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TEM Study of Aluminum Oxynitride Films Prepared by Reactive R.F.-Sputtering

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The versatile of physical properties of Aluminum Oxynitride (AlO_XN_Y) offers many applications in various technologies, e. g., microelectronics, optics and protective coatings [1-3]. AlO_XN_Y films are generally prepared by reactive sputtering in a mixture of nitrogen and oxygen ($N_2 + O_2$) gases [4] with aluminum target. However, control of the oxygen concentration in the reactive sputtering system is difficult due to the high reactivity of oxygen to aluminum. To improve the reactive sputtering system, a modified reactive rf-sputtering using N_2 + Ar gas mixture with Al_2O_3 target was initiated. This study reports the microstructure of AlO_XN_Y films obtained by this new method.

Al_2O_3 (99.6%) target (Nikkato Co., Japan) was sputtered using a facing target-type sputtering system (FTS-2R Model, Osaka Vacuum Co.) with 980 W power in a mixture of highly purified Ar and N gases (99.9999%) with the flow rate of 10 and 30 sccm (standard cubic centimeter per minute), respectively. Si substrates were preheated to 350 °C with a bias of -100V during the deposition. Total thickness of the film was approximately 2 μm. Cross-sectional TEM specimens were prepared using the FIB (Focus Ion Beam) technique. Plane-view TEM specimens were prepared by mechanical griding/poilishing followed by ion milling at -60℃ with a final accelerating voltage at 0.5 kV. Film micro-structure and -chemistry were examined by JEM 2100 FEGTEM. Detail chemical composition was analyzed by JEOL EPMA.

Figure 1 shows the change in indentation hardness (H_{pl}) of the films produced at different biases and at different substrate temperatures. The indentation hardness increases with increasing bias as well as with substrate temperature. The softest film deposited at room temperature without substrate bias showed an amorphous-like and/or poly-nanocrystalline XRD pattern (Fig. 2a). However, the hardest film deposited at 350 ^{o}C with a substrate bias of -100V revealed a crystalline structure (Fig. 2b); this is also evidenced by the HRTEM and diffraction contrast images (Figs. 3 and 4). The cross-sectional TEM dark-field image also depicts that the elongated columnar crystal strucuture of film is parallel to the direction of film growth (Fig. 3). An armorphous interlayer was found at the interface between the film and Si substrate (Fig. 5). The electron diffraction patterns obtained from the film (Fig. 4) and the interface region (Fig. 6) suggest that the film consists mainly of hexagonal AlN (JCPDS 25-1133) poly-nanocrystallites with some degrees of preferred crystal growth orientation. In addition to the presence of Al and N in the film, a high concentration of oxygen (~10% or more) was found in the EPMA and TEM EDS results. Of particular interest, AlN was the only oxygenated material that matched well with the diffraction pattern, yet oxygen was found evenly distributed throughout the film. This finding is difficult to interpret, but one possible explanation is that the film was composed mainly of nanocrystalline AlN with small amounts of AlOx or AlO_XN_Y. Further investigation is in progress to confirm the speculation and clarify the behavior of oxygen during the process of film deposition.

