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Study of behaviors of aluminum overlayers deposited on uranium via AES, EELS, and XPS

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ABSTRACT

Aluminum overlayers on uranium were prepared by sputtering at room temperature in an ultra-high vacuum chamber. The growth mode of aluminum overlayers and behaviors of the Al/U interface reaction were studied in situ by auger electron spectroscopy, electron energy loss spectroscopy, and X-ray photoelectron spectroscopy. The results suggested that the interdiffusion took place at the Al/U interface during the initial stage of deposition. The U4f spectra of the Al/U interface showed strong correlation satellites at binding energies of 380.4 and 392.7 eV and plasma loss features at 404.2 eV, respectively. The interactions between aluminum and uranium yielded the intermetallic compound of UAl_x, inducing the shift to a low binding energy for Al2p peaks. The results indicated that aluminum overlayers were formed on the uranium by sputtering in an island growth mode.

1. Introduction

The corrosion reactions (e.g., oxidation of uranium) are of great concern due to the importance in military and commercial applications. It is easy for uranium to form oxide layers on the surface in atmosphere, which will greatly reduce its performance [1–3]. In order to improve the corrosion resistance, the deposition of compact films on the surface has been suggested as a feasible way, which can prevent uranium from further oxidation. Egert et al. [4] argued that the aluminum overlayer was a preferable choice. The interaction between Al and U, the behavior at Al/U interface, and their correlation seem to be of great importance. Gouder et al. studied the behavior of U overlayers on various substrates (e.g., Pt [5], Pd [6], graphite [7], and Al and Mg [8]) by auger electron spectroscopy (AES), X-ray photoelectron spectroscopy (XPS), and electron energy loss spectroscopy (EELS). In a previous work [9], the aluminum film on uranium was prepared by magnetron sputtering with and without a circulated-argon ion bombardment process, and the AES study displayed that the significant diffusion and chemical reactions took place at the interface with the formation of UAl_3 species due to the circulated-argon ion bombardment. Al and Ti coatings on uranium were also investigated by AES and SEM [10–13].

Although the aluminum film on uranium has been reported [4,9,11], the study of this issue still remains very limited. Precise knowledge of fundamental properties of aluminum overlayers is essential for a wide range of functional applications. In this work, the behaviors of the Al overlayer deposited in situ on the uranium substrate were studied in detail by AES, EELS, and XPS. The growth mode of aluminum overlayers was proposed.

2. Experimental

Al overlayers were deposited on uranium through argon-ion bombardment of aluminum target with energy of ~3 keV. During deposition, the aluminum target was set on the uranium substrate with an angle less than 90°, as shown in Fig. 1. The aluminum target was composed of polycrystalline Al with purity above 99.99%. The uranium disc was 9 mm in radius and 2 mm in thickness. Both targets were cleaned by mechanical polishing before being introduced into the spectrometers and by argon ion sputtering in the ultra high vacuum chamber. Different samples were obtained by varying the deposition time.

XPS data were acquired using the PHI-5600 ESCA instrument equipped with a hemispherical analyzer using the Mg Kα (1253.6 eV) radiation with a resolution of 0.8 eV. AES data were

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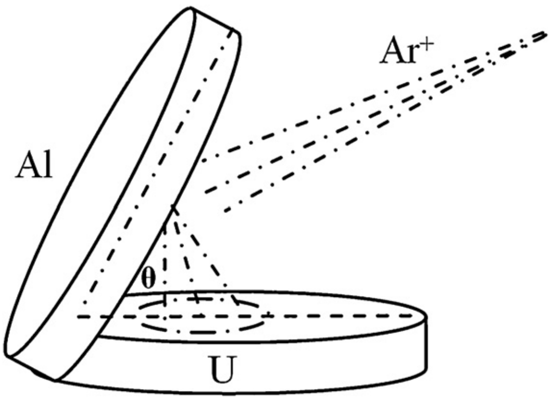

Fig. 1. Diagrammatic sketch of experimental setting.

acquired using the PHI-650 SAM instrument equipped with a LaB_6 electron gun and a cylindrical mirror analyzer (CMA), which had the function of EELS. The base pressure of analysis chamber was less than 5×10^{-7} Pa during the data record and less than 5×10^{-5} Pa during the deposition process. During AES data record, the electron gun worked with beam current of about 120 nA under 3 kV, while the beam current of about 30 nA under 200 V was used for EELS data acquirement.

