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ACCEPTED MANUSCRIPT

Environmental stability of solution processed Al-doped ZnO naoparticulate thin films using surface modification technique

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The environmental stability of solution processed Al-doped ZnO (AZO) thin films was enhanced by functionalizing the film surface with a thin self-assembled molecular layer. Functionalization of AZO films was performed using two types of molecules having identical 12-carbon alkyl chain termination but different functional groups: dodecanethiol (DDT) and dodecanoic acid (DDA). Surface modified AZO films were examined using electrical resistivity measurements, contact angle measurements and quantitative nanomechanical property mapping atomic force microscopy. The hydrophobic layer inhibits the penetration of oxygen and water into the AZO's grain boundaries thus significantly increasing the environmental stability over unmodified AZO. Surface modified AZO films using DDT exhibited lower electrical resistivity compared to DDA functionalized AZO films. Our study demonstrates a new approach for improving the physical properties of oxide based nanoparticulate films for device applications.

Keywords: Al-doped ZnO, Nanoparticulate thin films, Solution process, Surface modification, Environmental stability

1. Introduction

Transparent conductive oxides (TCOs) have been extensively applied to flat-panel displays [1], organic light-emitting diodes [2, 3], gas sensors [4] and solar cells [5] as transparent electrodes due to their high electrical conductivity and optical transparency. Making TCOs for these applications through low-cost, high-throughput techniques such as printing would be of great utility. Inks based on TCO nanoparticulate colloids dispersed in proper solvents would be of great advantage for printing TCOs, particularly via direct writing techniques. Direct write techniques, such as spray coating, inkjet/gravure/aerosol jet printing and spin coating, allow for the coating of functional materials on a broad choice of substrate materials and shapes at reasonable cost [6-9]. In particular, doped zinc oxide (ZnO) films are regarded as good substitutes for ITO films especially due to the lower cost and higher abundance of zinc with respect to indium and lower toxicity with respect to antimony [10, 11].

Oxygen vacancies play a vital role in electrical conduction of colloidal and polycrystalline ZnO films [12, 13]. Higher concentrations of oxygen vacancies provide extra electrons as carriers make the AZO films n-type transparent conducting oxides. The amount of charge carriers is controlled by oxygen-vacancy formation via annealing under a reducing atmosphere; in particular, hydrogen can act as both a shallow donor and a defect passivator in ZnO-based

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