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Original research article

Multi-response optimization of ZnO thin films using Grey-Taguchi technique and development of a model using ANN

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ABSTRACT

Spray pyrolysis deposited zinc oxide films have been prepared using zinc acetate precursor in *iso*-propyl alcohol (IPA). Based on deposition parameters, outputs were optimized for multiple performance characteristics such as thickness, roughness, and an optical transmittance of ZnO films using Grey-Taguchi technique. Optimum parameters from Grey relational analysis (GRA) were found to be 0.2 M, 1 bar, 365 °C, and 5 mL min^{-1}. The average thickness, roughness, and an optical transmittance obtained at the optimum condition using GRA were 79 nm, 1.88 nm, and 85.96% respectively as compared to the results obtained at optimized condition by a Taguchi method were 95 nm, 2.45 nm, and 85.19%, respectively. This indicated that the design proposed by Grey relational technique is a feasible approach for determining the optimal parameters. Further, the best condition was predicted by artificial neural network (ANN) and it was observed using cascade forward back propagation algorithm, tangent sigmoid transfer function as a hidden layer, and 4-8-3 topology.

1. Introduction

Zinc oxide (ZnO), indium tin oxide (ITO), and titanium oxide (TiO_2) are typical transparent conducting oxides (TCO) with a great commercial importance [1]. TCO are used for application in light-emitting diodes, piezoelectric transducers, touch panels, liquid crystal displays, display devices, electro-optical devices, conductive gas sensors, solar cells and optoelectronics device [2–6]. ZnO thin films are an attractive TCO because they are inexpensive, non-toxic, high thermal and chemical stability, a wide band gap (3.37 eV), large exciton binding energy (60 meV), high optical transmittance and low electrical resistivity [7,8]. There are several techniques used to prepare ZnO thin films such as sol-gel [9], spray pyrolysis [10], chemical vapor deposition [11], spin coating [12], electro-deposition technique [13] and laser ablation [14]. Spray pyrolysis is simple, versatile and less costly technique and it can be used to deposit films for several applications [15]. The main advantages of spray pyrolysis such as cost effective, easy handling, coating on substrates with complex geometries, uniform and high quality coatings, less temperature requirement, reproducibility make it superior over other techniques. In year 1966, Chamberlin and Skarman have introduced spray pyrolysis for the first time to deposit CdS thin films for photovoltaic application [16]. Since then, the process was used for various materials. Spray pyrolysis for ZnO thin film deposition was first used by Paraguay et al.

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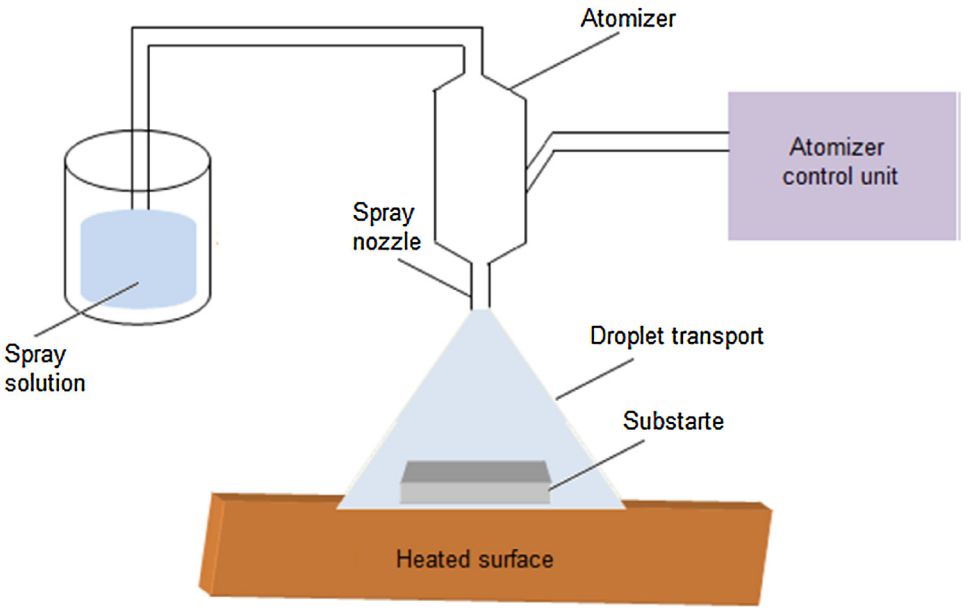

