

**A New Adsorbent for Simultaneous Removal of Cesium and Strontium – 14110**

Yuko Kani \*, Takashi Asano \*\*, Shin Tamata \*\*

\* Hitachi, Ltd.

\*\* Hitachi-GE Nuclear Energy, Ltd.

**ABSTRACT**

Contaminated water accumulated at the Fukushima Daiichi Nuclear Power Plant accident site contains many kinds of radionuclides. Radioactive cesium (Cs) and strontium (Sr) are included in the waste water at especially high concentrations ( $10^4$  to  $10^5$  Bq/cm<sup>3</sup>). For treatment of this waste water, we propose a new concept for adsorbents that remove Cs and Sr simultaneously. The developed adsorbent, a specially-treated titanate compound, removes Cs and Sr simultaneously and independently with high capacity and selectivity. The distribution coefficients (K<sub>d</sub>) of the developed adsorbent for Cs and Sr measured under the simulated sea water conditions are found to be similar to those of ferrocyanide (K<sub>d</sub>> $10^4$  L/kg) and sodium titanate (K<sub>d</sub>> $10^3$  L/kg), respectively. This result indicates that the developed adsorbent has high adsorption capacity for both Cs and Sr. The column testing for the adsorbent is also carried out to determine the dynamic adsorption capacity for Cs and Sr. The dynamic Cs adsorption capacity is about 28 times higher than that for Sr.

**INTRODUCTION**

Contaminated water accumulated at the Fukushima Daiichi Nuclear Power Plant accident site contains many kinds of radionuclides. Radioactive cesium (Cs) and strontium (Sr) are included in the waste water at especially high concentrations ( $10^4$  to  $10^5$  Bq/cm<sup>3</sup>).

The currently operated waste water treatment system at the Fukushima site has two main processes: the adsorption process for removal of Cs and the reverse-osmosis (RO) process for desalination. Most of the Cs is collected in the adsorbent medium and Sr and other radionuclides including residual Cs are in the RO concentrate (RO brine). A multiple nuclide removal system that removes all radionuclides from the RO brine stored in tanks has been constructed and two additional system are planned for risk reduction of radionuclide leakage.

Since Cs, an alkali metal element, and Sr, an alkaline earth, belong to different groups of the periodic table, they have been removed using separate adsorbent media, for example, natural zeolite and ferrocyanate for Cs, and sodium titanate and synthesized zeolite for Sr.

In this paper, we propose a new concept for adsorbents that remove Cs and Sr simultaneously. The adsorption ability of the new adsorbent is studied by batch and column tests using simulated waste water. Simultaneous removal of Cs and Sr promises a reduction in the number of processes for radionuclide removal in the waste water treatment system.

**METHODS**

The newly developed adsorbent is made of a kind of crystalline silicotitanate (CST). Pelletized

CST, for example, UOP Ionsiv™ R9120 (IE-911), was immersed in dilute NaOH solution for about 1 hour, washed with a small amount of deionized water, and air-dried. Figure 1 shows a photograph of the developed adsorbent.

Adsorption of radioactive Cs and Sr to the newly developed adsorbent was investigated by batch and column tests. Test feed solutions were prepared by appropriately diluting the simulated sea water (Marine Art SF-1) and adding radionuclide solutions containing Cs-137 and Sr-85 for the batch and column tests.

In the batch test, the distribution coefficient ( $K_d$ ) was obtained by a laboratory immersion test using the simulated sea water (no dilution). The adsorbent of known mass ( $M_{ad}$ ) was put into a plastic bottle and a test feed solution with known volume ( $V_{sol}$ ) and concentration of radionuclide ( $C_0$ ) was added. The  $V_{sol}/M_{ad}$  ratio was selected as 100:1 (v/w). The bottle was capped and kept on a rotary shaker at room temperature for 24 h. A portion of the solution was filtered using a 0.45  $\mu$  filter and the concentration of radionuclide ( $C_i$ ) was measured. The  $K_d$  was calculated by Eq.1.



Fig.1. Photograph of the developed adsorbent

$$K_d = \frac{V_{sol}(C_0 - C_i)}{M_{ad}C_i} \quad (\text{Eq.1})$$

The column test was performed using a plastic column (10 mm ID) filled with 5 mL of the adsorbent. Examination of the breakthrough curve was carried out with test feed solutions of different brine concentrations (TABLE I). The flow rate of the solution was kept at 75 mL/h (15 bed volume/h). At selected time intervals the concentrations of Cs-137 and Sr-85 in effluent were measured with a Ge detector.

