

## Durability Test of Cold-Immobilized Binder-Free Adsorbents

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### 1. Introduction

Some radioactive nuclides such as Cs<sup>+</sup>, Co<sup>2+</sup> produced as a result of the operation of nuclear power plants have to be removed and stored [1]. For removing the nuclides in the effluent and liquid wastes, both ion exchange and adsorption technologies are used because of easy and economical application. After capturing them, the as-spent materials are mixed with glass materials as binders and fixed through the vitrification, which is performed at high temperature (>1,000 °C), and there is a disadvantage of volatilization of low boiling point elements [2]. Thus, replacing glass binders for vitrification is a more safe and economical way. Recently, the low-temperature consolidation technology called cold-sintering is developed for making spent adsorbents into the waste matrix without binders [3].

The main element of the Allophanes is amorphous aluminosilicate, which can adsorb the bulky cations which microporous material could not capture [4]. Moreover, it is an easy and inexpensive to synthesize amorphous aluminosilicate powder. Thus, we used the material for adsorption and analyzed the durability of the immobilized binder-free matrix.

### 2. Methods/Experimental

#### 2.1. Forming As-Spent Matrix

Amorphous aluminosilicate was synthesized following the co-precipitation method [4]. Synthesized amorphous aluminosilicate powder used for batch adsorption tests with nuclides (Cs<sup>+</sup>, Co<sup>2+</sup>, Ni<sup>2+</sup>) followed by OECD guideline. After finishing the adsorption test, the batch was divided into liquid and as-spent adsorbent through a centrifugation process and dried at 90 °C for 24 h. The dried powder was pressed at a low temperature (200 °C) within 10 min.

#### 2.2. Characterization

The scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDS, TEAM<sup>TM</sup>) were used to investigate sorption sites on the adsorbents after the adsorption test and surface integrity after the long term leaching test.

The chemical durability of cold sintered amorphous aluminosilicates adsorbing nuclides was investigated under static leaching conditions. The 7-Days product consistency test (7-Days PCT) as per the ASTM

standard (C1285) was performed using MQ water as the leaching medium[6]. The 90-days leaching test (ANSI/ANS16.1) using deionized water was carried out[7].

The concentrations of ions in the solution were measured by the inductively coupled plasma optical emission spectroscopy (Agilent ICP-OES 720).

### 3. Results and Discussion

#### 3.1. Synthesized Amorphous Aluminosilicate

The synthesized aluminosilicate is an amorphous phase confirmed by XRD [5]. The appearance of cold sintered material is shown in Figure 1. It was well consolidated and structural integrity was retained when it submerged underwater.

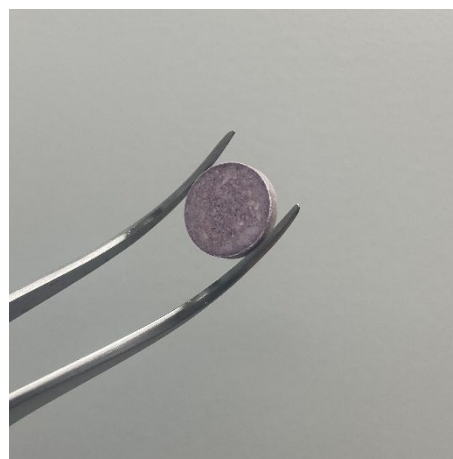


Fig. 1 Cold-sintered as-spent amorphous aluminosilicates.

#### 3.2. 7-Days Product Consistency Test (7-Days PCT)

For measuring the chemical stability and leaching-resistance of the formed waste matrix, ASTM C1285-14 product consistency test was done. The normalized leaching rate was calculated by equation 1.

$$NLR_i = \frac{C_i}{t \times f_i \times S/V} \quad (1)$$

where C<sub>i</sub> [g/L] is the concentration of element i in the leachate after a certain leaching time t (days). f<sub>i</sub> is the weight fraction of element i in the green sample, and S/V

( $m^2/L$ ) is the surface area of the sintered waste matrix divided by the leachate original volume.

The calculated normalized leaching rate with nuclides exhibited that amorphous aluminosilicate possessed 100-10,000 times higher durability.

### 3.3. ANSI/ANS 16.1 Leaching Test

The test is a simple and economical method for the quantitative comparison of the resistance of different radionuclides. The leaching indices and diffusion coefficient were calculated by the theoretical formula in ANSI/ANS-16.1-2019 document [7].

The calculated value showed higher durability than Portland cement type and zeolite. The leaching index fulfilled the threshold value (i.e. 6) for the acceptance criteria of a given matrix as radioactive waste immobilization.

The higher chemical durability was confirmed from the microstructure also. The post-leaching test surface of the cold-sintered matrix did not have any cracks or voids.



**Fig. 2** Post-leaching test of cold-sintered amorphous aluminosilicate with cesium

## 4. Conclusions

The study demonstrated that the chemical durability of easily and economically synthesized amorphous aluminosilicate. It showed higher durability than other adsorbents with a glass binder, without requiring high processing temperature over 1,000 °C temperature. Thus, this material can be used for not only adsorbents but also binders because it can be sintered at low temperatures.

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