Fig. 1. Schematic diagram of spray pyrolysis.

in year 1999 [17]. The quality of deposited ZnO films is a key factor for its application as TCO. The quality of films directly or indirectly depends on the process parameters [18]. Precursor concentration, substrate temperature, annealing temperature, carrier gas pressure, spray nozzle's distance from the substrate, and flow rate of precursor solution are the main deposition parameters involved with a spray pyrolysis process [19–21].

It is necessary to understand the effect of each important process parameter to identify the optimize condition [22]. It is also necessary to identify the most influencing operating parameter. However, to the best of Author's knowledge, an exhaustive study on simultaneous effect of each of the four major parameters for optimization of output is not yet reported. Most of the study optimized the output by considering one factor at a time method. For example, effect of temperature on output is reported in detail [19,23,24]. Effect of flow rate on output is reported in detail [20,25]. Effect of precursor concentration on output is reported by Boneto et al. [26]. So, there is a great need of such exhaustive study. The general schematic diagram of spray pyrolysis is shown in Fig. 1.

Taguchi and Komesh were developed a statistical method to optimize the output based on input parameters. Optimization of output by taking one factor at a time requires a large number of experiments [27]. Taguchi method is a technique used to identify and optimize those outputs based on process parameters which affects the performance of a process significantly.

This method reduces the number of experiments in a process during robust design [28]. The product with high quality and low cost is the main objective of Taguchi method [29]. In this method, control factors reduce inconsistency of a process and the effect of uncontrollable factors minimizes [30]. A set of experiments is conducted with the help of orthogonal array (OA) to calculate the signal to noise (S/N) ratio. S/N ratio is used to measure the change in response with respect to the target value under various noise conditions [31]. There are three kinds of performance characteristics (larger is better, smaller is better, and nominal is best). Despite the kind of performance characteristics, better is that, which has a larger S/N ratio. So, the combination of the levels with the highest S/N ratio is the set of process parameters that gives optimum result [32]. However, the Taguchi method is not applicable for multiple response optimizations. If more than one response is considered simultaneously, it is possible that the higher S/N ratio for one response may cause a lower S/N ratio for other response. To solve this type of problem, Dr. Deng in the year 1982 proposed a method known as Grey relational analysis (GRA) method [33]. Further, GRA gives a relationship between parameters for multiple factors using fewer data. GRA converts multiple characteristics optimization problem in single characteristics using gray relational grade (GRG) [34]. However, to compare the optimization conditions obtain from single response optimization by Taguchi method with optimum condition obtain from multi-responses optimization by Grey's Technique was carried out in the present study. Similar, studies have already been reported by Kao et al. [35] and Thakker et al. [36]. Artificial neural network (ANN) is used to correlate different factors [37] and to develop models to correlate various performances from a given set of input-output data. The experimental data obtained by Taguchi technique is sufficient and can be used to train the ANN model [38,39]. The aim of present study is to minimize the thickness and roughness and maximize the optical transmittance of ZnO film as the quality characteristic. Taguchi and Grey's techniques have been applied to optimize the outputs based on deposition parameters considering multiple performance characteristics to prepare ZnO films by spray pyrolysis method. The effects of four deposition parameters (precursor concentration, substrate temperature, carrier gas pressure, and solution flow rate) on three performance characteristics (thickness, surface roughness, and optical transmittance of ZnO films) were examined.