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Effects of parameters on the performance of amorphous IGZO thin films prepared by RF magnetron sputtering

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Amorphous indium-gallium-zinc oxide (IGZO) transparent conductive thin films are prepared on glass substrates by radio frequency (RF) magnetron sputtering. The effects of seven factors, which are substrate temperature, sputtering atmosphere, working pressure, sputtering power, annealing temperature, negative bias voltage and sputtering time, on Hall mobility, transmittance and surface roughness are studied through orthogonal experiments. The results show that the effects of working pressure, substrate temperature and sputtering atmosphere on performance of films are the most prominent. According to the experimental results and discussion, relatively reasonable process parameters are obtained, which are working pressure of 0.35 Pa, substrate temperature of 200 °C, sputtering atmosphere of Ar, sputtering power of 125 W, sputtering time of 30 min, negative bias voltage of 0 V and annealing temperature of 300 °C.

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Thin film transistor liquid crystal display (TFT-LCD), which has been widely used in large scale integrated circuits and display panels, is an important display technology[1]. With the rapid development of information technology and display technology, the higher requirements are raised, such as faster response, higher brightness, lower energy consumption, lower cost and transparent and flexible display[2]. Various materials were applied to the TFT-LCD as the channel layer, such as organic semiconductors[3], hydrogenated amorphous silicon (a-Si:H)[4,5] and metal oxide semiconductors[6]. Nowadays, traditional TFT channel material cannot meet the requirements of the increasing LCD size and the driving frequency in a circuit. Therefore, new materials for the TFT channel layer have been investigated intensively. Amorphous indium-gallium-zinc-oxide (a-IGZO) is an n-type semiconductor with band width of 3.5 eV, and currently it has become a hot research field of field-effect transistors[7]. In 2004, by doping In_2O_3 and GaO with high concentrations in ZnO, Nomura et al[8] prepared a-IGZO transparent conductive film on a plastic substrate, and its mobility is over 10 $cm^2V^{-1}s^{-1}$. These semiconductor materials can be used in many types of flexible, light-weight and impact-resistant electronic devices, including flexible displays, electronic papers and durable computers. Since then, the a-IGZO transparent conductive film and the TFT using it as channel layer have drawn the increasing attention, and the a-IGZO transparent conductive films and TFT prepared by different fabrication processes have also been reported[9,10].

In this paper, a-IGZO transparent conductive films are prepared on the glass substrates by radio frequency (RF) magnetron sputtering, and the effects of seven factors on the Hall mobility, transmittance and surface roughness are studied through orthogonal experiments.

The IGZO sputtering target was prepared by mixing In_2O_3 (AR, 99.99%), Ga_2O_3 (AR, 99.99%) and ZnO (AR, 99.99%) powders with molar ratio of 1:1:2 and then compressed at room temperature to form a disc. The compressed disc was then sintered for 12 h at 1250 °C [11]. The diameter of the target is 50 mm.

Corning Eagle 2000 non-alkali glass with main compositions of SiO_2, Al_2O_3, B_2O_3 and BaO was used as substrate, and was ultrasonically cleaned every 10 min by acetone, deionized water and alcohol in turn. Then the substrates were put into a vacuum oven.

The a-IGZO thin films were prepared on the glass substrates using JPC-200 RF magnetron sputtering system. High-purity argon and oxygen were introduced into the sputtering chamber with working pressure from 0.30 Pa to 1.0 Pa, substrate temperature from 25 °C to 300 °C, sputtering power from 20 W to 150 W and negative bias voltage from 0 V to 200 V. Moreover, the distance between the target and the substrate was 60 mm while the

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base pressure was 1.0×10^{-4} Pa.

Orthogonal experimental design uses a ready-made normalized form of orthogonal table to scientifically select experimental conditions and reasonably arrange the experiments[12]. It is a method for research on problem of multi-factors and multi-levels. It needs to pick out part of the representative points from the full-scale experiments according to orthogonality. Using the mathematical statistic principles, we can analyze the effect degree of each factor.

