Optimization of CHA-PCFC Hybrid Material for the Removal of Radioactive Cs from Waste Seawater

Keun-Young Lee^{*}, Jimin Kim, Minsung Park, Kwang-Wook Kim, Eil-Hee Lee, Dong-Yong Chung, Jei-Kwon Moon Korea Atomic Energy Research Institute (KAERI), 989-111 Daedeok-daero, Yuseong-gu, Daejeon, 305-353, Korea ^{*}Corresponding author: lky@kaeri.re.kr

1. Introduction

Nuclear power plant accident occurred in Fukushima Daiichi has shown that the technologies for removing the hazardous radionuclides, such as Cs, Sr, I etc., from waste seawater are very important in case of emergency. Even though the liquid waste treatment processes in the normal operation of nuclear power plant are commercialized, those in the abnormal accidents have not been fully developed until now. In the present study, as a preliminary research for the development of precipitation-based treatment process specialized for the removal of Cs from waste seawater generated in the emergency case, the performance test of a hybrid material combining chabazite and potassium cobalt ferrocyanide was conducted. Also the synthesis method for the hybrid adsorbent was optimized for the best Cs removal efficiency on the actual contamination level of waste seawater.

2. Experimental section

Three types of synthesized adsorbents, such as chabazite(CHA), potassium cobalt ferrocyanide(PCFC), ferrocyanide-embedded and potassium cobalt chabazite(CHA-PCFC), were used for Cs adsorption experiments using simulated waste seawater contaminated by inactive Cs with Cs-137 as tracer level, and the reaction kinetics and adsorption capacities were investigated depending on the various synthesis and adsorption conditions. The simulated waste seawater was prepared by dissolving inactive Cs as a concentration of 1 mg/l and Cs-137 as an activity of 3.7×10^3 Bq/ml into the seawater collected from Korean East Sea. The activities of Cs-137 before and after the adsorption experiments were analyzed by MCA (Multi Channel Analyzer, TC702, Oxford, UK). In order to optimize the synthesis method of CHA-PCFC hybrid material, Cs removal efficiencies of CHA, PCFC, and CHA-PCFC synthesized with different concentrations, temperatures, washing and drying conditions.

Also, various physicochemical and mineralogical properties of the materials before and after the experiments were observed by PSA (3500S, Microtrac, USA), DLS (ZS90, Malvern, UK), SEM (SNE 4500M, SEC, Korea), EDS (QUANTAX, Bruker, Germany), TEM (2100 FXII, JEOL, Japan) and XRD (D2 Phaser, Bruker, Germany), which made it possible to explain the Cs removal properties of the materials.

Table I: Mean	particle sizes	of synthesi	zed adsorbents	
measured by different methods.				

	$LD^{1)}$	DLS ²⁾
CHA	13.4 µm	
PCFC		635 nm
CHA-	20.7. um	
PCFC	20.7 µili	

¹⁾Mean volume diameter of dried sample by LD (Laser Diffraction)

²⁾Mean volume diameter of colloidal sample by DLS (Dynamic Light Scattering)

3. Results and discussion

3.1 Synthesis results of hybrid adsorbent

The synthesized chabazite was observed as aggregated particles with relatively smooth surface of the chabazite pure crystals and the synthesized potassium cobalt ferrocyanide was observed as aggregated forms with nano-sized crystals in the TEM image (Fig. 1A and B). The potassium cobalt ferrocyanide-embedded chabazite showed more complex aggregates with a number of potassium cobalt ferrocyanide nano clusters on the surface of chabazite (Fig. 1C and D).



Fig. 1. TEM images of synthesized (A) CHA, (B) PCFC, and (C, D) CHA-PCFC.

Elemental compositions measured by EDS directly supported that the synthesized material was the combination of chabazite as a micro-scale support and potassium cobalt ferrocyanide as a nano-scale cluster (Fig. 2).



Fig. 2. EDS results of synthesized (A) CHA, (B) PCFC, and (C) CHA-PCFC.

3.2 Cs removal efficiency

In order to define the optimized synthesis conditions specialized for the best Cs removal and its application to the precipitation-based process, some comparative experiments were performed.

Fig 3 showed variations of distribution coefficients (K_d) of Cs with time by the different amounts of CHA-PCFC per unit volume. All the cases of m/V showed more than 10^4 of distribution coefficient, therefore the synthesized material could be regarded as an effective adsorbent for the removal of an extremely low concentration of Cs in the waste seawater. Fig. 4 represented changes of Cs removal efficiencies with time by CHA-PCFC synthesized with different temperatures. Because the temperature effect on the synthesis of PCFC was confirmed by preliminary experiments, the optimization of CHA-PCFC synthesis was also conducted. The hybrid material synthesized at 40°C showed the highest distribution coefficient of Cs in the same manner of the performance of PCFC synthesized at the lower temperature than that of conventional methods.



Fig. 3. Distribution coefficient (K_d) of Cs by CHA-PCFC with time and different m/V in seawater containing Cs 1ppm with Cs-137(tracer).



Fig. 4. Distribution coefficient (K_d) of Cs by CHA-PCFC synthesized with different temperatures in seawater containing Cs 1ppm with Cs-137(tracer).

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