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A nitriding treatment of aluminum films by atmospheric pressure discharge (1) Effects of substrate temperature for nitriding treatment of aluminum films

大気圧プラズマを用いたアルミニウム膜の窒化処理（１）表面窒化反応に対する温度の影響

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Nitride process for aluminum film was investigated under the atmospheric pressure condition with the mixing of helium and nitrogen gases. At low temperature of 100 ºC, oxygen atoms were released from the surface and nitrogen atoms were bounded to Al atoms. The small peak of XRD pattern corresponding to AlN crystalline was detected when the flow ratio of N_2 gas to He gas is larger.

1. Introduction

The non-destructive inspection technique using ultrasonic waves, generated by an electromagnetic acoustic transducer (EMAT), has been introduced into the in-service inspection of fast breeder reactors. The EMAT allows us to inspect the defect without any contact material between the transducer [1-3]. However, the EMAT consists of permanent magnets and transmission coil so that it is hard to apply to the inspection of the pipe at high temperature condition of 550 ºC for the operating plant system. In these circumstances, a proposal has been made of a new EMAT driven by a pulse current method. This EMAT allows us to make some multi-layers piled alternately with insulation and conduction materials, which works as a current circuit on a substrate. The author has already studied the deposition multi-layers of (AlN) by introducing both an insulator with high thermal conductivity and a copper conductor with high electric conductivity, but the bad adhesion on the substrate and the micro-cracks on the film were observed due to large residual stresses. This is a problem to be solved for the current driven EMAT. The present study aims to investigate the fundamental process for nitriding treatments of aluminum films by atmospheric pressure plasma in order to improve the deposition system.

2. Experimental setup

The experimental setup for depositing AlN films is shown in Fig. 1. A cylindrical chamber has a substrate holder and a large electrode at the top and bottom, respectively, operating at a frequency of 100 MHz. Evacuation was done at the base pressure of 1×10^{-5} Pa with a turbo molecular pump in order to remove the influence of residual gases. After this process, the chamber was filled with the mixture of He and N_2 gases up to a working pressure at 9×10^4 Pa. The gas mixing ratio was steadily adjusted by the mass flow controllers (MFCs).

Cupper tungsten plates were used as a substrate. Before the exposure of atmospheric pressure plasma, a aluminum film was deposited on the substrate using magnetron sputtering device which generates Ar plasma at input power of 200 W. The aluminum film on the substrate was heated up to 400 ºC by a resistive heater mounted inside the substrate holder, which was kept at this temperature within ± 10 ºC while making deposition. A disk of pure aluminum (99.99 % in purity) with 3 inch in diameter and 2 mm in thickness was mounted on the electrode. The substrate was moved in position

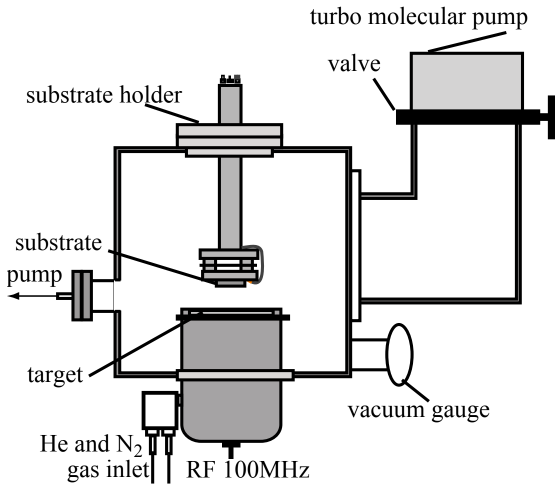

Fig.1. Schematic of experimental setup.

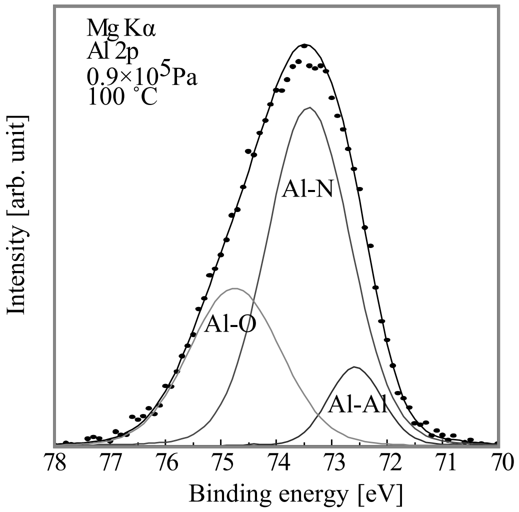

Fig.2 XPS spectrum for Al 2p.

