Armenian Journal of Physics, 2016, vol. 9, issue 1, pp. 15-19

Microstructure and Residual Stress Measurement of Ag/Glass Thin Films Using In-Situ High-Temperature X-ray Diffraction

A. Zolanvari*, S. Hasan Sagha, F. Eshaghi, Z. Shahedi, A. Zendehnam
Department of Physics, Faculty of Science, Arak University, Arak, 38156-8-8349, Iran

*E-mail: a-zolanvar@araku.ac.ir

Received 19 January 2016

Abstract. In this study pure Ag particles were deposited on the glass substrate using DC magnetron sputtering at room temperature and Ag thin film with 80nm thickness was prepared. To investigate the effect of high temperature on the crystal structure of Ag thin film, an in-situ HT-XRD instrument has been used. Microstructural properties of Ag thin film have been studied in several temperatures. Micrograin size of different atomic planes of Ag thin film during HT-XRD process was calculated. Residual stress of as-deposited and post annealed Ag thin film at (111) planes was measured using $\mathrm{Sin}^2\psi$ method.

Keywords: Ag/glass thin films, DC magnetron sputtering, In-situ HT-XRD, Residual stress

Introduction

Ag thin films have been studied widely because of their extensive applications in different industries, medical science. Also they are good candidate as interconnectors in electronic devices and electrical contacts in high temperature superconductor (HTSC) thin film devices [1].

Thin films properties strongly depend on their microstructure parameters such as preferred orientations, grain size, stress and strain values which have important roles in design, production and reliability of different devices.

Thin films deposition usually accompanied with residual stress [2]. The stress produced during film growth is an important factor affect on thin film operation [3]. On the other hand residual stress in thin films is a crucial challenge in design and fabrication of micro-electromechanical systems since increasing residual stress results in mechanical defects in thin films such as creating cracks and reducing adhesion of thin film with the substrate [4-5]. Therefore existing high stress in thin films may affect on the optical, electrical and magnetic properties of them [6].

Precise calculations of residual stresses with analytical methods are difficult, thus these stresses usually measure using experimental methods. Most of the methods are destructive. X-ray diffraction technique is the main non-destructive method for residual stress measurement. XRD methods use the distance between crystallographic planes (d-spacing) as a strain gauge. Stress can be calculated from comparison between d-spacing of strained and non-strained thin films. Some post deposition process can change the residual stress of thin films and improve the structural and mechanical properties of thin films. Annealing thin films at the high temperature is one of these methods. Heating treatment supplies energy to rearrangement of grains. High temperature X-Ray diffraction (HT-XRD) is an important instrument to study the structural changes at high temperatures, thermal behavior, and crystallization processes.

DC magnetron sputtering is a standard method for Ag deposition on glass substrates, because of its high deposition rate and ability to control parameters and characteristics of layer such as thickness and grain size to produce uniform films with high mechanical quality [7-10]. Also glass due to its characteristic features such as low porosity and uniform surface is a good choice as substrate in the most deposition methods which performed at low temperature [11].

In this study, Ag thin film with 80nm thickness was grown on glass substrates using DC magnetron sputtering at room temperature. Evolution of structure properties during HTK process has been measured using in-situ HT-XRD instrument. The residual stresses of as-deposited and post annealed have been measured to investigate the impact of high temperature on the residual stress of thin film.

Experimental

Silver thin films were deposited onto glass substrates using DC magnetron sputtering system. The pri-vacuum pressure of the chamber and the Ar gas pressure were respectively 10^{-5} and 3×10^{-2}mbar. The Ag (99.999%) target with 12.5 cm diameter and 3 mm thickness was placed at a distance of 12 cm from the substrate. Sputtering power was kept in 140 W resulted in deposition rate of 1 A^0/s. The thickness of thin film was 80nm.

