Treatment of Radionuclide From CRUD Using Underwater Microwave Plasma

Seongsik Nam and Wooyong Um*
Pohang University of Science and Technology, 77, Cheongam-ro, Nam-gu, Pohang-si, Gyeongsangbuk-do, Republic of Korea
*wooyongum@postech.ac.kr

1. Introduction

As nuclear power plants (NPPs) are getting older, the levels of radioactive contamination are also increased. CRUD (Chalk River Unidentified Deposit) is a kind of corrosion product accumulated on the metal surfaces of NPP as the operation time goes by. Decontamination of CRUD is really important task because radionuclides such as ^{60}Co (γ-emitter) can be deposited and react with the constituents of CRUD. The objective of this study is to decontaminate radionuclides such as ^{60}Co from the CRUD waste using microwave plasma approach in underwater system. In comparison with other techniques, microwave plasma in water technique was developed to provide high reliability for the environmental aspect using, efficient generation of plasma and high frequency radical.

2. Materials and Methods

2.1 Experimental setups of underwater plasma system

The experimental set-up of CRUD decontamination by underwater plasma system is described in Fig. 1. It includes argon and hydrogen supply system, microwave plasma torch, high voltage power supply and radical analysis (optical emission spectrometer) instruments. This system can generate microwave plasma that can react with synthesized CRUD samples inside of the acid solution.

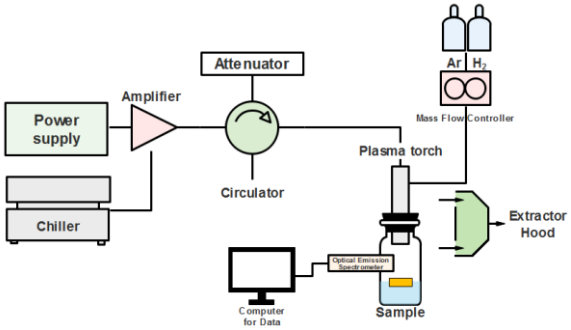

Fig. 1. Schematic diagram of Underwater Microwave Plasma system.

Inside of acid solution, typical chelate agent, EDTA was added to synthetic decontamination liquid waste and measured the kinds of generated radical at the same time with optical emission spectrometer. In addition, UV-VIS spectrometer was used to measure the oxidation state of cobalt ions.

2.2 Synthesis of Co-doped simulated CRUD

A doping method was used to deposit CRUD on stainless steel SUS 304 (Ni: 8~11%, Cr: 18~20%, Fe). The surfaces of metallic (SUS 304) samples were abraded using silicon carbide paper (#1200), then washed in acetone (99.5%, KANTO) with ultrasonic cleaner (BRANSON 1510) for 5 min. After that, 0.25 mL of cobalt nitrate solution (150 g/mL) was spread on the metallic coupon and dried at 100℃. This procedure was repeated four times. Finally, 1 mL of cobalt nitrate was doped on the metal coupon. To form an oxide layer on the coupon surface, it was heat treated at 700℃ for 24 h. XRD and XPS were conducted to characterize CRUD samples.

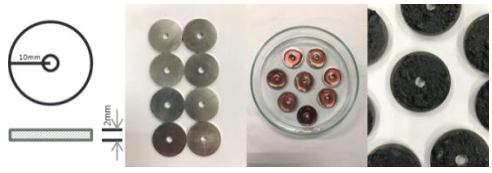

Fig. 2. Schematic diagram and doped metal coupon.

3. Results & Discussion

To see the effect of plasma, the synthesized simulated CRUD samples were characterized using x-ray diffraction (XRD) to identify the mineralogy. Compared with original data, the intensity of cobalt ferrite was reduced when it was treated by H_2 gas plasma. In details, Co^{2+} and Fe^{3+} changed theirs oxidation state to reduced state or elemental metal ions such as Co^0, Fe^0, and Fe^{2+} respectively.

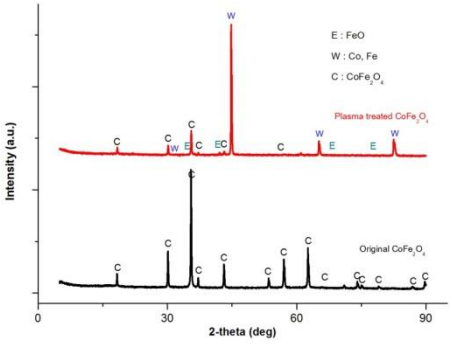

Fig. 3. XRD patterns of original $CoFe_2O_4$ (black) and after treatment with H_2 gas plasma (red).

The chemical composition of the samples is further studied using XPS analysis. The counts of Co, Fe, and O elements are decreased after H_2 plasma treatment (Fig.4).

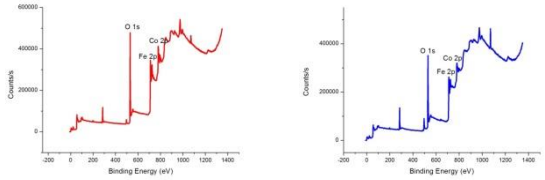

Fig. 4. XPS spectra of the CRUD (a) survey spectrum, (b) survey spectrum treated with H_2 gas plasma.

Additional microwave plasma generated by argon gases in the liquid condition was applied and the oxidation number of cobalt(Ⅱ) was changed to oxidized state, Co(Ⅲ), which is different from the result of plasma with H_2 gas condition. Reference peaks of cobalt(Ⅲ), 382 nm and 530 nm increased with reaction time (Table 1). Cobalt(Ⅱ)-EDTA complex was also found in the solution. According to Table 1, oxidation state of cobalt ions is changed to 2^+ to 3^+, during the argon microwave plasma reacted with simulated CRUD.

Table 1. Change of UV absorbance at 532 nm with increasing reaction time

Time	Absorbance at 532 nm wavelength
5 min	0.103
10 min	0.115
20 min	0.132
30 min	0.147

4. Conclusion

The microwave plasma in underwater system can be applied to decontamination process for the selective decontamination and volume reduction of the metal wastes. Further study will be investigated with varying decontamination reagent, power, temperature, ion conductivity, and pH. Development and application of our results will contribute to make an effective and safe decontamination process of nuclear power plants.

ACKNOWLEDGEMENT

This work was supported by Nuclear Research & Development Program of the National Research Foundation (NRF) grant (NRF- 2018M2B2A9065874) funded by the Korean government (MSICT).

REFERENCES

[1] Kim W-S, Nam S, Chang S, Kim H, Um W. Removal of Chalk River unidentified deposit (CRUD) radioactive waste by enhanced electro-kinetic process. Journal of Industrial and Engineering Chemistry,7:89-96, 2015.

[2] NEA, The Decommissioning and Dismantling of Nuclear Facilities, 2002.