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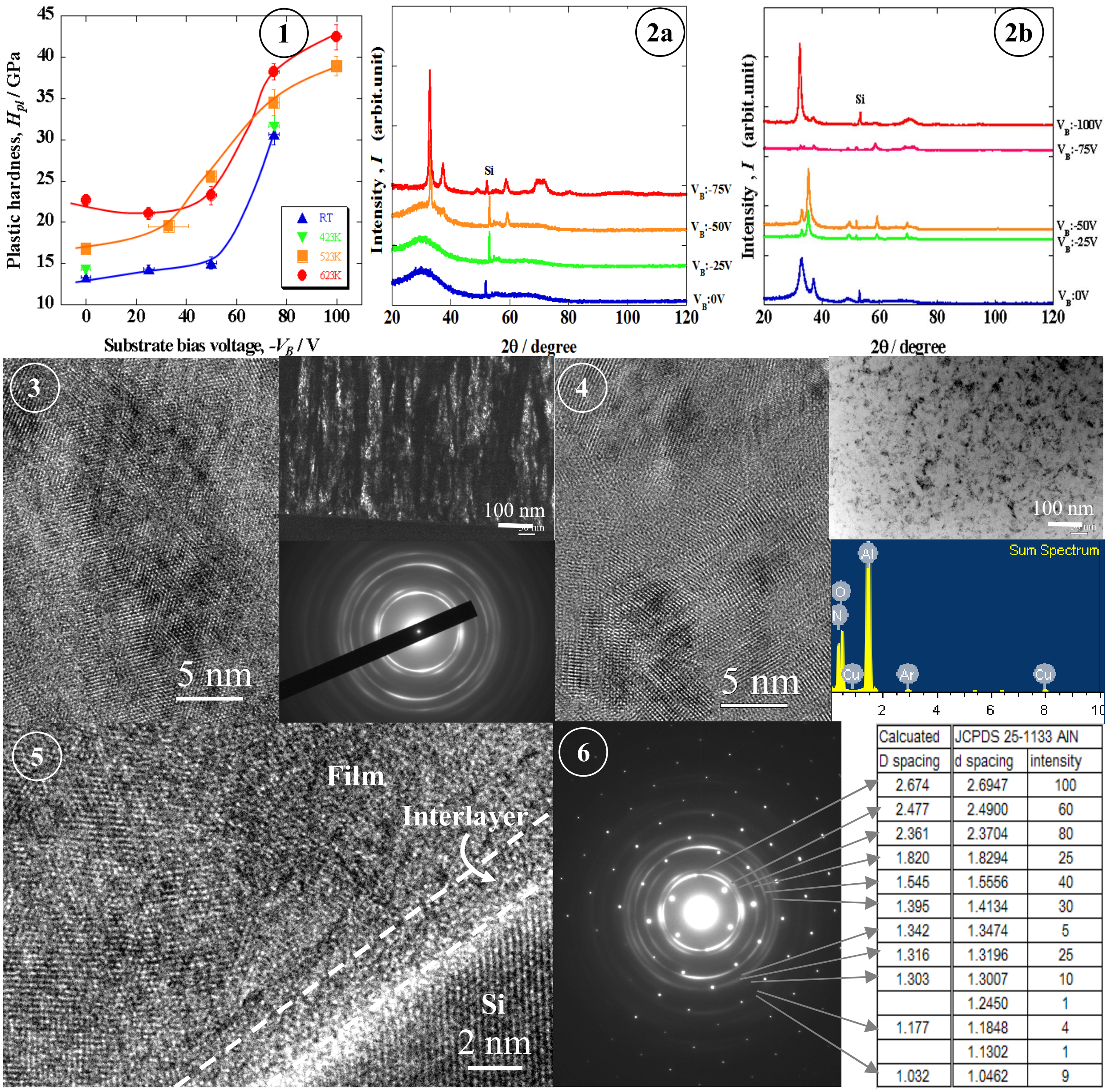

Calcuated	JCPDS 25-1133 AlN	
D spacing	d spacing	intensity
2.674	2.6947	100
2.477	2.4900	60
2.361	2.3704	80
1.820	1.8294	25
1.545	1.5556	40
1.395	1.4134	30
1.342	1.3474	5
1.316	1.3196	25
1.303	1.3007	10
	1.2450	1
1.177	1.1848	4
	1.1302	1
1.032	1.0462	9

Fig. 1. Diagram shows plastic hardness (H_{pl}) of film with different bias voltage and substrate temperature.

Fig. 2. XRD patterns of films deposited on different substrate biases: at 25 °C (2a) and 350 °C (2b).

Fig. 3. Cross-sectional DF and HRTEM images of the film show poly-nanocrystalline columnar structure along the film growth direction. SAD pattern matches with typical AlN (Fig. 6).

Fig. 4. Plane-view BF and HRTEM images of the film show a typical poly-crystalline structure. EDS spectrum showing Al, N and O throughout the film.

Fig. 5. Cross-sectional HRTEM image shows an amorphous interlayer at film/Si interface.

Fig. 6. Analysis of SAD pattern obtained from film/Si interface area (Fig. 5) reveals the film is composed mainly of nanocrystalline AlN. Diffraction spots are contributed from Si substrate.

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