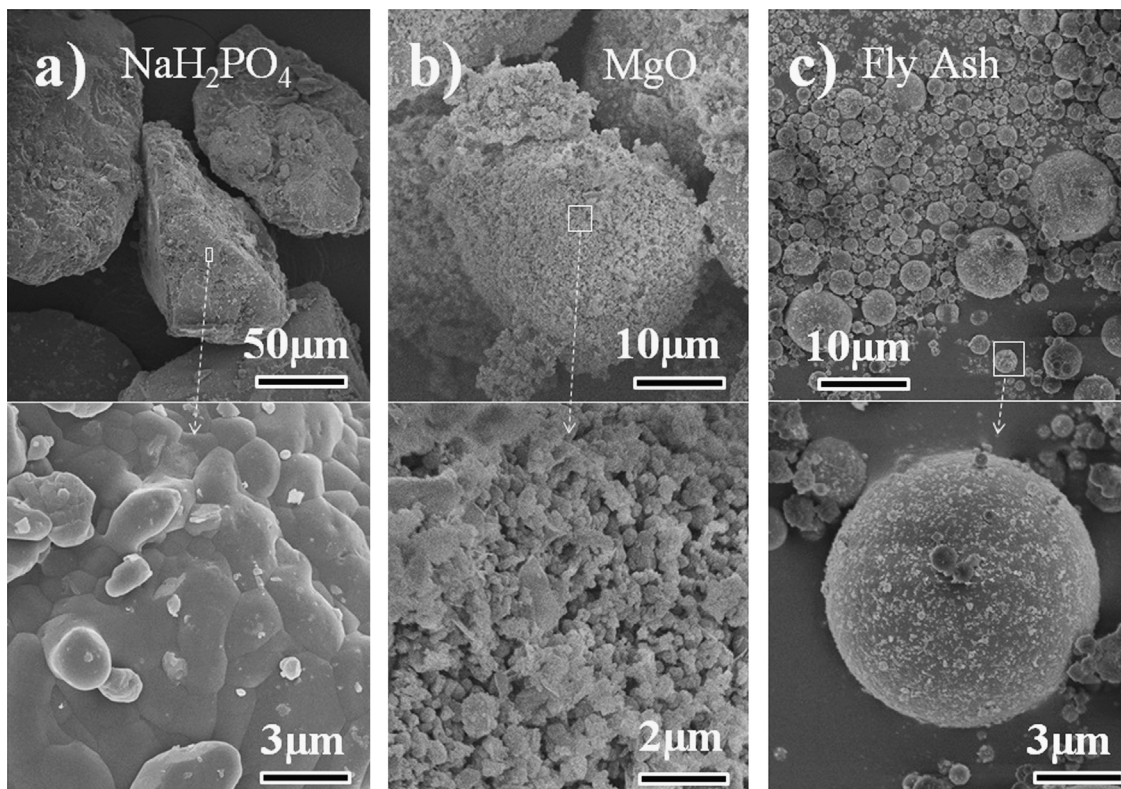


Fig. 1. Raw materials used in the fabrication of the high-sodium waste streams.

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