TABLE I. Concentration of ions in the test feed solutions used in the column testing (ppm)

No.	Cl <sup>-</sup>	Na <sup>+</sup>	Sr <sup>2+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>
1	6000	5000	2.4	136	395
2	3200	1777	1.3	73	210
3	2000	1111	0.8	45	132
4	1000	555	0.4	23	66

## RESULTS AND DISCUSSION

### Distribution coefficient ( $K_d$ )

Figure 2 shows the distribution coefficients ( $K_d$ s) of various adsorbents for Cs and Sr obtained for the non-diluted simulated sea water. The  $K_d$  data of the developed adsorbent and CST plotted in Fig.2 were obtained in this study, and those of other adsorbents have been reported

elsewhere [1]. Natural zeolite, ferrocyanate and CST are known as Cs adsorbents, and synthesized zeolites and titanate as Sr adsorbents. Our developed adsorbent has the  $K_d$  as large as those of CST and ferrocyanate for Cs, and as large as those of titanate and synthesized zeolite for Sr. The developed adsorbent, a specially-treated CST-like compound, has a high Sr adsorption ability with the same Cs adsorption ability as CST. This means that the developed adsorbent is capable of adsorbing Cs and Sr simultaneously.

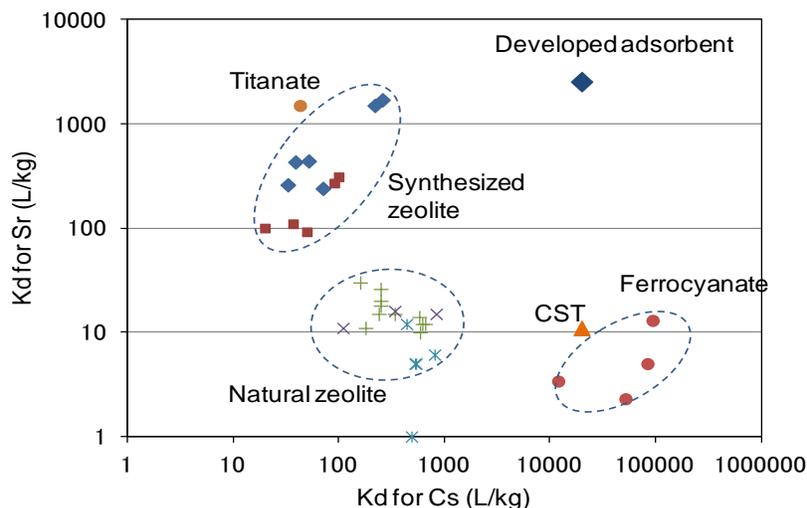


Fig.2. Distribution coefficients ( $K_d$ s) of the various adsorbents

### Breakthrough curves for Sr

Figure 3 shows the breakthrough curves of the developed adsorbent for Sr obtained with various test feed solution conditions. Breakthrough of the developed adsorbent has a clear dependency on the test feed solution condition: the test feed solution No.1 with the highest concentration of Sr exhibits the fastest breakthrough among the test feed solutions examined.

Table II summarizes the breakthrough point and the dynamic Sr adsorption capacity at  $C/C_0=0.1$  for the test feed solution conditions. The dynamic Sr adsorption capacity is defined as the amount of Sr (mg) adsorbed per 1 g of the developed adsorbent from throughput=0 to breakthrough point at  $C/C_0=0.1$ . The higher the Cl and Sr concentrations in the test feed solution are, the shorter the breakthrough and the lower the Sr adsorption capacity that are obtained. The test feed solution of higher Cl concentration contains higher concentrations of Ca and Mg, the interfering elements for Sr adsorption, and the dependency of dynamic Sr adsorption capacity of the developed adsorbent on the Cl concentration of the test feed solution suggests that Sr adsorption onto the developed adsorbent is somewhat affected by a competitive reaction with Ca and Mg.

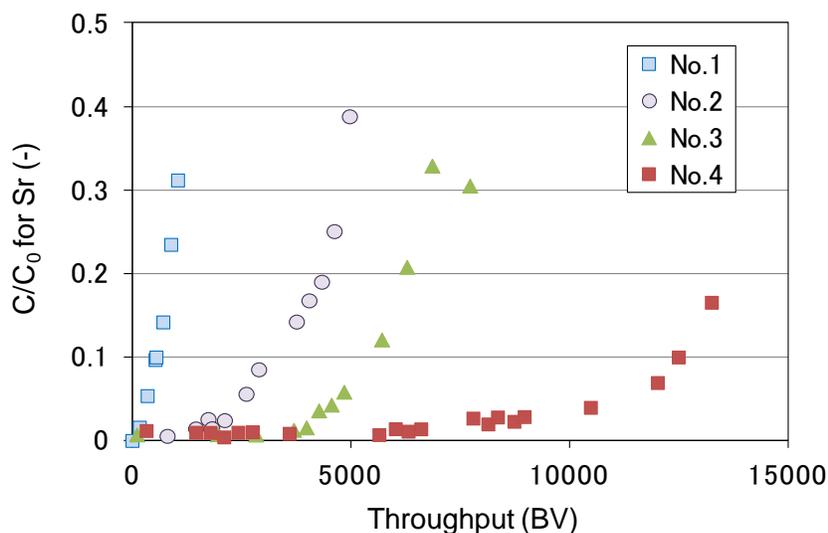


Fig.3. Breakthrough curves for Sr obtained with various test feed solution conditions

Table II. Breakthrough and dynamic Sr adsorption capacity at  $C/C_0=0.1$  for various test feed solution conditions.

