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Fabrication and characterization of $Zn_{1-x}Al_xO$ nanoparticles by DC arc plasma

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ABSTRACT

In this paper we have introduced a simple method for the fabrication of aluminum doped zinc oxide (AZO) nanoparticles. The $Zn_{1-x}Al_xO$ nanoparticles with different concentrations of Al (x=0.01, 0.03, 0.06, 0.09, 0.12) were fabricated successfully by this method. The samples were analyzed by the use of several techniques such as SEM, EDX, XRD, PL and UV–vis spectroscopy. The SEM images showed that the fabricated nanoparticles had spherical shapes. The XRD patterns of the samples indicated that the Al atoms substituted in the Zn positions in the crystal lattice of ZnO and there were some changes in the lattice parameters. A blue shift in the λ_{max} of the absorption and a red shift in the λ_{max} of the emission were observed. The results also indicated that the amount of shifts had a direct relationship with the changes in the lattice parameters.

1. Introduction

Semiconductors nanoparticles have increasingly attracted attention because of their applicability in different devices. Zinc oxide is a semiconductor with unique properties. The hexagonal wurtzite structure of the zinc oxide nanoparticles with the lack of symmetry combined with the large electromechanical coupling causes strong piezoelectric and pyroeclectic properties of ZnO [1]. Furthermore, wide band gap and large exciton binding energy of ZnO make it suitable for different purposes in optics [2].

In recent years, there have been some efforts to modify the chemical and physical properties of zinc oxide nanoparticles by doping with other elements. There is a wide variety of elements such as tin [3], indium [4], aluminum [5], cadmium [6], copper [7], sulfur [8], etc, which have been used as a dopant by researchers to modify the properties of the zinc oxide. Optical properties, band gap changing, catalytic properties, making changes in lattice parameters and grains' size are the properties that have drawn more attention among others.

Doping of ZnO with aluminum could cause several improvements in the optical and electrical properties of the zinc oxide. The aluminum doped zinc oxide (AZO) is a good candidate to be used as a transparent electrode in optical devices. The AZO is advantageous to materials such as indium tin oxide (ITO) because of its low cost [9].

Among different fabrication methods of the AZO, the chemical based methods have the advantages of simplicity and cheapness but suffer from unwanted impurities and the presence of solvents' trace in the final products. Moreover, because of the equilibrium conditions, there was almost a limitation in the amount of dopant which could be inserted in the crystal lattice of ZnO.

In developing our previous work of the fabrication of zinc oxide nanoparticles by DC arc [10], we have introduced a simple method of fabricating the aluminum doped zinc oxide nanoparticles in the present study. The method has the advantages of simplicity and the ability of fabricating high purity products. As this method uses a non-equilibrium condition for the fabrication of nanoparticles, there is a possibility for samples with higher concentrations of impurities to be fabricated.

In principle, the method is based on ionizing the elements which should be present in the final compound and simultaneously forcing them to merge with each other. To this end, in a certain atmosphere, a high DC current passes through a pair of electrodes which are kept apart from each other by a short distance. In the present study, as it was intended to fabricate AZO nanoparticles, the electrodes were constructed from the mixture of zinc and aluminum, and the reaction atmosphere was pure oxygen.

The medium gas (oxygen) has a dual role in this method: first as a constituent element which should be present in the final products and second as an agent which defines the pressure of the chamber. This pressure has an important role in producing the final product.

2. Experimental procedure

2.1. Preparation of aluminum doped zinc oxide nanoparticles

In order to fabricate $Zn_{1-x}Al_xO$ nanoparticles, we have modified our previous method of fabricating the zinc oxide nanoparticles [10] as illustrated.

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Various amounts of pure aluminum powder (Merck-1.01056.1000) and pure zinc powder (Merck-1.08789.0500) were mixed together as in Table 1. Then, the mixed powders were shaped under a 10 ton pressure to obtain a rod with rectangular cross section. The approximate dimensions of each rod were $1 \times 1 \times 10$ cm^3. Two bars were prepared for each ratio of mixtures. Then the prepared bars were placed in the furnace and heated for 3 h at the temperature of 350 °C.

The remained process for the fabrication of AZO nanoparticles was the same as the method illustrated in Ref. [10]. The method involved passing high current through a pair of electrodes when they were approximately 1 mm far from each other in a medium of pure oxygen gas.

Here, in order to fabricate the AZO nanoparticles, each pair of bars was placed in a homemade reactor with an approximate volume of 7000 cm^3. One of the rods was fixed at the center of the reactor and the other one could easily move to the right and left by the aid of a DC motor. There were side windows in the middle of the reactor so that the inside of the chamber, and the rods movements could easily be seen.

