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Realization and Characterizations of Conducting Ceramics Based on ZnO Doped by TiO_2, Al_2O_3 and MgO

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Abstract : ZnO with wurtzite structure is a well-known semiconducting oxide (SCO), being applied in thermoelectric devices, varistors, gas sensors, transparent electrodes, solar cells, liquid crystal displays, piezoelectric and electro-optical devices. Intrinsically, ZnO is weakly n-type SCO due to native defects (Zn_j, V_o). However, the substitutional doping by metallic elements as (Al, Ti) gives rise to a high n-type conductivity ensured by donor centers. Under $CO+N_2$ sintering atmosphere, Schottky barriers of ZnO ceramics will be suppressed by lowering the concentration of acceptors at grain boundaries and then inducing a large increase in the Hall mobility, thereby increasing the conductivity. The presented work concerns ZnO based ceramics, which are fabricated with doping by TiO_2 (0.50mol%), Al_2O_3 (0.25mol%) and MgO (1.00mol%) and sintering in different atmospheres (Air (A), N_2 (N), $CO+N_2$(C)). We obtained uniform, dense ceramics with ZnO as the main phase and Zn_2TiO_4 spinel as a secondary and minor phase. An important increase of the conductivity was shown for the samples A, N, and C which were sintered under different atmospheres. The highest conductivity ($\sigma = 1.52\times10^5$ $S\cdot m^{-1}$) was obtained under the reducing atmosphere (CO). The role of doping was investigated with the aim to identify the local environment and valence states of the doping elements. Thus, Electron paramagnetic spectroscopy (EPR) determines the concentration of defects and the effects of charge carriers in ZnO ceramics as a function of the sintering atmospheres. The relation between conductivity and defects concentration shows the opposite behavior between these parameters suggesting that defects act as traps for charge carriers. For Al ions, nuclear magnetic resonance (NMR) technique was used to identify the involved local coordination of these ions. Beyond the six and forth coordinated Al, an additional NMR signature of ZnO based TCO requires analysis taking into account the grain boundaries and the conductivity through the Knight shift effects. From the thermal evolution of the conductivity as a function of the sintering atmosphere, we succeed in defining the conditions to realize ZnO based TCO ceramics with an important thermal coefficient of resistance (TCR) which is promising for electrical safety of devices.

Keywords : ceramics, conductivity, defects, TCO, ZnO
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