3. Results and discussion

3.1. AES and EELS study of Al/U interface

The surface composition was determined by using the U(OPV) and Al(LVV) auger emissions. Because of the similar kinetic energies of these auger emissions, the partial overlap prevented a simple peak-to-peak intensity analysis. Fig. 2 shows the AES spectra of the uranium substrate during deposition. Obvious changes can be found in the AES spectra. The intensities of uranium auger peaks decrease gradually while those of aluminum increase with the increasing of aluminum coverage. When the thickness is about 0.745 monolayer (ML), the kinetic energy of Al(LVV) is 66.96 eV, with a 1.04 eV shift to low kinetic energy compared to that of metallic Al. The U(OPV) emission shows no chemical shift or decrease in intensity. It should be noted that Al(LVV) emissions overlapped with the tail of U(OPV), which broadens the FWHM of both peaks and leads to the shift of Al(LVV) or some changes to chemical environment induced by the aluminum deposition. With the increase of aluminum coverage, uranium auger peaks decrease in intensity. When the thickness of aluminum overlayers is 2.678 ML, the kinetic energy of Al(LVV) is 68.0 eV, which corresponds to the metallic aluminum. Note that the thicknesses of aluminum overlayers mentioned in this work always refer to those calculated by AES spectra.

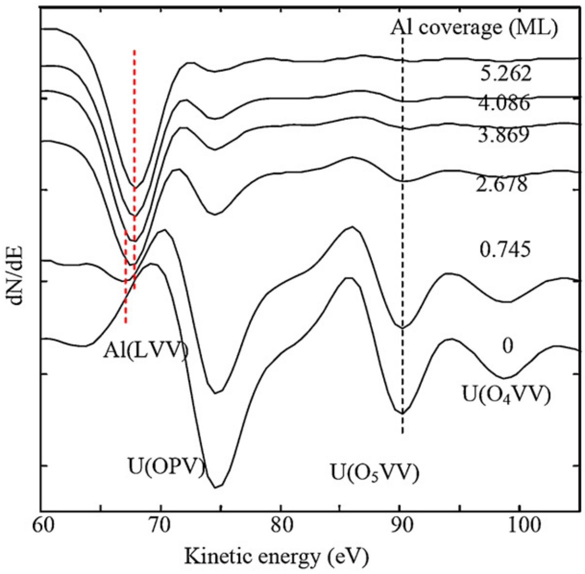

Fig. 2. AES spectra of uranium substrate with increasing Al coverage at room temperature.

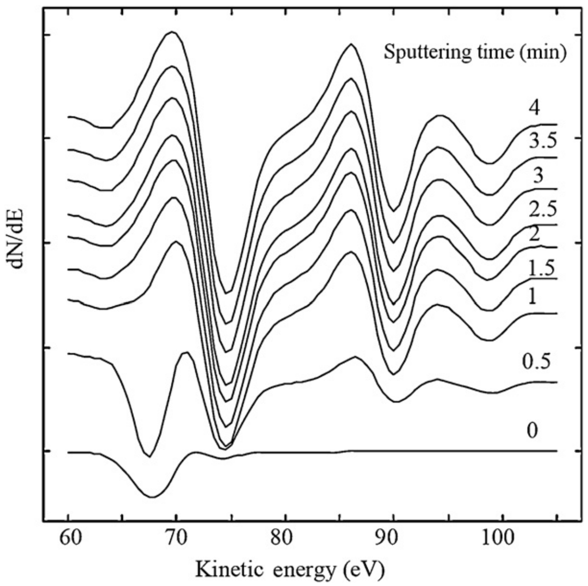

Fig. 3. AES spectra in depth profile of aluminum overlayers with etching time at room temperature.

The depth profile was carried out to reveal the behavior at the interface. Fig. 3 shows the AES spectra of aluminum overlayers with etching time of 0–4 min. The Al(LVV) peaks decrease in intensity, while those of uranium increase as a function of etching time, with no obvious shift for both peaks. After etching for 1.5 min, the weak signal for aluminum is detected. In the study of Lv et al. [9], UAl_3 was found at the interface of the aluminum film on the uranium substrate deposited by magnetic sputtering. However, the similar phenomenon is not observed in our study. It may be related to the different growth processes in both studies, such as the ion bombardment process and the resultant energies of aluminum atoms and substrate temperatures. Further investigations are necessary to clarify this issue.

In the initial stage of deposition (coverage less than 0.745 ML), EELS was taken on the same surface using an electron beam with a kinetic energy of 200 eV close to that of AES spectra. Fig. 4 shows EELS spectra of the clean surfaces of uranium and aluminum. The peaks with energy loss of 4.2, 13.3, and 20.0 eV correspond to the surface plasma (SP), bulk plasma (BP), and secondary surface plasma energy losses of uranium, respectively. The peaks at energies of 10.6, 15.8, 25.8, and 31.5 eV are due to the energy loss of aluminum SP, BP, composite of SP and BP, and secondary BP, respectively. Fig. 5 shows EELS spectra of the uranium substrate with increasing Al coverage. The SP energy loss is more sensitive to the surface environment than BP, so the energy loss of 4.2 eV decreases in intensity rapidly with increasing Al coverage. Unfortunately, the overlap exists in the energy loss of 9–16 eV, so it is hard for us to identify whether the changes of spectra are evoked by the overlap or the interaction between Al and U. When the aluminum coverage is about 3.869 ML, it is almost the signal of pure metallic aluminum. Comparing the 4.2 eV peaks in Figs. 4(a) and 5, we know that the