In order to study the structure and characterization of Ag thin film X-ray diffraction pattern of Ag thin film were measured with Philips diffractometer using Cu-Kα radiation with 1.54 A^0 wavelength at 40kv voltage and 30mA current. To evaluate the effect of temperature on the structure of Ag thin film grown on the glass, an in-situ HT-XRD has been used. The thin film heating process was started from room temperature (25^0C) then the temperature was increased to 300^0C and 450^0C in defined steps. Cooling process have been done in the same way from 450^0C to 300^0C and returned to the room temperature (25^0C). X-ray diffraction pattern of Ag thin film during HTK process was measured at each temperature.

The residual stresses of as-deposited and post annealed Ag thin film were measured using $\mathrm{Sin}^2\psi$ method with X-ray diffractometer. Since the residual stress in thin films is strongly influenced by micro structure, calculation of the size of crystalline grains is important. The crystallite size (D) was estimated using the Scherrer formula [12]:

$$D = \frac{K\lambda}{\beta \cos\theta}$$

where β is the full width at half maximum (FWHM) of the XRD peak, K is constant (K= 0.9), θ is the diffraction angle and λ is the X-ray wavelength corresponding to CuKα radiation (λ= 1.54 A^0).

Result and discussion

Fig.1 shows in-situ X-ray pattern of Ag thin film before, during and after applying high temperature. At heating way, temperature increased stepwise from room temperature (25^0C) to 300^0C and 450^0C. At cooling way, the temperature decreased step by step from 450^0C to 300^0C and room temperature (25^0C). Ag peaks are indicated. Since glass was used as an amorphous substrate, no extra peak detected. The XRD pattern of 25^0C increasing temperature shows that as-deposited Ag film has several grains related to (111), (200), (220), (311) and (222) planes. The (111) peak are relatively sharp. When temperature increased to 300^0C and 450^0C, Ag peaks were heightened. At cooling process, during temperature decreasing from 450^0C to 300^0C the intensity of Ag peaks had no changes, but decreasing temperature to 25^0C leads to appearing stronger Ag peaks. At this point although the intensity of all Ag peaks were increased, but Ag(111) grains had the most enhancement. A little shift in the place of Ag peaks occurred, which related to the uniform strain due to applying high temperature.

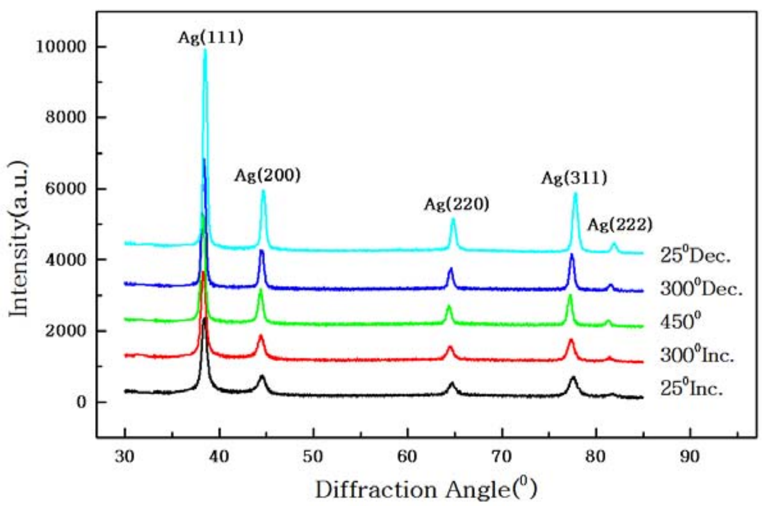

Fig.1: In-situ X-ray diffraction pattern of Ag thin films during HTK process, at increasing from the room temperature and decreasing to the room temperature.

To access detailed information about the effect of HTK process on the grain size of Ag thin film, the sizes of different Ag grains were estimated using Scherrer formula. Table 1 shows the sizes of (111), (200), (220), (311) and (222) grains during heating and cooling ways of HTK process. With comparison of first and last columns related to as-deposited (25^0C increasing) and post annealed (25^0C decreasing) thin film it is indicated that totally the size of all Ag grains increased due to the thermal treatment, but size enhancement of (111) grains is more than the other grains. The second place of size enhancement belongs to (220) grains. It is well known that Ag thin films have the face centered cubic (FCC) structure. In FCC metals, two planes are important: (111) and (220), which (111) planes have minimum surface energy and (220) planes have minimum strain energy. Competition between these planes defines preferred orientation of FCC metals. Here, although the size of (111) and (220) grains are more than the other planes but the size of (111) grains are larger than (220) grains.

