



Stable solidification of cesium with an allophane additive by a pressing/sintering method



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HIGHLIGHTS

- A pressing/sintering method was used to solidify AMP-Cs/SiO₂-allophane.
- The volatility of Cs₂O could be effectively restrained by allophane.
- Cs₄Al₄Si₂₀O₄₈ was formed for stable solidification of Cs.
- The leachability of Cs from sintered AMP-Cs/SiO₂-allophane was relatively low.

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ABSTRACT

Pyrolysis of AMP/SiO₂ adsorbed Cs (AMP-Cs/SiO₂) occurred at > 400 °C sintering temperature, and Cs immobilisation decreased from 100% to 40% after sintering at 1200 °C. To safely dispose radioactive Cs, allophane was immobilized with AMP-Cs/SiO₂ to prepare a stable form by using a pressing/sintering method. The structure of AMP-Cs/SiO₂ collapsed, and cesium aluminosilicate formed more easily under conditions of higher sintering temperature (>800 °C) or increasing mixing ratio of allophane (mass ratio = 1:3 AMP-Cs/SiO₂-allophane). The decomposition products of AMP-Cs/SiO₂ were Cs₂O, MoO₃ and P₂O₅ at 1200 °C. Cs₂O volatilisation was depressed by allophane addition, and a stable immobilisation phase of Cs₄Al₄Si₂₀O₄₈ formed. An immobilisation ratio of Cs of approximately 100% was maintained. The leachability of Cs for AMP-Cs/SiO₂-allophane (1:3, 1200 °C) in distilled water at 25 °C and 90 °C for 15 days was estimated as 0.174% and 1.55%, respectively.

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1. Introduction

In the Fukushima nuclear accident, large amounts of radioactive waste water were generated. The management and treatment of this waste water is an important issue in environmental safety. ¹³⁷Cs is one of the principal heat-generating isotopes in radioactive waste water, exhibiting strong beta, gamma-ray, high inherent solubility and leachability [1–3]. Numerous adsorbents are developed to separate ¹³⁷Cs from Fukushima radioactive waste water, including natural and synthetic zeolites with various cation types [4], conjugate adsorbent prepared by direct immobilisation of dibenzo-18-crown-6 ether onto mesoporous silica monoliths [5], insoluble ferrocyanide [6], silica based ammonium

molybdophosphate [7] and so on. However, large amounts of secondary solid wastes (used adsorbents) after removal of ¹³⁷Cs are accumulated and urgently require management. After volume reduction and valuable component recovery, the residue waste materials should be conditioned by solidification, storage and disposal [8–10]. The goal of solidification is to convert the wastes into forms with excellent physical and chemical stability for handing, transport and disposal. In addition, the release of radioactive nuclides from the forms of wastes will be slow even upon contact with complicated ground water. Numerous methods for the solidification of Cs such as glass solidification, cement solidification, ceramic solidification, and rock solidification cementation have been reported [11–14]. However, the above methods present several disadvantages, such as easy devitrification, low thermodynamic stability and high leaching rate [15–17]. In view of the above factors, aluminosilicate solidified bodies are easy to synthesize and present good chemical stability, thermal stability, radiation and

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leachability resistance [18–20].

Recent years, silica based adsorbents had attracted more and more attention because of excellent mechanical and chemical stability and low column pressure loss, such as silica based prussian blue analogues [21,22] and silica based ammonium molybdophosphate (AMP/SiO₂) [7,23]. Silica based Prussian Blue Analogues showed excellent affinities for removal of Cs from wastewater. For solidification of these type adsorbents, it should be concerned with the treatment of HCN gas during the solidification process [18,24]. Silica based ammonium molybdophosphate was successfully applied to remove Cs from radioactive waste water in our previous study. To solve the hydraulic problems of AMP, SiO₂ was used as support to load AMP into the pores. AMP/SiO₂ shows superior adsorption properties towards Cs than zeolites, including fast uptake rate, and high saturated adsorption capacity, resulting in high adsorption efficiency of Cs and reduction of the amount of secondary wastes. As a potential inorganic adsorbent for removing Cs from radioactive waste water, AMP was widely investigated [25–27], but final disposal, such as solidification and volume reduction is rarely reported. In addition, SiO₂ has been widely used as the support, and the solidification of SiO₂ has attracted increasing attention. Therefore, the solidification of AMP/SiO₂ will provide strong technical support for its practical application. Allophane, a type of hydrous aluminium silicate with amorphous structure, shows fine Cs adsorbability, gas trapping and self-sintering features. Therefore, a solidified substrate is expected to achieve the stable solidification of Cs.

For the stable solidification of AMP/SiO₂ absorbed Cs (AMP-Cs/SiO₂), allophane is selected as fixation material which possesses the abilities of self-sintering and Cs trapping to restrain the volatilisation and achieve the immobilisation of Cs. The present study deals with the preparation and characterisation of the sintered solidified products and estimation of the stability, mechanism, optimised solidification condition and Cs leachability.

