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Bias Voltage Effects on the Removal of Co and Mo by SF_6/O_2 Plasma for Radioactive Waste Decontamination

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1. Introduction

The equipments and components used in nuclear power plant such as piping and valves usually are contaminated by TRU elements and radioactive corrosion and fission products. In this experiment, the principle contaminants Co of corrosion product and Mo of fission product are examined by SF_6/O_2 plasma. To enhance the reaction rate, DC bias voltage is applied on the specimens. Experimental results demonstrate the applicability and effectiveness of plasma decontamination processing. Results show that the reaction rate of molybdenum is 10.21 *u*m/min. at 290 ^oC. In the case of cobalt, etching reaction hardly takes place at low temperatures. However, it begins to increase as temperature exceeds 350 ^oC. The reaction rate is 2.56 *u*m/min. at 420 ^oC. Ion-assisted experiments are under conduction. To support these results OES, SEM, and AES analysis are followed.

2. Methods and Results

Diode type and r.f. (13.56 MHz) plasma system power up to 600 W is used. Processing chamber has feed through for DC negative bias (-500 V, 1 A) which can be applied to the substrate (Figure 1). The distance between electrodes remains 10 cm during the current experiments. Sample can be heated up to 1200˚C with halogen heater. Mass flow controller can control SF_6 and O_2 gas finely and total flow rate is 50sccm. Total gas pressure of inside chamber is maintained at around 0.45 Torr during experiments.

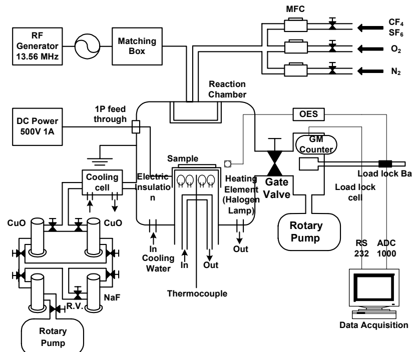


Figure 1. Schematic of plasma reaction apparatus.

2.1 Sample Preparation

Non-radioactive metal samples (Co, Mo) with 99.8% purity are used as specimens. Specimens are manufactured to a thin disk of 0.7mm thickness from rod 10 mm and 5mm each radius using low-speed diamond cutter. Prior to the sample loading, the surfaces of the specimens are polished as mirror-like by 1200 grit SiC paper, cleaned with ultrasonic cleaner and baked at 200 °C for 10 minutes in a vacuum to evaporate the absorbed moisture on the surface.

2.2 Experimental Procedure

Surface removal reaction carried out with 220 W r.f. power at 0.45 Torr of pressure during 30 min. To find out the most efficient SF_6 and O_2 gas mixture ratio, this experiment begins measuring the etching rate under changing O_2 ratio. At the most efficient O_2 ratio, etching rate of each specimen is measured under different temperatures. Etching rate is determined by weight loss measurement before and after the reaction with an electro-micro balance (BP210D, Satorius) whose sensitivity limit is 10^{-5}g. Weight loss is expressed in *u*m/min. OES (Optical Emission Spectroscopy) analysis is accompanied with the main experiments to diagnose and determine the plasma parameters and thus to understand the reaction mechanism. At the same condition, negative bias is applied. This experiment is under conduction.

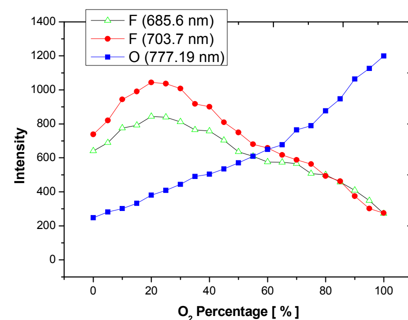


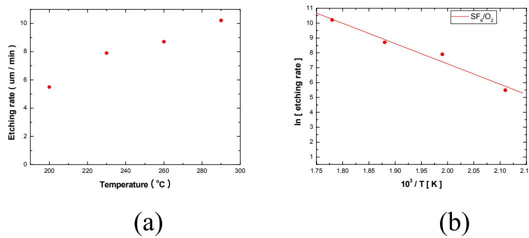
Figure 2. OES analysis of SF_6/O_2 plasma

Figure 2 shows that the most efficient O_2 rate of SF_6/O_2 plasma is about 20 %. The fluorine atom quantity has the maximum value at O_2 rate is 20 % It

appears that essential reaction of the surface is fluorination reaction.

2.3 Experimental Results

Mo etching rate is increased as temperature goes up under SF_6/O_2 plasma (Figure3). In the results of SEM analysis before and after, we can confirm the intensive reaction of surface (Figure 5)



(a) (b)

Figure 3. (a) Etching rate of Mo as temperature at SF_6/O_2 plasma. (b) Arrhenius plot

The etching rate of Co metal is very low (< 0.03 *u*m/min) below 350 °C temperature. But etching reaction begin to occur around 350 °C. Unfortunately, however, the etching rate attained at 380°C was not high enough to be useful for the practical decontamination. Therefore, in order to examine. The reaction rate enhancement through the ion-assisted etching mechanism negative bias voltage was applied to the substrate of the metallic cobalt specimen. In the case of applying bias, voltage etching rate increase rapidly about 4 times than without bias voltage at 400°C. as shown Fig. 5.

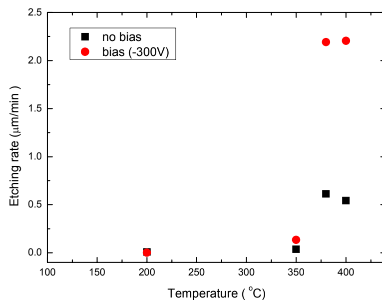
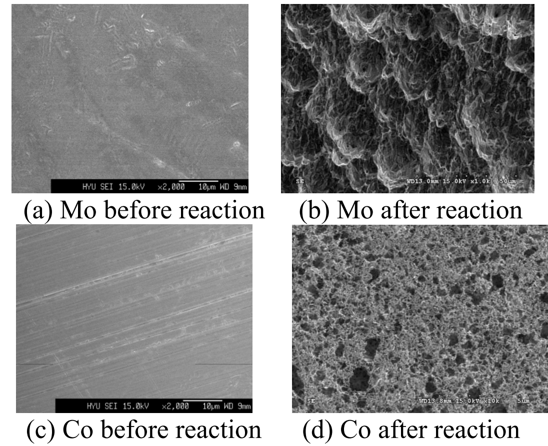


Figure 4. Etching rate of Co as temperature at SF_6/O_2 plasma

SEM analysis at 400 °C shows that reacted surface get rougher after the reaction (Figure 5.).



(a) Mo before reaction (b) Mo after reaction
(c) Co before reaction (d) Co after reaction

Figure 5. SEM analysis of Mo (290 °C) and Co (400 °C) before and after the reaction under SF_6/O_2 plasma (× 10K)

3. Conclusion

This experiment demonstrates the feasibility of plasma decontamination processing. Surface removal reaction is the most efficient at 20~25 % O_2 percentage. Under the same conditions, negative bias applying experiments are being carried out. At 400°C, etching rate of cobalt is 0.54 ㎛/min. and in the case of applying -300V bias voltage, maximum etching rate reached 2.2 ㎛/min. More through experiments are continued.

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