No.	Cl concentration (ppm)	Sr concentration (ppm)	Breakthrough (BV)	Dynamic Sr adsorption capacity (mg-Sr/g-media)
1	6000	2.4	550	1.6
2	3200	1.3	3200	4.9
3	2000	0.8	5300	5.2
4	1000	0.4	12500	6.1

**Breakthrough curves for Cs**

Figure 4 shows the breakthrough curve of the developed adsorbent for Cs obtained with the test feed solution No.1 but with 10 ppm of cold Cs added. Calculated dynamic Cs adsorption capacity at  $C/C_0=0.1$  for No.1 test feed solution is 44 mg-Cs/g-media, about 28 times higher than the Sr adsorption capacity for the same test feed solution condition. The developed adsorbent has much higher adsorption ability for Cs than for Sr.

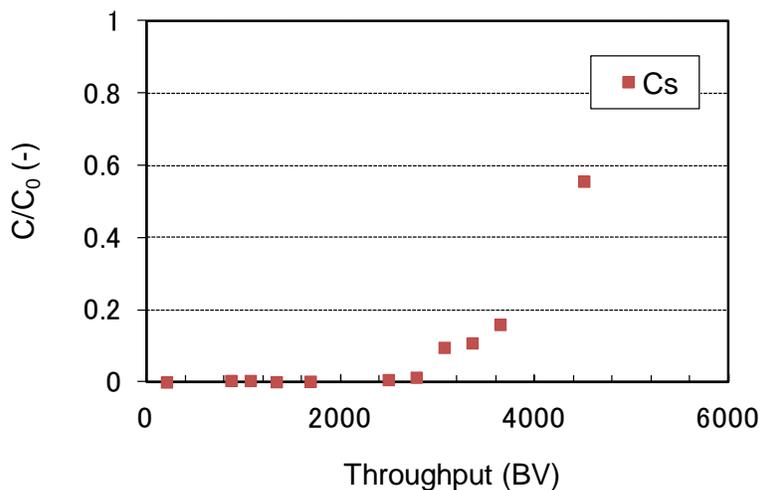


Fig.4. Breakthrough curve for Cs obtained with the test feed solution No.1 (Cs=10ppm)

**Breakthrough curves for Cs and Sr**

Figure 5 shows the breakthrough curves of the developed adsorbent for Cs and Sr obtained with the test feed solution No.1. The feed solutions for tests A and B contain 0.03 ppm and 0.6 ppm of cold Cs, respectively. As shown in Fig.5, the breakthrough curves for Sr of tests A and B are almost the same although the test feed solutions used in tests A and B contain different concentrations of Cs. The result indicates that Cs and Sr are adsorbed onto the developed adsorbent independently.

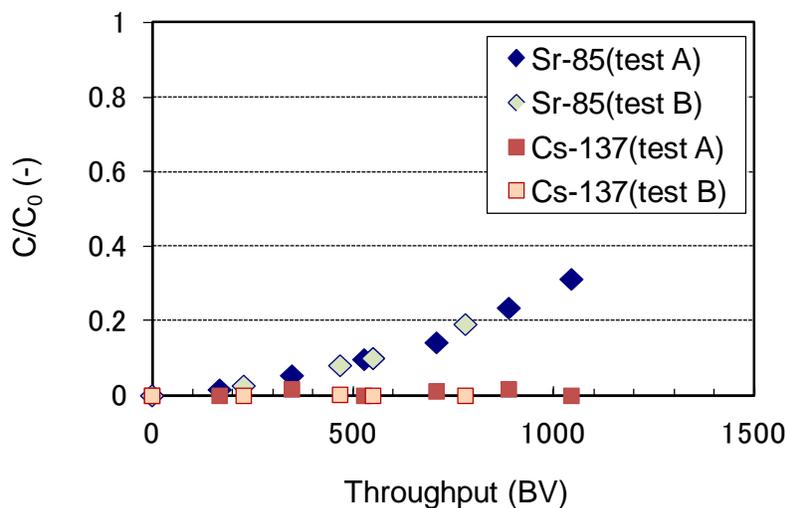


Fig.5. Breakthrough curves for Cs and Sr obtained with the test feed solution No.1 (test A, 0.03ppmCs; test B, 0.6ppmCs)

## **CONCLUSIONS**

A new adsorbent that adsorbs Cs and Sr simultaneously and independently has been developed for the treatment of waste water generated at the Fukushima Daiichi Nuclear Power Plant site. The batch and column tests for the developed adsorbent showed its high adsorption ability and capacity for both Cs and Sr even under sea water conditions containing interfering elements such as Na, Ca and Mg. The adsorbent is promising to remove Cs and Sr from the RO concentrate stored in tanks at the plant site.

## **REFERENCE**

1. Atomic Energy Society of Japan, Division of Nuclear Fuel Cycle and Environment, "Contaminated Liquid Water Treatment for Fukushima Daiichi NPS", <http://www.nuce-aesj.org/projects/clwt:start>