Factors affecting the performance of a-IGZO thin films include substrate temperature, working pressure, sputtering atmosphere and so on. Taking together, we select seven factors of substrate temperature, sputtering atmosphere, working pressure, sputtering power, annealing temperature, negative bias voltage and sputtering time, and they are respectively represented as A, B, C, D, E ,F and G. Substrate temperature is taken as two levels of 25 °C and 200 °C, sputtering atmosphere is taken as three levels of 100%Ar+0%O_2, 100%Ar+5%O_2 and 100%Ar+10%O_2, working pressure is taken as three levels of 0.35 Pa, 0.5 Pa and 0.7 Pa, sputtering power is taken as three levels of 75 W, 100 W and 125 W, annealing temperature is taken as three levels of 25 °C, 200 °C and 300 °C, negative bias voltage is taken as three levels of 0 V, 30 V and 60 V, and sputtering time is taken as three levels of 20 min, 30 min and 40 min.

The Hall mobility, transmittance and surface roughness of the thin films were respectively measured by Van-der-Pauw method, 722 grating spectrophotometer and atomic force microscopy, respectively.

Because the numbers of levels for factors in the orthogonal table are not equal, the range value needs to be modified[12], and the modified formula is

$$R' = R \cdot d\sqrt{n} , \quad (1)$$

where R is original range value, R' is modified range value, d is discount coefficient, and n is experimental times of each level.

X-ray diffraction (XRD) pattern of the IGZO thin film is shown in Fig.1. It can be seen that the IGZO thin film prepared by RF magnetron sputtering is obviously amorphous with a diffraction amorphous envelope at 20°–30° and there is no other obvious characteristic peak. It matches the XRD pattern of the a-IGZO thin film prepared by Hosono[13].

The morphology of a-IGZO film is obtained by the scanning electron microscopy (SEM). The surface of the film shown in Fig.2(a) is very smooth, and there is no significant bump structure on the fracture shown in Fig.2(b). In addition, as can be seen from the section morphology of the film, the thickness of the film is about 300 nm. It is emphasized that there is no significant difference in the prepared thin films.

Under each level of the factors, the average response values of Hall mobility, transmittance and surface roughness are obtained by calculating and analyzing the measured data. And then the variation trends of Hall mobility, transmittance and surface roughness along with the change of level of each factor are obtained, respectively. According to the calculated range values, the effect degree of each factor is analyzed. The larger the range value, the more significant the effect is.

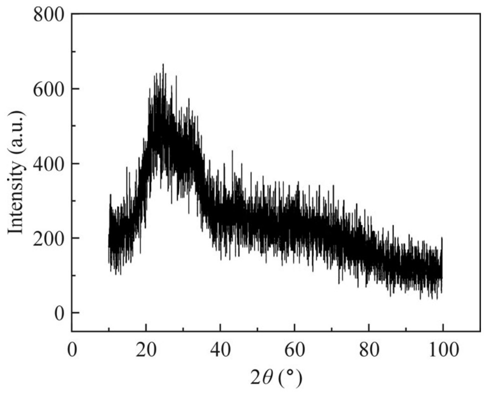
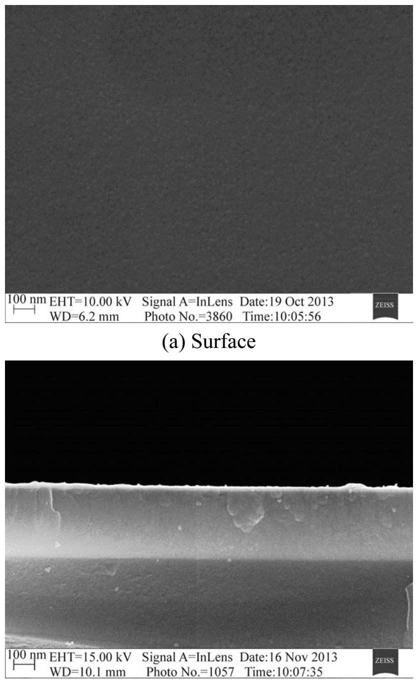


Fig.1 X-ray diffraction pattern of the prepared a-IGZO thin film



(a) Surface

(b) Fracture

Fig.2 SEM micrographs of the a-IGZO thin film

The range values of the Hall mobility under the effects of different factors are shown in Tab.1. According to the range values, the primary and secondary orders of the factors affecting the Hall mobility are concluded, which are substrate temperature, sputtering atmosphere, annealing temperature, working pressure, sputtering power, sputtering time and negative bias voltage.

