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Influence of Annealing Temperature on Structural and Magnetic Properties of Co Doped SnO_2 Thin Films Prepared by Sol-Gel Dip Coating Method

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Abstract: *Sol-gel dip coating technique has been used to prepare pristine and Co doped SnO_2 thin films. The prepared films were annealed at three different temperatures i.e. 300°C, 400°C and 500°C. X-ray diffraction studies show the tetragonal rutile structure of SnO_2 and Co-doped SnO_2 with no additional phases of SnO_2. For Co doped SnO_2 samples annealed at different temperatures the ferromagnetic ordering has been observed. However, coercive field, remnant magnetization as well as saturation magnetization decreases with increase in annealing temperature, showing a crucial phase transformation from ferromagnetism to paramagnetism.*

Keywords: *Semiconductor thin film, Sol-Gel Dip-coating, Annealing, XRD, VSM*

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1. Introduction

SnO_2 (Tin dioxide) is frequently investigated material having unique properties like wide band gap value of 3.6 eV[1], excellent optical transmittance[2], high conductivity[3] and good chemical stability[4]. There are certain studies on the alike material when cobalt (Co) is chosen as dopant atom due to its ferromagnetic and gas sensing properties[5]. Therefore, scientifically being an important material, magnetic characteristics of Co-doped SnO_2 thin films need to be investigated and analyzed in detail. Sol-gel dip coating method is preferred for preparation of thin films annealing at different temperature due to its low processing temperature and possibility of tailoring the starting solutions resulting in better composition and control of the final structure. Until now, great concern of studies has been endured on the incorporation of Co ions of different concentration into the SnO_2 lattice. But, not much work has been done on the study of influence of annealing temperature on the structural and magnetic properties of $Sn_{1-x}Co_xO_2$ thin films. In the present study, Co-doped SnO_2 thin films have been fabricated on glass substrate by sol-gel dip coating method and annealed at different temperatures to study the effect of variation of annealing temperature on their structural and magnetic properties.

2. Experimental Method

Pristine SnO_2 and Co-doped SnO_2 films were deposited by sol-gel dip coating method. To prepare the solution we had taken 0.5 mol of $SnCl_2.2H_2O$ and dissolved in 50 ml of ethanol. The solution was then stirred and refluxed at 80^{o}C for 2 hours. Then, 2% of CoCl ($CoCl_2$. $6H_2O$) was added as precursor into the solution. The solution was again refluxed for 4 hours at 80^{o}C, for homogenous mixing of the solution. The solution was then kept in air for 48 hours. For preparation of $Sn_{1-x}Co_xO_2$ thin films by dip coating method, the clear homogenous solution was ready. The glass slide was dipped in the homogenous solution for 1 hour and then dried at 100°C for 10 minutes. To get an even layer of solution on the substrate, this procedure was repeated three times. Then, the samples were annealed at three different temperatures in a furnace i.e. 300°C, 400°C and 500°C for1 hour.

3. Results and Discussion

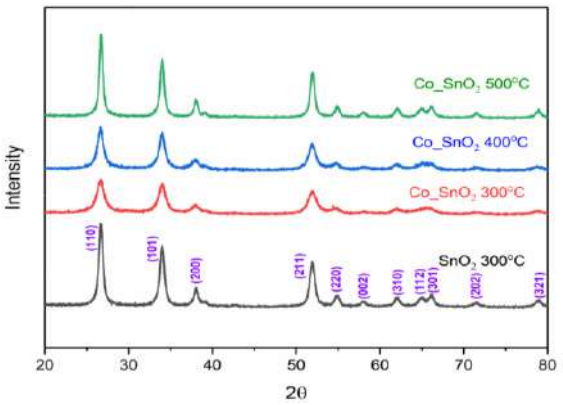

Fig.1.: XRD patterns of pristine SnO_2 annealed at 300°C and Co-doped SnO_2 annealed between 300°C to 500°C.

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From Fig. 1, the peaks revealed tetragonal rutile structure of SnO_2. It is observed that intensities of XRD peaks of Co doped SnO_2 samples increases as the annealing temperature increases. Diffraction peaks become more sharp and intense with increasing annealing temperature and peak broadening goes on narrowing. Broader peak signifies small particle size but with increase in annealing temperature, broadening decreases enhancing the particle size. Increase in peak intensities, with increase in annealing temperature, maybe due to replacement of Sn^{4+} ions with Co ions in the lattice of SnO_2 sample.This process leads to movement of Sn^{4+} ions in the interstitial sites which increasesthe disorders.

