The Synthesis, Characterizations and Application of One-Dimensional Nanostructured Fluorine Doped Tin Oxide Thin Films

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学位論文要旨

Abstract of Doctoral Thesis

専　　攻：
Course：Optoelectronics and Nanostructure Science

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論文題目：
Title of Thesis：The Synthesis, Characterization and Application of One-Dimensional Nanostructured Fluorine Doped Tin Oxide Thin Films

論文要旨：
Abstract：Transparent conducting oxides (TCO) materials have become technologically very important class of materials due to optically transparent and electrical conducting properties at the same time. An advanced version of spray pyrolysis technique which is known as rotational, pulsed and atomized spray pyrolysis (RPASP) was developed and deployed to fabricate One-dimensional (1-D) nanostructured fluorine doped tin oxides (FTO) thin films. This technique is versatile and deployed to prepare thin films of various nanotechnological architectures of fluorine-doped tin (IV) oxide (FTO) layers on glass surfaces. The different spray parameters have been controlled in order to fabricate FTO nanoparticles, cross-link nanorods, vertically aligned nanorods, hollow nanorods and needle type 1D nanostructured thin films on glass substrate.

RPASP was shown as a viable, and attractive, deposition technique for the synthesis of fluorine doped tin oxides one- dimensional nanostructured thin films, which allows for the perfect controllable spray directions for the fabricate vertically aligned FTO nanorods. Spraying at low angle to the substrate is mandatory for the growth of of vertically aligned nanorods. Spraying at low angle or horizontal to the substrate facilitate to sweep the aerosol spray of the fine atomized particles on the glass substrate and as a result density between the particles become high. Therefore, FTO crystal tends to grow along vertical direction with the pyrolysis process at higher substrate temperature. But, vertical or higher angle spraying of the SPD technique only facilitate to synthesize thin films consist with FTO nanoparticles. The preferential orientation of nanorods crystallites along the (101) direction and prepared nanorods thin film showed an excellent transparency of 84.8% and a low resistance of 26.7 Ω/sq.

This technique allows for the perfect control of morphology of nanotechnological architectures of FTO, which can be achieved simply by controlling spray duration. As such, 0-D nanocrystallites, 1-D uncapped nanorods and 1-D capped nanorods; all in 2-D thin layers, and extensively cross-linked 3-D nanotechnological architectures of FTO can be prepared, on soda lime glass surfaces. This is the first time study on kinetically-controlled growth of different nanotechnological architectures of FTO, using the same technique.

X-Ray Diffraction (XRD), Scanning Electron Microscopic (SEM) and X-Ray Photoelectron Spectroscopic (XPS) data show excellent correlations. The one-dimensional FTO thin films are crystallized in the tetragonal rutile structure, with a preferential orientation of their crystal along the (101) direction. They exhibit an excellent transparency in the visible range of the electromagnetic spectrum. As evident from XPS data, FTO uncapped nanorods were found to contain more Sn2+ sites on their surfaces when compared to those of other nanotechnological architectures. ATR-FT-IR spectrum shows F-Sn-F and F-Sn stretching vibrations in all forms of FTO nanostructures. Thin layers have optical transmissions in the range 70% to 85%, in the visible range of the electromagnetic spectrum. This technique is versatile and is not limited only to fabricate FTO nanostructures, but, it can also be used to fabricate thin layers of nanotechnological structures of different dimensionalities of various materials on various substrates, which is capable to withstand required pyrolytic temperatures.

The effect of the fluorine concentration to the growth of FTO hollow nanorods at higher spray direction to the substrate was also investigated. The experiment was conducted at higher spray angle to the substrate while controlling the fluorine concentration of the spray precursor solution. The incorporation of fluorine to tin dioxide has resulted to enhance the grain size of FTO thin films and hollow nanorods could be obtained with higher fluorine concentration. The fluorine ions substitute to the oxygen vacancies in the proper lattice position of tin oxide in the initial stage and the crystal size increased with the pyrolysis process. However, high concentration of fluorine affects as the impurities to the crystal structure and it restricted the growth of FTO crystal in horizontal direction but allowing to grow in the vertical direction. As a result, FTO hollow type nanorods has formed with the spray time duration. The fabricated nanorods show cubic structure of the tin dioxide with (111) direction as the preferred orientation. The deposition of sufficient amount of fine atomized FTO particles at fast rate with low spray nozzle distance has coursed to the growth of cubic crystal of tin oxide as it makes high pressure on the substrate. The film morphology, texture and transmittance properties of the thin films are show good interaction with each other.

The effect of the different additives for the crystal growth of FTO 1-D nanostructured thin films was investigated. It clearly shows that the different additives in the precursor solution are effected to improve both verticality and dimension of the FTO nanorods. Vertically align and well separated nanotubes easily fabricate with propanone and ethanol as additives. We suggest that propanone addition plays a role to form vertically align nanorods with (101) preferential orientation while (110) face was the predominant plane of well separated nanorods with ethanol added solution. The thin films prepared by using only water based solution was unable to grow separated 1-D nanostructures and growth of vertically aligned FTO nanorods could not be able to obtained with isopropanol as additives in the spray precursor solution. The conductivity of the 1-D nanostructured thin films also enhanced with commercial FTO glasses as the substrate. However, the preferred crystal growth orientation of the nanorods on commercial FTO glass substrate was along (200) direction due to the effect of the main FTO conducting layer.

The performance of the dye-sensitized solar cells was compared by assembling DSSCs with our 1-D nanostructured FTO thin films and commercially available FTO glasses. It clearly showed that the fill factor and open circuit voltage enhanced with FTO nanorods thin films as the front electrode of the DSSCs. However, the efficiency of the DSSCs with 1-D nanostructures was comparatively low due to the low film thickness of the photoactive electrode, and further development of the DSSCs fabrication technique with FTO nanorods need to be studied.