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ABSTRACT

Polycrystalline spinel ferrites have wide range of applications in industrial and home appliances due to their interesting electric and magnetic properties. It is well known that the characteristics of ferrites, depends upon, their chemical compositions, preparative conditions and substitutions. By introduction of a relatively small amount of foreign ion, important modifications in structural, magnetic and electrical properties can be obtained.

The spinel ferrites with different ion substitution have been the subject of many investigations (1-4). The literature survey show that Mn$^{2+}$ ferrite posses about 80% normal spinel structure (5) and its degree of inversion does not depend upon the heat treatment.

Influence of Si$^{4+}$ substitution on magnetic properties of CoFe$_2$O$_4$ spinel ferrite system was studied by Shinde et.al. (6) The ferrite system Mn-Mg-Fe-Cr-O is studied by N.K. Raut (7).
In the literature there is no mention of the systematic study of the effect of the substitution of Si$^{4+}$ on the ferrite system MnFe$_2$O$_4$ and MnFeCrO$_4$.

In view of this, in the present investigation, effort has been made to investigate the effect of the substitution of Si$^{4+}$ on the magnetic, electrical, transport and dielectric properties of the spinel ferrite systems.

$$\text{Mn}_{1+x} \text{Si}_x \text{Fe}_{2-2x} \text{O}_4$$

And

$$\text{Mn}_{1+x} \text{Si}_x \text{Fe}_{1-x} \text{Cr}_{1-x} \text{O}_4$$

Where x = 0.0 to 0.5 in steps of 0.1

This thesis has been logically arranged in five chapters as described below.

CHAPTER 1:

INTRODUCTION

This chapter deals with the introduction to the field of ferrites, different types of magnetism, application of ferrites, survey of the field of Mn ferrites and aim of the present investigation.
CHAPTER 2:

CRYSTAL STRUCTURE, ANALYSIS AND SYNTHESIS OF FERRITES.

This chapter gives description in detail about the spinel structure of ferrites, classification of ferrites, oxygen parameter, U – parameter, structural co-ordinations of cubic spinels, cation distribution, factors affecting cation distributions and infrared techniques used for the structural determination of ferrite system. It also includes the X-ray diffraction method used for the analysis of the spinel structure and different methods used for the synthesis of the ferrite.

CHAPTER 3:

MAGNETIC, ELECTRICAL AND DIELECTRIC PROPERTIES OF FERRITES.

This chapter includes description about magnetization, magnetic hysteresis loop, classification of ferrites depending upon
magnetization as soft and hard ferrites and models for magnetization (Neel’s model, Yafet-Kittel model)

A.C. susceptibility, electric and dielectric properties of ferrites, different models such as Hopping model of electrons (8,9) small polaron model (10). Suggested to account for electrical properties of ferrite are discussed at length. The method for the estimation of cation distribution is also described in detail.

CHAPTER 4:

EXPERIMENTAL METHODOLOGY

This chapter gives a brief description of the different experimental methods employed for the preparation and characterization of the ferrite materials studied under the present work. The measurement techniques employed during the investigation viz. X-ray diffraction, I.R. spectroscopy, magnetization, ac susceptibility measurement and measurement of ac/dc electric resistivity and dielectric properties are described in details.
CHAPTER 5:

RESULTS AND DISCUSSIONS

This chapter is divided in two parts A & B. In each part the results of investigations followed by the corresponding discussions for the systems under the present investigation are given.

The systems under the present investigation are

1] Mn_{1+x} Si_x Fe_{2-2x} O_4
2] Mn_{1+x} Si_x Fe_{1-x} Cr_{1-x} O_4

where x = 0.0 to 0.5 in steps of 0.1

The ferrite systems under the investigation are prepared using solid state double ceramic technique. These spinels were characterized by X-ray diffraction for their single phase formation. The magnetic properties of these systems were investigated by employing a.c. susceptibility measurements and magnetization.

The electrical properties are investigated by measuring ac resistivity, capacitance and Q factor at frequencies 100 Hz &
1KHz. The dc resistivity of the systems is measured from room temperature to 773°K.

The observed magnetic moment per formula unit \( n_B (\mu_B) \) for both the systems Mn-Si-Fe-O, Mn-Si-Fe-Cr-O exhibit conventional behaviour i.e. as concentration increases magnetic moment per formula unit decreases. On the basis of intensity calculations for X-rays, magnetization data, octahedral site preference energies and empirical distribution scheme of verwey and Heilmann [11] cation distributions are determined for the present ferrite systems and they are:

\[
(Mn_{0.8-x} Fe_{0.2+x})^A [Mn_{0.2+2x} Si_x Fe_{1.8-3x}]^B O_4
\]

\[
(Mn_{0.8-\frac{x}{2}} Fe_{0.2+\frac{x}{2}})^A [Mn_{0.2+3x/2} Si_x Fe_{0.8-3x/2} Cr_{1-x}]^B O_4
\]

where \( x = 0.0 \) to 0.5 in step of 0.1

For the system Mn\(^{2+}\) cation have strong tetrahedral site preference. But due to different tetrahedral site preference energies of Mn\(^{2+}\), these cations are also found in small proportion in B- site, which is confirm by X-ray intensity calculations and magnetization data. The calculated values of \( n_B (\mu_B) \) using cation
distribution agree well with the experimental values for the compositions.

X-ray intensity calculations have been done and the intensity ratios of the structural sensitive reflecting planes such as $I_{220}/I_{400}$, $I_{422}/I_{400}$, $I_{400}/I_{422}$, have been estimated and considered for determination of cation distribution. With the help of the cation distribution the theoretical values lattice parameter have also been determined.

In infrared spectra of all the samples shows the absorption of infrared radiation of the wave numbers corresponding of tetrahedral complexes of the spinel crystal structure. The force constants and bond lengths of tetrahedral and octahedral sites have been determined. The force constant at tetrahedral site ($K_t$) is found decreasing with increase in tetrahedral bond length ($R_A$) and force constant at octahedral site ($K_0$) is found decreasing with decrease in bond length ($R_0$). The force constant at tetrahedral site ($K_t$) is found greater than the force constant at octahedral site ($K_0$). These results are in agreement with the general behavior usually obtained in the mixed metal oxides.
The measurements of dc resistivity as a function of temperature are carried out from 323°K to 723°K for all the system. The activation energy values calculated from the resistivity (ρ)dc against 1000/T plots give information about transport properties of the system.

The measurements of a.c. resistivity (ρac), capacitance (c) and Q factor were carried out at fixed frequencies 100 Hz & 1KHz from 323 to 773K for Mn-Si-Fe-O and Mn-Si-Fe-Cr-O systems.

The dielectric constant (ε') dielectric loss (ε'') and dielectric loss tangent (tan δ) were calculated from the capacitance (c) and Q- factor at fixed frequencies 100 Hz to 1KHz. The large value of dielectric constant (ε') at low frequencies observed for all the system may be due to predominance of the species like Fe²⁺ ions, oxygen vacancies, grain boundary defect and interfacial dislocation pile up etc. (9). It is observed that ε' and ε'' increase with temperature for all the samples.