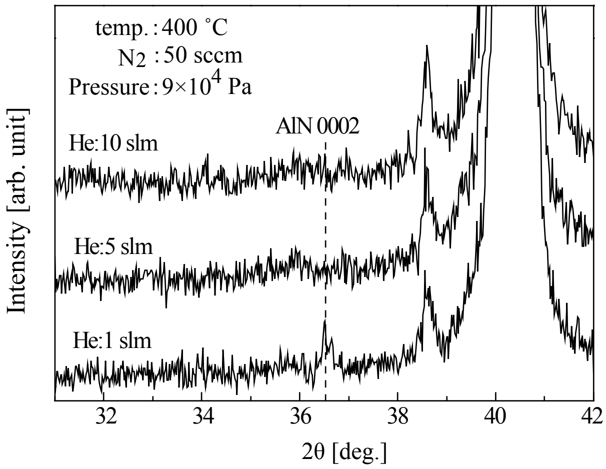

Fig.3. XRD patterns for various He gas flow.

so that the deposition flux varied spatially from the target. The distance between the substrate holder and the electrode was within 1mm. The two kinds of highly purified gases, helium (He) and nitrogen (N_2), were supplied to the system as the reactive gases. The monitored gas flow of N_2 was always set at 50 sccm during operation.

X-ray photoelectron spectroscopy (XPS, Shimazu AXIS-165), X-ray diffraction (XRD, Rigaku Rint2000), scanning electron microscopy (SEM: HITACHI HF-3000) were equipped to do the measurements and analyses of the chemical bonding states, surface morphology.

3. Results and discussion

Fig.2 shows an XPS spectrum for Al 2p core level of the aluminum film exposed by He/N_2 mixture atmospheric plasma at 100 ºC. Main spectrum was separated to three spectra corresponding to Al-N, Al-O and Al-Al. The atomic ratio of Al-N is about 30-50 % at the investigated temperature. It is noted that Al-O and Al-Al components only exist before exposure. By the effect of the atmospheric plasma, oxygen atoms were released from the surface and nitrogen atoms were bounded to Al atoms at low temperature of 100 ºC.

The surface morphologies were not noticeably different up to 400 ºC. On the other hand, AlN crystalline was grown on the aluminum surface when the total pressure was 2 x 10^4 Pa, and the flow rate of He and N_2 gases was 50 sccm and 20 sccm, respectively. In this case, the flow ratio of N_2 gas to He gas was considerably high. It suggests that it might be enough nitrogen atoms to form the AlN crystalline.

XRD patterns of the aluminum films were investigated at atmospheric pressure condition, when He gas flow was controlled from 1 slm to 10 slm, and the N_2 gas flow is constant flow of 50 sccm. Fig.3 shows the XRD patterns of the aluminum films for the different He gas flow. The weak diffraction pattern corresponding to AlN 0002 was detected at He gas flow of 1 slm, while no peaks related to AlN crystalline were observed at 5 slm and 10 slm.

4. Summary

Nitride process for aluminum films by He and N_2 mixture plasma at 9 x 10^4 Pa were investigated from 100 ºC to 400 ºC. At low temperature of 100 ºC, nitrogen atoms were bounded to Al atoms on the surface. AlN crystalline was found when the flow ratio of N_2 gas to He gas is larger.

Acknowledgement

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References

[1] R.B. Thompson, IEEE Transactions on Sonic and Ultrasonics, Vol. **SU-20** (1973) 340

[2] B.W .Maxfield, A. Kuramoto, J.K. Hulbert, Material Evaluation **45** (1987) 1166

[3] Y. Kurozumi, M. Higashi, T. Satou, M. Nishikawa, Material. Evaluation **59** (2001) 638