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Removal of Radioactive Cobalt(II) Ions in High Temperature•Pressure Chemical & Volume Control System (CVCS) by Ceramic Nanoparticles Adsorbents

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1. Introduction

Recently, one of the biggest issues on nuclear industry was waste treatment and purification. Several accidents such as Fukushima provoked public concerns even the actual damage directly from radioactivity disposal was small.

A few types of waste produced during the nuclear operation are treated in several ways. Large particulates are filtered out, and gaseous waste are ventilated and adsorbed. Liquid waste involving the radioactive ions are the major concern, which are decontaminated with ion exchange resin bed. In the ion exchange resin bed, bulk of small ion exchange resin are embedded, and the letdown flows through the bed where radioactive ions are adsorbed to the resin beads. This conventional way have been widely used, but still have big limitation on the stability of the resin beads. Mostly ion exchange resin beads have operation temperature lower than 100 ℃ and operation pressure lower than several psi, while the temperature and pressure of the primary cycle exceeds 300 ℃ and 2000 psi. Therefore lowering the temperature and pressure is very essential for the purification system, which make the system complex, enlarged, and costly.

To address the issue, we adopted the ceramic nanoparticles adsorbent as a substitute for the ion exchange resin. Ceramic nanoparticles have benefit from their intrinsic mechanical and chemical stability. In addition, ceramic nanoparticles undergoes endothermic adsorption process which represent higher adsorption performance under high temperature environment, which is very beneficial for the primary cycle of the nuclear power plant.

In this study, we selected cobalt as a target ion, whose radioactive forms (^{60}Co, ^{58}Co) are well known as the main source of radiation worker exposure. We will synthesized various types of ceramic nanoparticles by electrochemical anodization and test the cobalt ion adsorption ability under ambient temperature and high temperature (~300 ℃). Also, the adsorption mechanisms will be studied in terms of the adsorption species change and morphological change of the adsorbent.

This results are expected to give an alternative to design the CVCS structure, in particular for new type reactors such as Small Modular Reactor (SMR) which requires the simplest and optimized structures.

2. Method

2.1 Equipment and Materials

Metal wires (Al, Fe, Ni, Sn, Ti, V, Zr) of diameter 1.0 mm were purchased from Sigma-Aldrich for the fabrication of ceramic nanoparticles through anodization. Potassium Chloride (KCl) and Potassium Iodide (KI) powder, as electrolytes for anodization, was supplied by Sigma-Aldrich. Distilled Water was purchased from Daejung, Korea. Stainless steel Cathode frame as a counter electrode and anodization stand to hold the frame and adjust the level of iron wire was custom-made from Gumto Engineering, Korea. After anodization, the solution contains nanoparticles were filtered with *Steritop* of pore size 0.22 μm and 0.1 μm, obtained from Sigma-Aldrich.

For the adsorption experiment, cobalt chloride hexahydrate ($CoCl_2$•$6H_2O$) was obtained from Sigma-Aldrich. For the high temperature•pressure adsorption experiments, 1 Liter HC-276 Autoclave system was utilized (youngsung Tech, Korea).

2.2 Fabrication of Ceramic Nanoparticles through Electrochemical Anodization

300 mL of 1 M KCl and 1 M KI solution were served as an anodization electrolytes. For the stabilization, 6 hours of stirring was performed on the solutions. Each metal wire was prepared with length of 3 cm and served as a working electrode. [5].

15 V of voltage and 2 A of the electric current were applied to the system. In particular, for the fabrication of FeOOH, the temperature of the system was set to be lower than 5 ℃ by iced bath [5]. After the anodization, solution was filtered and dried, and the dehydrated samples were grinded as nanoparticles.

2.3 Cobalt Ion Adsorption Test

Cobalt solution was prepared by dissolving cobalt chloride ($CoCl_2$) in deionized water. The concentration of the solution was adjusted to 100 mg/L. The solution was stirred in 300 rpm for 6 hours to reach the equilibrium [7, 8].

For the ambient temperature adsorption test, solution test volume was set to 100 mL. Each sample was sealed with paraffin film to prevent the volume change by evaporation.

For the high temperature adsorption test, 100 mL test solution was put in autoclave. The system reaches to 350 ℃ in 30 min. In every 30 min, the temperature was decreased to 340 ℃ and soon increased to 350 ℃ to induce the convection inside the system, which mix the adsorbent particles.

After the electrosorption, the solution were filtered out and the concentration was measured. ICP-OES was utilized for the measurement of the cobalt concentration of the final product.

3. Results

3.1 Adsorption result under ambient temperature

Several types of ceramic nanoparticles adsorbents were tested. In all cases, initial cobalt concentration was 100 mg/L. Amount of the adsorbent in each experiment was 200 mg.

Table I. Cobalt concentration and adsorption capacity of various ceramic nanoparticle adsorbents under ambient temperature

Adsorbent	Cobalt Concentration after 2 h (mg/L)	Cobalt concentration after 24 h (mg/L)	Adsorption capacity after 24 h (mg/g)
AlOOH	90.50	86.14	6.93
Fe_3O_4	89.36	56.10	21.95
$Ni(OH)_2$	84.23	66.76	16.62
SnOOH	93.14	92.89	3.56
TiO_2	86.21	85.22	7.39
V_2O_5	95.16	86.16	6.92
ZrO_2	91.73	93.42	3.29

As the results, Fe_3O_4 and $Ni(OH)_2$ showed the best adsorption performance, 21.95 and 16.62 mg/g, respectively. In particular, $Ni(OH)_2$ showed better performance in the earlier time (2 h) than Fe_3O_4, but Fe_3O_4 exceeded the performance of $Ni(OH)_2$ in the later time (24 h). Other ceramic species showed relatively low performances.

3.2 Adsorption result under high temperature

Cobalt adsorption performance of Fe_3O_4 and $Ni(OH)_2$ nanoparticle adsorbents, which showed the best cobalt removal performance under ambient temperature, were tested under high temperature (90 ℃) and reactor temperature (350 ℃). Tests were conducted for 24 h, and initial cobalt concentration was 100 mg/L for each test.

Table II. Cobalt concentration and adsorption capacity of Fe_3O_4 and $Ni(OH)_2$ nanoparticle adsorbents under high temperature

Adsorbent	Final concentration (mg/L)		
	Ambient Temperature	High Temperature (90 ℃)	Reactor Temperature (350 ℃)
Fe_3O_4	56.10	39.40	5.69
$Ni(OH)_2$	66.76	48.53	7.53

Under the high temperature, both adsorbents showed improved adsorption performance than the samples under the ambient temperature, which was originated from the endothermic behavior of ceramic nanoparticle surface. Unlike the ion exchange reaction, surface adsorption on the ceramic nanoparticles are known to be endothermic, which gives superior performance under the higher temperature.

Besides, under the reactor temperature, the removal performances were superior. In the super high temperature water, several removal pathways participates in together. Cobalt ions form precipitates on the nucleation surface, and alloying reaction can be involved in the reaction. This will be shown in the further works.

4. Conclusions

In this study, we proposed a ceramic nanoparticle adsorbent based high temperature purification of CVCS. Under the high temperature close to the reactor coolant system environment, the adsorbents represented almost perfect performance, while the conventional ion exchange resin degrades and totally fails.

In future work, the adsorption environment will be further studied, along with adsorption species change and phase change of the adsorbents. Also, the mechanisms studies will be conducted throughout. The kinetics of the system will be further studied. This study will suggest an alternative purification strategy, and even will suggest optimized and simplified CVCS structure, in particular for SMR.

REFERENCES

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