Effects of Ag-doping and oxygen partial pressure on the physical properties of the $Mg(In_{2-x}Ag_x)O_4$ system

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Abstract

Ag-doping tansparent conductor $Mg(In_{2-x}Ag_x)O_4$ (x = 0.0 - 0.5) oxides with a space group Fd$\bar{3}$m are grown by a usual solid state reaction technique. The addition of Ag lowers the growth temperature and reduces the growth time. However, the resistivity increases following Ag doping. Post annealing in hydrogen gas increases the grain size and the connections among grains.

Key words: transparent conducting oxide, indium oxide, $MgIn_{2-x}O_4$

Transparent conductive oxides, such as tin-doped indium oxide (ITO), are widely used in optoelectronic devices. These materials exhibit high optical transmission in the visible range and electrical conductivities comparable to those of metals. Recently magnesium indium oxide, $MgIn_{2-x}O_4$, was found to have similar transparent conductive properties and could be used as an alternative for ITO in the future [1-4]. This work discusses the effects of Ag doping, oxygen pressure and hydrogen annealing on the synthesis and physical properties of the $Mg(In_{2-x}Ag_x)O_4$ compounds.

A series of nominal composition $Mg(In_{2-x}Ag_x)O_4$ oxides were prepared from high-purity MgO, In_2O_3 and Ag_2O powders by the standard solid-state reaction technique. The powders were ground, calcined, sintered, pressed into pellets, then heated in a tube furnace at 1400 ^{o}C in air for 72 h. The prepared pellets exhibit a single-phase $MgIn_2O_4$ structure with a space group Fd$\bar{3}$m, as revealed by the X-ray diffraction (XRD) analysis shown in Fig. 1. If the sintered temperature was decreased to 1350 ^{o}C, XRD patterns showed that the In_2O_3 phase remained in the sample in addition to the original $MgIn_2O_4$ phase. It indicats incomplete solid-state reactions. By adding a small amount of Ag to the $MgIn_2O_4$ phase, the growth temperature was greatly reduced. Fig. 2 illustrates that as some In was replaced by Ag, the $Mg(In_{2-x}Ag_x)O_4$ single phase can be obtained at 1300 ^{o}C; in addition, the heating time was also reduced to 6 h. The XRD pattern revealed that the Ag-doped sample $Mg(In_{2-x}Ag_x)O_4$ has the single phase structure identical to that of the parent $MgIn_2O_4$ oxide.

The addition of Ag increases the resistance of $Mg(In_{2-x}Ag_x)O_4$ series. Fig. 3 displays the variation of resistivity ρ, measured by a four-point method, with Ag content x. The resistivity exhibits a monotonically increase with x. The electrical resistivity is also strongly influenced by oxygen partial pressure during the growth process. We prepared samples grew under different ambient atmosphere. The sample grown in pure O_2 gas has the largest ρ of 52.8 Ω-cm; the sample in air has ρ of 11.4 Ω-cm and the one in Ar gas has the lowest ρ of 0.13 Ω-cm. The resistivity of the sample annealed in Ar gas is about two orders of magnitude smaller than the one in O_2. It indicats that high oxygen content reduces the electrical conductivity of the sample and suggests electron-type carriers in the system. Future Hall measurements can verify

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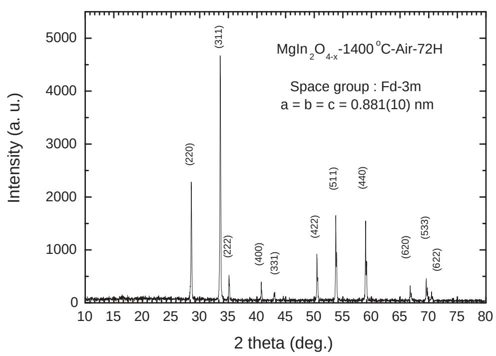

Fig. 1. X-ray diffraction pattern of the pure $MgIn_2O_4$ phase.

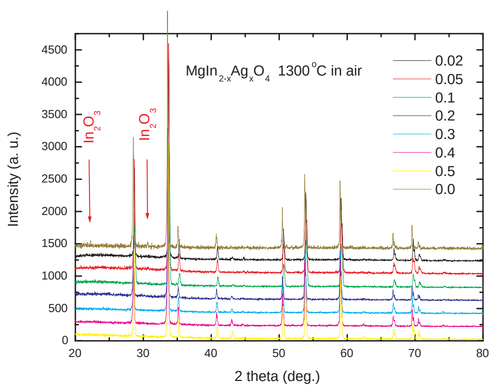

Fig. 2. X-ray diffraction patterns of $Mg(In_{2-x}Ag_x)O_4$ series heated at 1300^{o}C for 6 h in air. For parent compound of x = 0.0, there appeared a small amount of the In_2O_3 phase.

this interpretation.

Post annealing in flowing 10%H_2/Ar gas has a positive effect on the crystalline quality of the sample. Figs. 4 and 5 show the scanning electron microscopy (SEM) graphs of the samples before and after the H_2 reduced gas treatment at 300 ^{o}C for 3 h. The grain sizes exhibit substantial increase after the annealing treatment. Better connectivity among grains is also observed in the micrographs. These results reveal that H_2 reducing treatment improves both the electrical and the structural properties of the samples.

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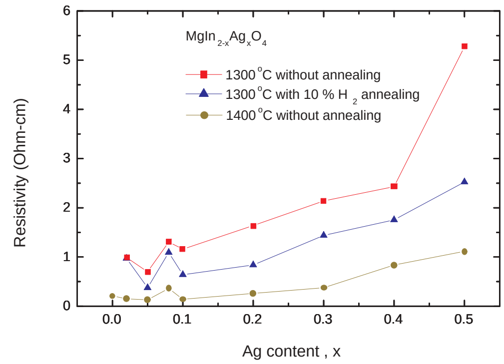

Fig. 3. Variaction of the room-temperature electrical resistivity of $Mg(In_{2-x}Ag_x)O_4$ oxides with Ag content.

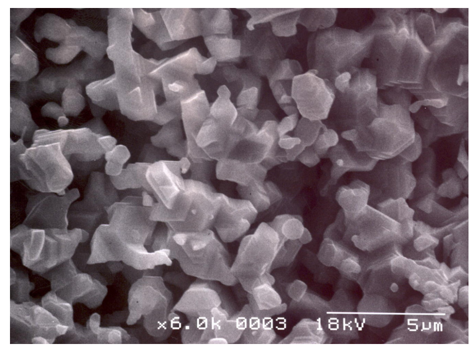

Fig. 4. Scanning electron micrograph of as-sintered $Mg(In_{1.92}Ag_{0.08})O_4$ oxides heated at 1300^{o}C for 6 h in air.

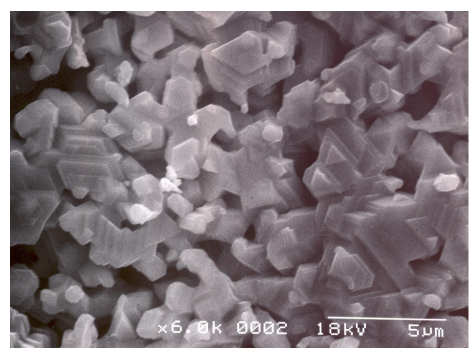

Fig. 5. SEM graph of the $Mg(In_{1.92}Ag_{0.08})O_4$ sample which was annealed at 300 ^{o}C for 3 h in flowing 10%H_2/Ar gas.

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