The chamber was evacuated down to 10^{-2} mbar and then filled with oxygen gas until it reached the atmosphere pressure. The above procedure was repeated three times to make sure of the purity of the chamber atmosphere. Finally, the chamber was filled with oxygen gas until the reactor pressure reached 4 bar. Then the bars were connected to the current source. The value of the current source was adjusted to 320 A. After that, approaching of the moveable rod started and after a few moments an arc established between the two electrodes and the dusts of the AZO nanoparticles was produced and deposited on the reactor walls. It was tried to establish a stable arc between the electrodes during the procedure by adjusting the position of the movable electrode.

The above steps were repeated for all the different mixtures and at the end of each experiment the dusts were gathered for analysis.

Table 1
Various amounts of zinc and aluminum used for the fabrication of molded rods.

x	Zn		Al		Formula
	(gm)	(mol)	(gm)	(mol)	
0.01	64.7262	0.99	0.2698	0.01	$Zn_{0.99}Al_{0.01}O$
0.03	63.4185	0.97	0.8094	0.03	$Zn_{0.97}Al_{0.03}O$
0.06	61.4572	0.94	1.6188	0.06	$Zn_{0.94}Al_{0.06}O$
0.09	59.4958	0.91	2.4283	0.09	$Zn_{0.91}Al_{0.09}O$
0.12	57.5344	0.88	3.2377	0.12	$Zn_{0.88}Al_{0.12}O$

2.2. Characterization

The resulted powders had a yellowish green color, and they became greener as the percentage of dopant increased (Fig. 1). The morphology and the structure of samples were analyzed using several measurements such as SEM (Cam-Scan MV2300), EDX and XRD (Philips—PW1840). The optical behavior of samples was investigated by UV–visible (Cintra 101) and photoluminescence spectroscopy (Perkin Elmer LS50B).

3. Results and discussion

3.1. Morphological and structural properties

Fig. 2 shows the SEM images of samples with different amounts of dopants. As shown in the images, nanoparticles in all the samples have spherical shapes. The samples have homogeneous distributions of nanoparticles and the grains size of them varies from 50 to 100 nm.

In Fig. 2f, the typical results of EDX analysis for the samples are shown. The EDX analyses were captured in the limitation range of one nanoparticle to investigate the actual amount of dopant in each nanoparticle. Measurements on different points showed that the ratio of Al to Zn concentrations in each nanoparticle has an acceptable accordance with the fabrication ratio.

Fig. 3 shows the XRD patterns of the samples. The overall XRD patterns of samples have the same feature as the pure zinc oxide but there were some changes in the lattice parameters. The lattice parameters of the samples were calculated from XRD data by the following formula:

$$\frac{1}{d_{hkl}^2} = \frac{4}{3}\left(\frac{h^2+hk+l^2}{a^2}\right)+\left(\frac{l^2}{c^2}\right)$$

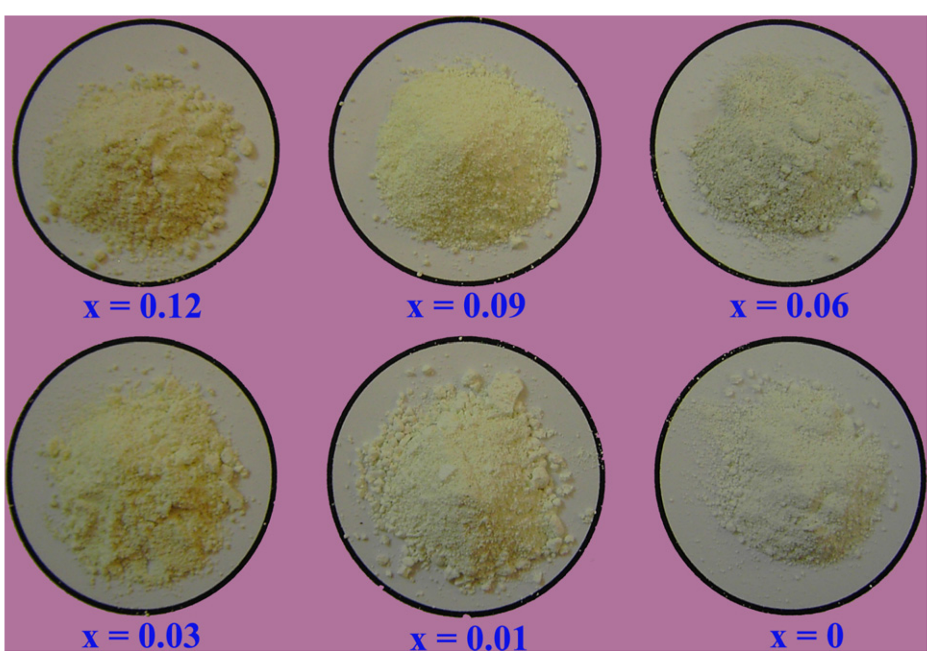

Fig. 1. Images of fabricated $Zn_{1-x}Al_xO$ nanoparticles with different amounts of aluminum concentrations.