# Abstract

To date state of art transparent conducting oxides (TCOs) material is tin doped indium oxide (ITO). It is due to its low electrical resistivity (~ $10^{-4}$ Ω-cm) and high optical transparency (more than 85 %) in the visible region. Though that material possesses superior optical and electrical properties but the production cost is very high due to limited resources of indium on earth. From application point of view, these films were prepared using very sophisticated techniques like sputtering and atomic layer deposition, which increases its cost. The inherent brittleness of ITO is big hindrance in its use on flexible substrates. To overcome these limitations, one has to explore an alternative material that is abundantly available and inexpensive too. Among various TCOs, zinc oxide (ZnO) is inexpensive, available in abundance, and it seems to be an alternative of ITO. Its optical properties are comparable to that of ITO but its electrical resistivity is rather on higher side. So the objective of thesis is to improve electrical and optical properties of ZnO thin films using inexpensive solution based method.

The main objective of the thesis is to deposit ZnO thin film with pure *c*-texture because thin films with high *c*-texture possess better electrical and optical properties as compared to *a*-texture and *m*-texture ZnO thin films. The texture of ZnO thin films can be easily controlled in magnetron sputtering and pulsed laser deposition technique but in solution based method it is very tricky. The effect of various dopants like Al, Cd, Ba, Li, and In, on the structural, optical, morphological, and electrical properties of the highly *c*-axis oriented ZnO thin films prepared using solution based method have been rarely reported. In most of the investigations, magnetron sputtering and pulsed laser deposition technique have been used.

Some other objectives include: (i) investigations of the effect of dopants mean-square amplitude of atomic vibrations $< u^2 >$ or their Debye-Waller thermal parameter $B = 8\pi^2 < u^2 >$ on the structural and optical properties of ZnO thin film, (ii) improvement in the electrical conductivity of un-doped and In-doped ZnO thin films using thermal treatment in $N_2$ atmosphere, and (iii) to explore whether thin films prepared using solution based method possess any gas sensing ability or not?

The thesis comprises of seven chapters:

**Chapter # 1** describes general, electrical, and optical properties of transparent conducting oxides.

**Chapter # 2** presents a discussion on ZnO thin films with *c*-, *a*-, and *m*-texture as well as a brief review about adjusting these textures by controlling the various deposition parameters in magnetron sputtering, pulsed laser deposition, chemical vapor deposition and solution based method. The effect of various dopants on the *c*-texture of ZnO thin films (mostly prepared by magnetron sputtering technique) have also been reviewed.

**Chapter # 3** reviews various deposition and characterization techniques for thin films. Deposition techniques include sol-gel spin coating and spray pyrolysis. For characterization, x-ray diffraction (XRD), UV-Visible spectroscopy, Scanning electron microscopy (SEM), Fourier Transform Infrared Spectroscopy, Two Point and Four Point Probe technique, Hall effect, and Photoluminescence Spectroscopy have been used.

**Chapter # 4** (original contribution) highlights the influence of Al, Ba, and Cd (concentration range 1%, 3%, and 5%) on the optical, structural and electrical properties of highly *c*-axis oriented ZnO thin films. These thin films were prepared using sol-gel spin coating method. The effect of dopants mean-square amplitude of atomic vibrations $< u^2 >$ or their Debye-Waller thermal parameter $B =$

$8\pi^2 < u^2 >$ on the properties mentioned above have also been discussed. This aspect has been explored for the first time and never been reported before.

**Chapter # 5** (original contribution), deals with the effect of Li dopant (concentration range 1%, 3%, and 5%) on the enhancement of the crystallite orientation along *c*-axis of ZnO thin film have been discussed. Its influence on the optical and electrical properties have also been studied. These films were also deposited using sol-gel spin coating method.

**Chapter # 6** (original contribution), presents the influence of a very wide range of In dopant concentration (0.2%, 0.4%, 0.6%, 0.8%, 1.0%, 1.25%, 1.5%, 2.0%, 2.5%, 5.0%, and 10%) on the optical, structural, and electrical properties of ZnO thin films using spray pyrolysis technique. Such a broad range of dopant concentration has never been explored and reported in the past. The effect of thermal treatment in $N_2$ atmosphere on the physical properties of these thin films have also been investigated.

**Chapter # 7** (original contribution), highlights the gas sensing ability of In-doped ZnO thin films prepared using spray pyrolysis technique. These films show their potential ability to detect oxygen, and can be used as oxygen gas sensor.

**Chapter # 8** presents the conclusion from the original contribution

Finally some suggestions have been given to extent this work in future.

**(A) Papers Published in Peer Reviewed International Journals from the Thesis**

1. **Dilawar Ali**, M. Z. Butt, Conor Coughlan, David Caffrey, Igor V. Shvets, Karsten Fleischer:
*Nitrogen grain boundary passivation of In-doped ZnO transparent conducting oxide*
Physical Review Materials 2 (2018) 043402

2. **Dilawar Ali**, M.Z. Butt, Bilal Arif, Ahmed A. Al-Ghamdi, Fahrettin Yakuphanoglu:
*The role of Al, Ba, and Cd dopant elements in tailoring the properties of c-axis oriented ZnO thin films*
Physica B 506 (2017) 83–93

3. **Dilawar Ali**, M Z Butt, Bilal Arif, Abdullah G Al-Sehemi, Ahmed A Al-Ghamdi, Fahrettin Yakuphanoglu:
*Li induced enhancement in c-axis orientation and its effect on structural, optical, and electrical properties of ZnO thin films*
Mater. Res. Express 4 (2017) 026405

**(B) Papers Published/Submitted in Peer Reviewed International Journals not included in the Thesis**

1. **Dilawar Ali**, M. Z. Butt, Iqra Muneer, Farooq Bashir, Murtaza Saleem:
*Correlation between structural and optoelectronic properties of tin doped indium oxide thin films*
Optik 128 (2017) 235–246

2. **Dilawar Ali**, M. Z. Butt, I. Muneer, M. A. Farrukh, M. Aftab, M. Saleem, F. Bashir, A. U. Khan:
*Synthesis and characterization of sol-gel derived La and Sm doped ZnO thin films: A solar light photo catalyst for methylene blue*
Submitted for publication in Thin Solid Films (Submission date 16-07-2018)