It seems thorough applying high temperature, (111) Ag grains gain enough energy to compete with (220) planes and reach to minimum surface energy.

Table1: the sizes of (111), (200), (220), (311) and (222) grains during heating and cooling ways of HTK process.

Temperture(^{0}C) / D(nm)	25^{0}C Inc.	300^{0}C Inc.	450^{0}C	300^{0}C Dec.	25^{0}C Dec.
D(111)	**16.49**	**54.37**	**50.62**	**35.81**	**73.35**
D(200)	**13.47**	**31.84**	**45.37**	**45.36**	**18.28**
D(220)	**34.9**	**17.05**	**24.44**	**16.4**	**60.83**
D(311)	**14.45**	**17.21**	**26.49**	**15.18**	**18.55**
D(222)	**13.68**	**22.02**	**18.27**	**18.31**	**22.11**

Table 2 displays measured residual stress of an as-deposited and post annealed Ag thin film sputtered on glass substrate with 80nm of thickness. Applying thermal treatment up to 450^{0}C resulted in decreasing of residual stress. Due to high temperature, a part of stress which caused by deposition process in Ag thin film was relaxed. Simultaneous consideration of Table 1 and Table 2 and Fig. 1 revealed that decreasing residual stress in Ag thin film encouraged Ag grains to orient along (111) planes. High intensity of Ag(111) implys the high compact of Ag grains along the (111) direction. Although the crystallite size of (220), (200) and (311) Ag grains of post annealed thin film increased but high compact of (111) grains shows that thermal treatment empowered Ag (111) grains and allowed Ag thin film to have the (111) direction as its preferred orientation.

Table2: Measured residual stress of as-deposited and post annealed Ag thin film with 80nm thickness sputtered on glass substrate.

	Before HTK Process	After HTK Process
Stress(GPa)	**2.1**	**1.1**

Conclusion

Ag thin film with 80nm thickness grown on the glass substrate has been structurally characterized using XRD instrument. To investigate the effect of high temperature on the microstructure of Ag thin film, an in-situ HT-XRD has been employed. It was found that applying high temperature up to 450^{0}C yield to enhancement of Ag(111) grains growth. Calculating the several Ag grains size and measuring the residual stress revealed that surface energy plays an important role to control the preferred orientation of Ag thin film. X-ray pattern of HTK process confirmed that Ag (111) grains have been strengthened and compacted after applying thermal treatment.

References

[1] D. Winau, R. Koch, M. Weber, K.-H. Rieder, Appl. Phys. Lett. 61, 279 (1992).

[2] A. G. Gómez, A.A.C. Recco, L. G. Martinez, Activity Report LNLS (2007).

[3] B.W. Sheldon, K. H. A. Lau, A. Rajamani, J. Appl. Phys. 90, 5097 (2001).

[4] E. Klokholm, IBM J. Res. Develop. 58531 (1987).

[5] M. D. Drory, M. D. Thouless, Acta Metal 36, 2019 (1988).

[6] H. Awano and T. Sato, Japan. J. Appl. Phys. 27, L880 (1988).

[7] M. Madou, Fundamentals of Microfabrication, CRC Press, Boca Raton, New York, 1997.

[8] D.W. Hoffman, J.A. Thornton, Thin Solid Films 40, 355 (1977).

[9] J.A. Thornton, J. Tabock, D.W. Hoffman, Thin Solid Films 64 (1979).

[10] D.W. Hoffman, J.A. Thornton, J. Vac. Sci. Technol. 20355 (1982).

[11] H. Savaloni, Surface science in nanotechnology, Universities Central Publishing Company, Tehran, Iran (in Persian), Vol. 1 (2004).

[12] T.C. Huang, G. Lim, F.Parmigiani, E. Kak , J. Vac.Sci. Technol. A, 3 (1985).