Evolution of Dielectric Relaxation and High k dielectricity in Doped Oxides: Interplay of crystal structure and Electronic states in the lattice

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By

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Chapter-7

SUMMARY AND FUTURE SCOPE

7.1 Summary

I have studied several compositions of doped ZnO in wurtzite structure. The data containing the dielectric constant of different samples at 1000 Hz frequency is compiled in the table given below. The increase in dielectric constant was observed with increasing dopant concentration. Thus in general high entropy in the lattice results superior dielectric constant of the material.

Table 7.1: List of materials and their obtained dielectric constant at 1000 Hz.

Sample	Dielectric constant
ZnO	450[1]
$Zn_{0.95}Cu_{0.05}O_{1-\delta}$	140000
$Zn_{0.92}Cu_{0.05}Li_{0.03}O_{1-\delta}$	18000
$Zn_{0.9}Cu_{0.05}Li_{0.05}O_{1-\delta}$	20000
$Zn_{0.9}Fe_{0.1}O_{1+\delta}$	612
$Zn_{0.9}Li_{0.05}Fe_{0.05}O$	90000

Further, I have studied single-phase Ti^{4+} ion substituted $CoNb_2O_6$ in the form of $CoNb_{2-x}Ti_xO_6$. The data containing the dielectric constant of different samples at 100 Hz frequency is compiled in the table given below. The increase in dielectric constant is because of bond strength or relative ionicity in the M-O bond. Which plays a major role in the change in net dipole moment (i.e. because of entropy change of the

Sample	Dielectric constant
CoNb2O6	500
$CoNb_{1.95}Ti_{0.05}O_6$	700
$CoNb_{1.9}Ti_{0.1}O_6$	**14000**

The key findings of the above results are that the dielectric constant increases with an increase in doping. Thus high entropy doped oxide can be a choice of material for the development of high k dielectrics. The high entropy or doping/ presence of multiple cations in a lattice results in the formation of different M-O bonds having different relative ionic strengths or iconicity. This results in a net dipole moment or polarisation vector in the lattice that can be responsible for to result of high k dielectrics. Further relaxor nature of high k dielectric is also observed in high entropy doped oxides. Further, the details finding of this dissertation is presented below:

Chapter 1 contains a general introduction to complex materials like $CoNb_2O_6$ and Binary Oxides. These materials have received great interest in high dielectrics, ferroelectrics, and magnetic effects resulting from their mutual interaction. The details of complex $CoNb_2O_6$ and ZnO materials considered in the current study and the purpose of the thesis are mentioned in this chapter.

Chapter 2 described the experimental procedure used for the preparation and characterization of the materials. The sol-gel route and the solid-state route were used for the preparation of materials. X-ray diffraction has been studied for the determination of phase purity. Scanning electron microscope (SEM) and energy-dispersive X-ray spectroscopy (EDX) analysis have been used for microstructural studies and elemental analysis of materials respectively. The oxidation state of the ceramics was to be examined by X-ray photoelectron spectroscopy; Magnetic properties of the materials were measured by MPMS with the variation in temperature and applied field. Dielectric properties of materials were measured by EIS study with the variation of temperature and frequency. The ferroelectric study was done by a P-E Loop tester with the variation of the applied field.

In Chapter 3, I successfully synthesis the Cu^+ doped ZnO and Up to 8% of Cu^+ ions were substituted in the ZnO lattice and the highest dielectric constant (~6300) was obtained at 600°C for $Zn_{0.95}Cu_{0.05}O_{1-\delta}$ at 100 KHz frequency. The materials $Zn_{0.95}Cu_{0.05}O_{1-\delta}$ also exhibit ferroelectricity at room temperature with remnant polarization Pr and Vc equal to $9.60*10^{-03}$ μC/cm^2 and $3.83*10^{+02}$ V/cm respectively.

In Chapter 4, I have done a synthesis of single-phase Cu/Li co-doped ZnO. In this chapter, $Zn_{0.92}Cu_{0.05}Li_{0.03}O_{1-\delta}$ and $Zn_{0.9}Cu_{0.05}Li_{0.05}O_{1-\delta}$ ceramics samples were prepared by the modified sol-gel method. However, we are not able to achieve more than 10% doping of Cu and Li in ZnO. We have done X-ray diffraction analysis and scanning electron microscopy which shows the polycrystalline nature of samples and the composition of the sample was analyzed by an EDX probe attached SEM instrument. The material has having high dielectric constant as compared to pure ZnO. The material $Zn_{0.9}Cu_{0.05}Li_{0.05}O_{1-\delta}$ also exhibits ferroelectricity and dilute magnetic properties.

In Chapter 5, $Zn_{1-x}Fe_xO_{1+\delta}$ and $Zn_{1-2x}Li_xFe_xO$ ceramic samples were prepared by the conventional sol-gel method where; (x=0.05, 0.1). The structural parameters and phase purity have been studied using powder X-ray diffraction patterns. The SEM study showed that the grains are of sub-micrometer sizes in the range of 0.5-1μm. The electronic structure of the sample was investigated by the XPS study. The impedance spectroscopy was carried out at a variable temperature in the air to study the dielectric behavior of the material in the frequency range of 1 MHz to 1 kHz. The dielectric constant (ε_r') is equivalent to 612 for $Zn_{0.9}Fe_{0.1}O_{1+\delta}$ and 90000 for $Zn_{0.8}Li_{0.1}Fe_{0.1}O$. at 1000 Hz frequency at 400°C. Also with an increase in frequency, the dielectric constant and dielectric loss were found to decrease.

In Chapter 6, Ceramic samples of $CoNb_{2-x}Ti_xO_6$ were generated using a modified solid-state approach. Powder X-ray diffraction patterns have been used to investigate the structural characteristics as well as the phase purity. Images captured by an SEM show a dense pellet that is devoid of porosity and voids at the surface. The material shows both dielectric constant and dielectric loss were decreasing with increasing frequencies. The dielectric constant (ε_r') for $CoNb_2O_6$ was found to be 500, $CoNb_{1.95}Ti_{0.05}O_6$ is 700 and $CoNb_{1.9}Ti_{0.1}O_6$ is 14000 respectively at 100 Hz frequency at 200^{o}C and then decreases, clearly shows relaxor type behavior. Samples also exhibit ferroelectric behavior.

Further, this dissertation can be extended to develop novel high-k dielectrics and ferroelectric materials. The detailed future scope of this dissertation is represented below.

7.2 Future Scope

As far as the material application is concerned, the following work objective can be taken for further advancements and growth of the dielectric materials:

- ❑ The dielectric study can give better results in thin film.
- ❑ To utilize the materials in the real application.
- ❑ At higher frequencies the behaviour of materials.

References:

[1]. Khalid Omar, M. D. Johan Ooi & M. M. Hassin, Investigation on Dielectric Constant of Zinc Oxide, Modern Applied Science, Vol. 3, No. 2, February 2009.