Fig.3 shows the variation trends of Hall mobility with the changes of different factors. It can be seen that when the substrate temperature is 200 °C, the Hall mobility of the films is increased significantly compared with that at 25 °C, which means that the higher deposition temperature of the films is helpful to get higher Hall mobility, because the higher substrate temperature is propitious to the diffusion ability of deposited atom and a more complete crystal can be achieved. The Hall mobility decreases significantly with the increase of O_2, which is in accord with the result reported by Hisato et al[14]. It may be attributed to that O_2 makes more defects which reduce the Hall mobility of the films. The maximum Hall mobility occurs when the sputtering atmosphere is pure argon. The Hall mobility of the annealed film is higher than that of film without annealing, but it decreases with the further increase of the annealing temperature. The film annealed at 200 °C has higher mobility. With the increase of working pressure, the Hall mobility has declining trend, which may be attributed to that under higher pressure, gas molecules and ions are so many that the probability of generating impurities increases. Hall mobility increases with the increase of sputtering power, and the maximum Hall mobility is obtained when sputtering power is 125 W. The sputtering time principally affects the thickness of the films. The longer the sputtering time is, the thicker the film is. The Hall mobility is relatively large when the sputtering time is 30 min, which indicates that the moderate film thickness can get larger Hall mobility. Negative bias voltage has little effect on Hall mobility.

Tab.1 The range values of Hall mobility under different factors

Range value	A	B	C	D	E	F	G
R ($cm^2V^{-1}s^{-1}$)	7.05	7.07	2.81	2.33	3.39	0.33	1.25
R' ($cm^2V^{-1}s^{-1}$)	6.76	6.41	2.55	2.11	3.08	0.30	1.14

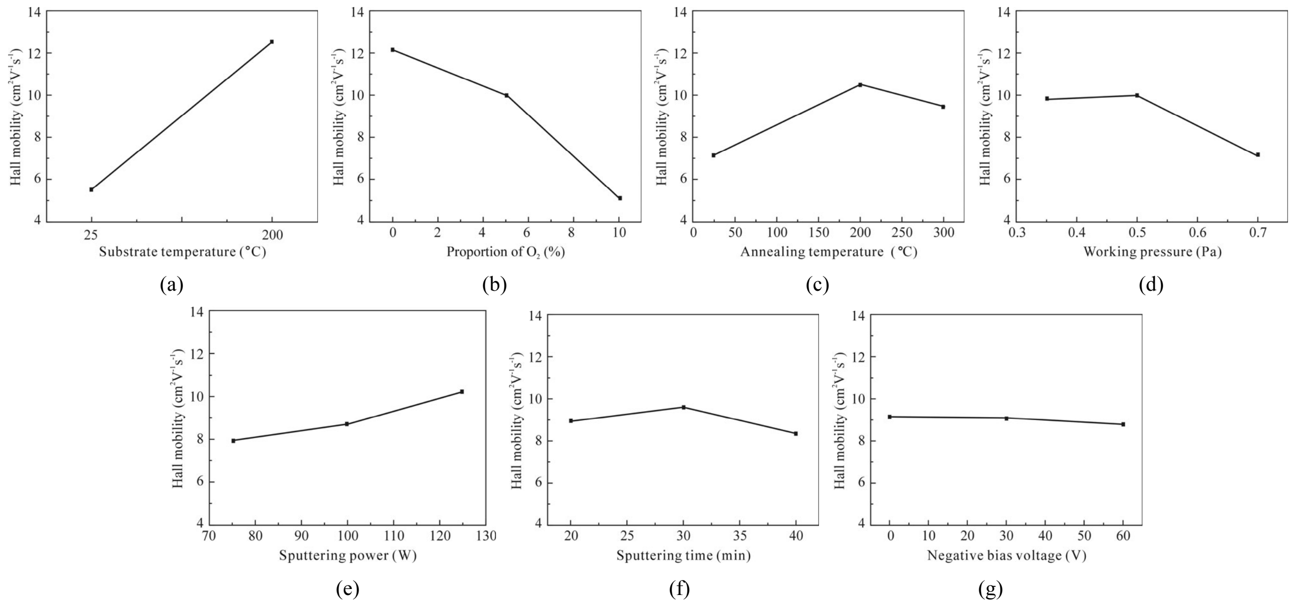


Fig.3 Variation trends of Hall mobility with the changes of different factors

The range values of the transmittance under the effects of different factors are shown in Tab.2. According to the range values, the primary and secondary orders of the factors affecting the transmittance are concluded.

