Study on Impurity Doped ZnO Terrace-truncated Nanocone Structures Grown by Advanced Spray Pyrolysis Deposition Technique

（課程博士・様式７）（Doctoral qualification by course work, Form 7）

学 位 論 文 要 旨

Abstract of Doctoral Thesis

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論文題目：

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Abstract :

Investigations on semiconductor oxide materials, with high optical transmittance in the visible range and high electrical conductivity (Transparent conductive oxides - TCO), have gained the interest of researchers in last few decades. Due to their wide range of applications, still there is a great demand for new TCO materials. ZnO is known as an interesting semiconductor oxide material with unique physical and chemical properties such as direct wide band gap (3.37 eV), large exciting binding energy at room temperature (~60 meV), high thermal stability, radiation hardness, nontoxicity. In order to enhance the electrical conductivity and optical transparency, ZnO is dope with many impurities. Al and Ga are known as the most successful dopant due to its' compatible ionic and covalent radii with Zn. Few reports were found about the synthesize of ZnO nanocone structured thin films. We have investigated the structural properties of Al doped zinc oxide terrace-truncated nanocone structure which was grown by using advanced spray pyrolysis deposition method. The optimum growth temperature for Al doped ZnO was investigated in the range of 300 to 450 ºC. The major structural differences were found when changing the growth temperature. The optimum growth temperature was decided to be ~400 ºC with 40 nm, 290.10 per µm2 and 80 % of average top diameter, average structure density and optical transparency in the visible range, respectively. Optimum Al doping ratio was found to be 2 at % of Al doping for optimum transparent conductive oxide properties. The spray angle is the crucial factor deciding the nano structure in spray pyrolysis deposition technique. This value was investigated in three ranges, ~15º, ~30º and ~45º. Properties of nanostructure highly depended on spraying angle. An excellent nanostructure with

good separation, lowest number of structural defects was observed in ~15º of spraying angle. The differences of horizontal and vertical velocities of atomized particles in each spraying angle, were correspond to these structural changes. The lowest resistivity of 1.06×10^{-4} Ω.cm and highest optical transparency of 80.3 % in the visible range were observed in Al doped ZnO sample, which was prepared at ~15º of spraying angle. However, the nanostructure was not clearly identified up to this stage. The spraying volume was increased from 100, 200 and 400 ml to identify the nano structure. The terraced truncated nanocone structure was confirmed at this stage. Due to the higher growth rate along the (002) direction increased the decay rate along the same direction and end up with terrace truncated nanostructure. Even though the top was decaying, the bottom of the nanostructure was narrow as the polar O2- face tightly attached to the nucleation sites of the FTO glass substrate.

We have conducted research on Ga doped ZnO nanostructure using advanced spray pyrolysis method. The growth temperature was changed from 300 to 450 ºC and an excellent nanostructure of Ga doped ZnO was found in 400 ºC of growth temperature with 22.9 nm of average top diameter and 366 per µm2 of average nanostructure density. Ga doping level was varied from 1 to 3 at %. The optimum nanostructure was found when sample was doped with 2 at % of Ga doping, average top diameter and average structure density were 37.0 nm and 230.4 per µm2, respectively. All the Ga doped ZnO samples with varied Ga to Zn ratio, show high optical transmittance in the visible range. The spray angle dependence for the nanostructure growth was investigated in three different angles of ~15º, ~30º and ~45º. The ideal transparent conductive oxide properties with 82 % of optical transmittance in the visible range and 3.9×10^{-4} Ω.cm of low resistivity were grasped at ~15º of spraying angle. The average top diameter, average structure density and height were 22.8 nm, 195 per µm2 and ~200 nm, respectively. The terrace-truncated nanocone structure was not confirmed until this stage. The spraying volume was changed 100, 200 and 400 ml to identify the nanostructure. Heights of the nanostructure were 200, ~380 and ~700 nm for 100, 200, and 400 ml volumes, respectively. The terraced truncated nanocone structure was found to be formed at this stage by FE-SEM images.