2. Experimental

2.1. Materials and preparation procedure

AMP/SiO₂ adsorbent was prepared by successive impregnation of molybdophosphoric acid and ammonium chloride into the macropores of SiO₂ to form ammonium molybdophosphate precipitation [7,21]. AMP/SiO₂ was then treated with 0.2 M CsNO₃ solution to obtain AMP-Cs/SiO₂. Allophane, with a porous and hollow structure, was purchased from Hattori company, Ltd. Japan. The diameter of allophane particle is ranged from 3.5 to 5.0 nm and the chemical formula is shown as 1~2SiO₂·Al₂O₃·5~6H₂O. The main components of allophane used as matrices are analysed by X-ray fluorescence (XRF) (Table 1). The mixtures of AMP-Cs/SiO₂-allophane before sintering at different mass ratios (mass ratios of AMP-Cs/SiO₂ and allophane = 1:1, 1:2 and 1:3) are also presented in Table 1.

Table 1
Main components of allophane and mixtures of AMP-Cs/SiO₂-allophane.

Components	SiO ₂	Fe ₂ O ₃	K ₂ O	Al ₂ O ₃	CaO	Na ₂ O
Allophane	78.78%	3.51%	1.64%	12.39%	2.38%	0.48%
Mixtures	SiO ₂	Cs ₂ O	P ₂ O ₅	MoO ₃	Al ₂ O ₃	Fe ₂ O ₃
1:1	32.16%	1.78%	0.40%	48.53%	15.19%	1.49%
1:2	39.82%	1.63%	1.01%	27.56%	15.19%	1.49%
1:3	42.21%	1.24%	1.02%	21.00%	30.79%	2.64%

2.2. Solidification and Cs immobilisation

The AMP/SiO₂ was treated with CsNO₃, and the resulting AMP-Cs/SiO₂ was sintered up to 1200 °C for 1 h, the immobilisation ratio (%) of Cs was determined by the difference of Cs content before and after sintering. For stable solidification of Cs, allophane was selected as the fixation material. The solidification process was as follows: the mixtures of AMP-Cs/SiO₂-allophane (1:1, 1:2 and 1:3) were pulverised and then moulded as a disc by cold-pressing (40 MPa). The moulded discs were sintered at different temperature (400–1200 °C) for 1 h. The flowsheet for the preparation of solidified products is shown in Fig. 1.

2.3. Characterisation

The surface morphologies of fresh adsorbents and sintered specimens were examined by scanning electron microscopy (SEM, NovaSEM NPE218, FEI Company, USA). The elemental distribution was tested by energy dispersive spectrometry (EDS, NovaSEM NPE218, FEI Company, USA) and chemical composition was determined using a Sequential XRF Spectrometer (XRF, XRF-1800, Daojin Company, Japan). The crystal structure of the specimens before and after sintering was identified by X-ray powder diffraction spectroscopy. (XRD, D8 ADVANCED DA VINCI, Germany).

2.4. Leaching test

For estimating the leachability resistance of the solidified products (10 mm Ø disc, sintered products of AMP-Cs/SiO₂-allophane (1:1, 1:2 and 1:3) at 1200 °C), the solidified products were contacted with different leachants in glass bottles stored at 25 °C or 90 °C for 15 days. After 15d leaching, the leachant was filtered and the Cs concentration in the leachant was determined by an atomic absorption spectrophotometer (AAS, Shanghai Guangpu-SP3880). The solubility of Cs for sintered products in the leaching test was calculated from the specific values of the Cs content in the leachant and the Cs content in the solidified products without the leaching test. The parameters for the leaching test are listed in Table 2.

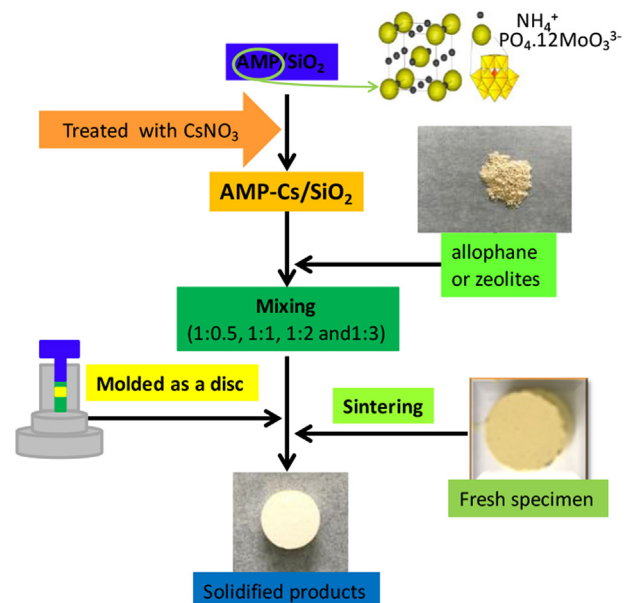


Fig. 1. Flowsheet for the preparation of solidified products.

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