Tab.2 The range values of transmittance under different factors

Range value	A	B	C	D	E	F	G
R (%)	2.17	0.68	2.83	2.15	1.88	2.04	0.95
R' (%)	2.08	0.62	2.57	1.95	1.70	1.85	0.86

Fig.4 shows the variation trends of transmittance with the changes of different factors. As shown in Fig.4(a), the transmittance of the films increases obviously with the increase of working pressure, and the maximum transmittance occurs at 0.7 Pa. When the substrate temperature is 25 °C, the transmittance of the film is higher than that at 200 °C. For the polycrystalline transparent conductive film, the increase of substrate temperature reduces the crystal defects so as to improve the transmittance, which may not work for the a-IGZO thin film. With the increase of sputtering power, the transmittance has a decreasing trend. It may be that the greater the sputtering power is, the faster the deposition rate is, and the smaller the range of short-range order is, which has a weakening effect on the transmittance. Along with the increase of negative bias voltage, the transmittance first decreases and then increases, but on the whole, it reduces. When the negative bias voltage is 0 V, the transmittance is the largest. The annealing temperature, sputtering time and sputtering atmosphere have little effect on transmittance.

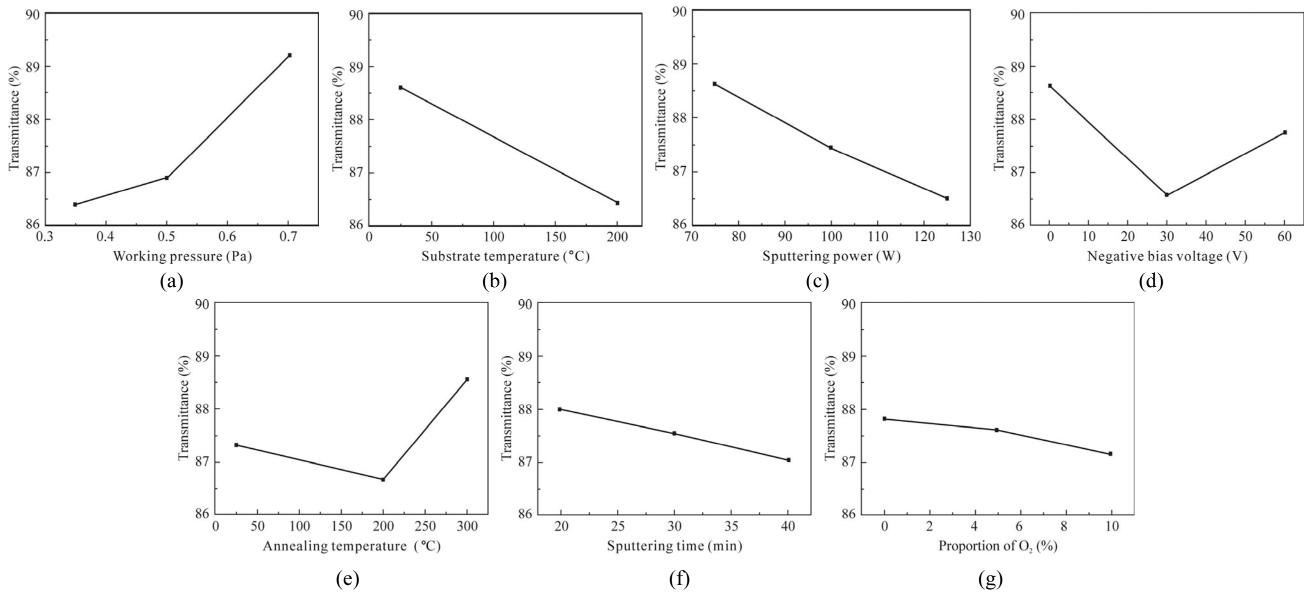


Fig.4 Variation trends of transmittance with the changes of different factors

The range values of the surface roughness under the effects of different factors are shown in Tab.3. According to the range values, the primary and secondary orders of the factors affecting the surface roughness are concluded.

Tab.3 The range values of surface roughness under different factors

	A	B	C	D	E	F	G
R (nm)	0.225	0.413	0.451	0.159	0.317	0.168	0.314
R' (nm)	0.216	0.374	0.409	0.144	0.287	0.152	0.284

Fig.5 shows the variation trends of surface roughness with the changes of different factors. As shown in Fig.5(a), with the increase of working pressure, the surface roughness of the films increases obviously, and it is the lowest when the pressure is 0.35 Pa. With adding O_2 in the sputtering atmosphere, the surface roughness decreases obviously, but increases with a further increase of O_2. But on the whole, the surface roughness falls after adding O_2. When the proportion of O_2 is 5%, the surface roughness is the minimum. The surface roughness of the films is relatively low when the annealing temperature is 200 °C, but with the further increasing of annealing temperature, it will increase. The roughness increases with the increase of sputtering time, and it indicates that roughness increases along with the increase of thickness. When the substrate temperature is 200 °C, the surface roughness of the films is relatively low, which indicates that the higher substrate temperature is helpful to smooth the film surface. The negative bias voltage and sputtering power have little effect on surface roughness.

