Removal of Radioactive Cobalt(II) Ions in Chemical & Volume Control System (CVCS) by Electrosorption with Ceramic Nanoparticles Adsorbents Active Material

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1. Introduction

In the primary system of a nuclear reactor, metal corrosion produces a variety of metal ions, of which cobalt (Co) ions are of particular importance as the primary corrosion product [1, 2]. Cobalt ions react with neutrons to form radioactive activation products, most notably ^{60}Co and ^{58}Co, which emit strong gamma radiation, increasing the reactor radiation level and the risk of radiation exposure to workers [3]. Therefore, the effective removal of cobalt ions from the reactor primary system is an important research challenge to improve radiation management and operational safety of nuclear reactors [1].

Conventional methods of cobalt ion removal include chemical precipitation, ion exchange resins, and filtration technologies [2]. However, these technologies require a continuous supply of chemicals or periodic regeneration processes, and have limitations in terms of long-term operational reliability and economic viability. Therefore, technologies that selectively adsorb and remove metal ions using electrochemical methods are attracting attention, and cobalt ion adsorption using electrodes has great potential for practical applications due to its high efficiency and simple process [4].

In this study, we aim to develop a cobalt ion adsorption technology using electrodes and evaluate the feasibility of applying it in the primary system of a nuclear reactor. In particular, we propose the use of ceramic nanoparticles as cathode active materials to effectively adsorb cobalt ions [4]. Ceramic nanoparticles have a high specific surface area, good chemical and physical stability, and these properties play an important role in maximizing the adsorption performance of the electrode [4].

The results of this research are expected to improve the efficiency of reactor radiation management and contribute to reducing operating costs and enhancing safety of nuclear power plants in the long term.

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2. Method

2.1 Equipment and Materials

For the fabrication of ceramic nanoparticles through anodization, iron, aluminum, tin and titanium wires of diameter 1.0 mm were purchased from Sigma-Aldrich. Potassium Chloride (KCl) and Potassium Iodide (KI) powder, as electrolytes for anodization, of purity higher than 99% 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 fabrication of electrode, Copper foil of thickness 0.03 mm was purchased from GFM, Korea. Platinum sheet of thickness 0.1 mm, which were used as a counter electrode of electrosorption test, were also obtained from GFM, Korea. For the fabrication of the electrodes, Super P as a conductive additive, Carboxymethyl cellulose (CMC) and Poly(acrylic acid) (PAA) as binders were supplied by Sigma-Aldrich. For the adsorption test, cobalt chloride was purchased from Sigma-Aldrich.

2.2 Fabrication of Ceramic Nanoparticles through Electrochemical Anodization

As an electrolyte, 1 M KCl and 1 M KI were dissolved in 300 mL deionized water. These solutions were stirred in 300 rpm for 6 hours to reach the equilibrium. For the anodization setup, each metal wire was used as a working electrode, while the stainless steel plate was used as a counter electrolyte. Metal wires were prepared with length of 3 cm [5].

15 V of voltage were applied to the system. Electric current was set to be lower than 2 A. During the fabrication, the electrolyte were stirred in 100 rpm. In particular, for the fabrication of FeOOH, the temperature of the system was set to be lower than 5 ℃ by chiller [5].

After the anodization, solution with the fabricated nanoparticle was filtered with the vacuum filter. During the filtration, deionized water is continuously supplied to completely rinse out the electrolyte component. The nanoparticles were collected and dried at 50 ℃ for 24 hours. After the drying, samples were grinded with 45 μm sized sieve.

2.3 Fabrication of Ceramic Nanoparticles based Anode

Anode, a working electrode of the electrosorption experiments, was fabricated by slurry-casting method. Super P was used as conductive additive, and the mixture of CMC and PAA was used as binder. Aqueous binder was produced by dissolving 2.5 wt% of CMC and 2.5 wt% of PAA in deionized water [6].

Active materials, conductive additive and binder were blended in mass ratio of 8:1:1. The mixture of electrode materials were dissolved in DI water by ball-milling method. Prepared slurry was casted on the copper foil with doctor blade. After the coating, the electrode was dried in vacuum oven at 70 ℃ for 12 hours.

2.4 Electrosorption Test

Cobalt solution was prepared by dissolving cobalt chloride ($CoCl_2$) in deionized water. The concentration of the solution was adjusted to 1000 mg/kg. The solution was stirred in 300 rpm for 6 hours to reach the equilibrium [7, 8].

Experimental setup for the electrosorption test was as figure 1. Distance between the electrodes was 5 cm. Surface area of the electrodes under the solution was 2.5 x 3.3 cm^2. During the experiment, 5 V of voltage was applied, and the solution was stirred in 100 rpm.

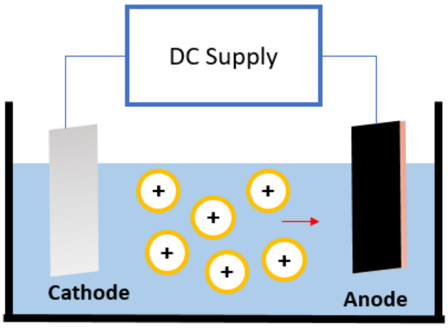

Fig. 1. Experimental setup for the electrosorption test

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 Cobalt Adsorption Capacity Results

3.1.1. TiO_2 based electrode test

Two samples of Co solution of volume 100 mL and Co concentration 1000 mg/L was tested. Total reaction time was 12 hours, which was considered to reach the equilibrium. The results was shown as below.

Table I. Cobalt adsorption capacity of TiO_2 based electrode

	Final Concentration (mg/L)	Adsorption Capacity (mg)
Sample 1	550.5	44.95
Sample 2	595.6	40.44
Average	573.05	42.695

As the results, average adsorption capacity was 42.695 mg, and the average adsorption efficiency of the system was about 57.3 % for high concentration Co solution.

4. Conclusions

In this study, we proposed an electrochemical adsorption technology for the effective removal of cobalt ions from nuclear reactor primary systems. In order to overcome the limitations of conventional chemical precipitation, ion exchange resin, and filtration methods, an electrode-based selective adsorption method was studied, and in particular, a strategy of applying ceramic nanoparticles as cathode active material was proposed. It was found that ceramic nanoparticles offer high specific surface area and good chemical and physical stability, which can maximize the adsorption performance of the electrode [9].

In this study, we also introduced the use of anodization technology to prepare the nanoparticles. The anodization process is highly reproducible and mass-producible, and the safety of the process makes it a practical technology that can be applied in a reactor environment. The electrode-based cobalt ion adsorption technology developed in this study can contribute to the reduction of radioactive activation products in the reactor primary system, and is expected to be useful in lowering radiation levels and reducing the risk of exposure to workers.

In future work, optimization studies of different electrode materials and electrochemical operation parameters will be conducted to further evaluate the applicability in real reactor environments. In addition, the research will be expanded to maximize practicality by studying long-term durability and repeated use. The results of this research are expected to contribute to improving radiation safety and increasing reactor operation efficiency in the nuclear industry.

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