Table 1.XRD Data of Co doped SnO_2thin film sample at different annealing temperature

Sample	Crystallite Size (nm)	FWHM	Lattice Constants a (Å)	c (Å)	ε (*10^{-3})	ρ (*10^{15})
SnO_2 (300°C)	25.2	0.0057	4.72	3.18	1.3	1.5
Co_SnO_2 (300°C)	11.13	0.0126	4.65	3.02	2.6	9.1
Co_SnO_2 (400°C)	15.09	0.0118	4.67	3.14	1.8	8.5
Co_SnO_2 (500°C)	23.58	0.0055	4.69	3.11	1.5	5.4

With increasing annealing temperature, the Sn atoms move to grain boundaries from interstitial sites which reduce the lattice mismatch due to recrystallization. Due to which micro-strain and dislocation density of the film decreases. Particle size is small and crystal growth is slow, at lower annealing temperature. But on increasing the annealing temperature, growth of the particle has been noticed. This might be due to coalescence phenomenon that is responsible for reorganization of grains in pristine and doped thin films, thus leading to densification of thin film occurring by filling of the gaps and reducing strains between the grain boundaries.

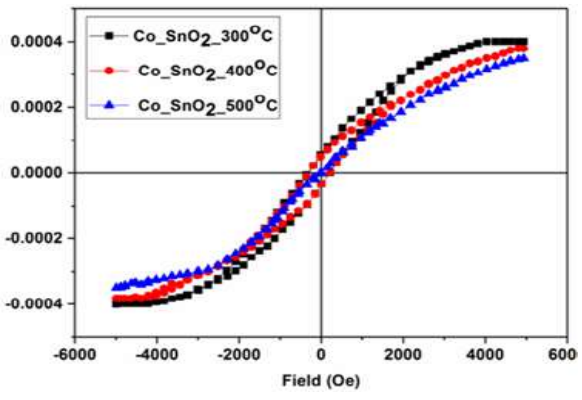

Fig. 2.M-H magnetization curves of Co doped SnO_2 thin film samples annealed at 300°C, 400°C and 500°C.

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Thin film of pristine SnO_2 exhibits diamagnetic behavior at lower annealing temperature (300^oC). It is observed that thin films of Co doped SnO_2 exhibits ferromagnetic ordering at lower annealing temperature.As annealing temperature increases, phase transformation from ferromagnetism to paramagnetism is observed. Saturation magnetization decreases with increase in annealing temperature. This reduction in saturation magnetization has been attributed to the coupling due to antiferromagnetic exchange interaction. From results, it is quite visible that thin film samples of Co doped SnO_2 shows decreasing trend for coercive field and retentivity on increasing the annealing temperature, indicating crucial phase transformation from ferromagnetism to paramagnetism [13].

Table 2.Variation of Saturation Magnetization (M_s) (emu/gm), Coercivity (H_c) (Oe) and Remnant Magnetization (M_r) (emu/gm) of Co doped SnO_2 thinfilms at different annealing temperatures.

Annealing Temperature	Saturation Magnetization	Coercivity	Remnant Magnetization
300^{o}C	2.22	77	1.5×10^{-5}
400^{o}C	2.01	35	4.9×10^{-6}
500^{o}C	1.89	0	0

4. Conclusion

XRD patterns of pristine and Co doped SnO_2 thin films samples exhibit rutile type tetragonal structure with no impurity phase observed. This implies that Co ions were successfully substituted at the Sn site and Co impurities do not change the tetragonal structure of SnO_2. Pristine SnO_2 at 300^{o}C shows diamagnetic behavior. On increasing the annealing temperature to 400^{o}C, ferromagnetic ordering occurs.

With Co doping in SnO_2 lattice diamagnetic character vanishes and ferromagnetic ordering appears even at lower annealing temperature300^{o}C and 400^{o}C. However, at higher annealing temperature i.e. 500^{o}C saturation magnetization, coercive field and remnant magnetization decreases indicating crucial phase transformation from ferromagnetism to paramagnetism. This phase transformation might be due to lesser direct and indirect exchange interactions which might arise because of diffusion of dopant ions which cause distortion in the crystal lattice at higher annealing temperatures.

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