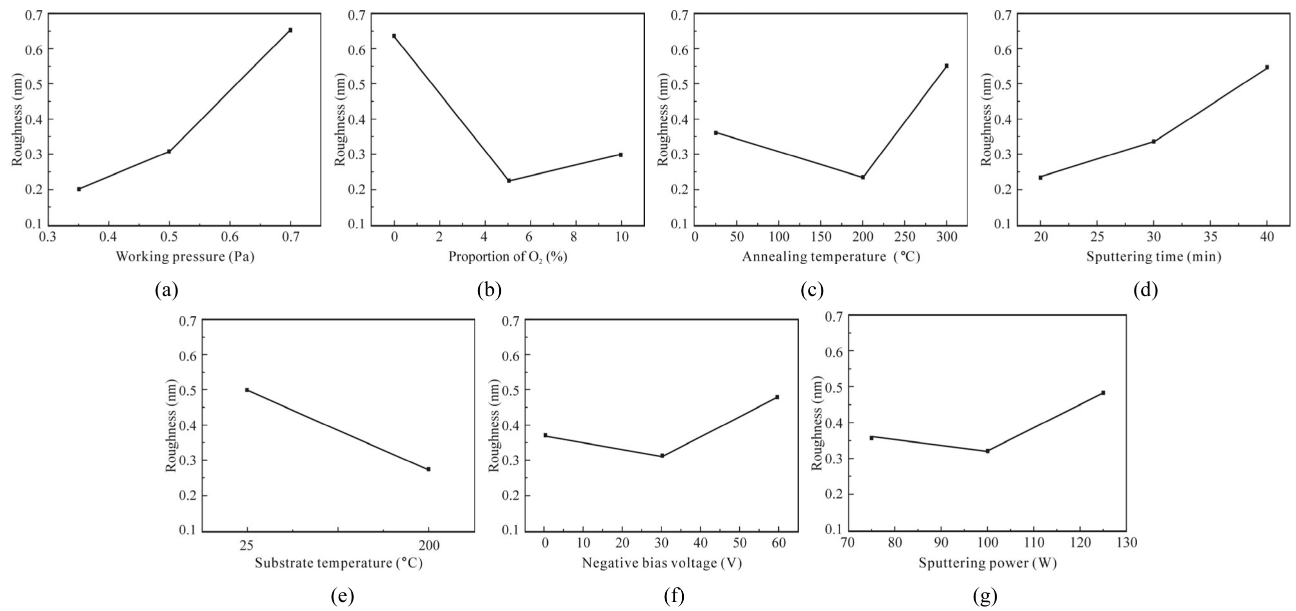


Fig.5 Variation trends of surface roughness with the changes of different factors

In conclusion, the effects of working pressure, substrate temperature and sputtering atmosphere on the performance of the films are more prominent. With the increase of the working pressure, the Hall mobility declines overall, while the roughness and transmittance increase. Hence, when the working pressure is relatively low, the performance of the thin films is better. Considering the heat resistant ability of flexible substrate, we need to balance the substrate temperature and the performance of the films, and select the temperature at which substrate can withstand in the preparation of a-IGZO thin films. With the increase of the proportion of O_2 in the sputtering atmosphere, the Hall mobility and transmittance decline, and the roughness also declines overall. So, on the whole, the effect of O_2 is detrimental to the performance of the films. In addition, with the increase of annealing temperature, Hall mobility, transmittance and surface roughness increase overall. Just as the substrate temperature, annealing temperature should be balanced with heat resistant ability of the substrate. With the increase of the sputtering power, the transmittance declines, while the Hall mobility and roughness increase. The negative bias voltage and sputtering time have little effect on the performance of the films.

According to the experimental results and discussion, the relatively reasonable process parameters are working pressure of 0.35 Pa, substrate temperature of 200 °C, sputtering atmosphere of Ar, sputtering power of 125 W, sputtering time of 30 min, negative bias voltage of 0 V and annealing temperature of 300 °C.

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