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VARIATION OF TRANSPORT PROPERTIES AND APPARENT DENSITY OF SOFT NICKEL FERRITE

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ABSTRACT

Polycrystalline $NiFe_2O_4$ was prepared from Analytical Grade powder of NiO and Fe_2O_3 by conventional double sintering technique. The apparent density of the prepared sample increases with the increase in sintering temperature and consequently the porosity decreases which has a great influence on electrical and dielectric properties of soft ferrites. It is observed that with the increase in temperature, the DC resistivity of the prepared sample decreases which confirms the semiconducting behavior of the prepared $NiFe_2O_4$. In addition to this, at room temperature it is observed that the resistivity decreases with the increase in sintering temperature. Variations of activation energy have been observed for different sintering temperature and are in agreement with the resistivity measurement. Frequency dependence of dielectric constant (κ) has been measured and shows the normal dielectric behavior of the prepared sample which can be explained on the basis of Koop's two layer model and Maxwell-Wagner polarization theory. Along with these, enhancement of dielectric constant has been observed with the increase in sintering temperature.

Key words: Apparent density, DC resistivity, Activation energy, Dielectric constant

INTRODUCTION

Soft ferrites are the most versatile electronic materials suitable for higher frequency application in the telecommunication field (Sugimoto 1999). Due to their high resistivity and low eddy current losses these ferrites are used in radio frequency circuits, high quality filters, rod antennas, transformer cores, read/write heads for high speed digital tape and operating devices (Igarash and Okazaki 1977, Tseng and Jou 1989, Riches 1972). They are more stable, relatively inexpensive, easily manufactured, have good magnetic properties, low dielectric loss and high electrical resistivity. The electrical and dielectric properties are the most important properties for ferrites. So, the study of the electrical resistivity produces valuable information on the behavior of free and localized electric charge carriers in the samples.

It is known for ferrite materials that the transport and magnetic properties are sensitive to their microstructures, especially grain size, apparent density and porosity

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(Zaag *et al.* 1993) depending on the processing conditions, sintering temperature and time, chemical composition and the amount and type of the additives (Gopalan *et al.* 2009, Patil and Chougule 2009). So, in order to reduce the losses many parameters such as density, grain size, porosity and their intra- and intergranular distribution must be controlled (Ishino and Narumiya 1987). With the increase of sintering temperature, grain size, porosity and density of the $NiFe_2O_4$ can be controlled, which influence the transport and magnetic properties of the ferrites. Thus obtaining optimum sintering temperature is important. Effect of sintering temperature on microstructures, magnetic and transport properties of $NiFe_2O_4$ has been reported for the samples prepared from nano size powder of NiO and Fe_2O_3 (Choudhury *et al.* 2011, Bhuiyan *et al.* 2010). In this study, therefore, effect of sintering temperature on apparent density and transport properties of $NiFe_2O_4$ synthesized from Analytical Grade powder of NiO and Fe_2O_3 have been reported.

EXPERIMENTAL DETAILS

Polycrystalline $NiFe_2O_4$ was prepared through the solid state reaction using conventional double sintering ceramic technique from Analytical Grade powder of NiO and Fe_2O_3. The purity of the constituent elements was NiO (99.9%) and Fe_2O_3 (99.9%) and was obtained from E. Mark of Germany at the Materials Science Division, Atomic Energy Center Dhaka. After thorough mixing the powder was presintered at 1100°C for 3 hours. The presintered ferrite powder was crushed and mixed with 1 wt. % polyvinyl alcohol (PVA) as a binder and uniaxially pressed into toroid and pellets. The compacts were successively sintered in a muffle furnace in air from the temperature range 1000°C to1400°C to eliminate the PVA and finally furnace cooled to room temperature. The bulk densities of the pellet samples were determined by measuring volume and mass after sintering. DC electrical resistivity was measured up to 300°C using Electrometer Keithley model 6514. Frequency dependence dielectric constant (κ) of the pellet samples has been measured up to 13 MHz by Wayne Kerr 3255 B Impedance Analyzer.

RESULTS AND DISCUSSION

Sintering temperature dependent apparent density has been presented in Fig 1. It has been observed that with the increase in sintering temperature, the apparent density of the prepared samples increases and consequently the porosity decreases which is due to the diffusion kinetics.

High resistivity is a pre-requisite for high frequency applications of ferrites to counter the eddy current losses, which degrade the ferrite performance. In Fig. 2, DC electrical resistivity as a function of inverse temperature has been presented for the samples for various sintering temperatures. It is clear from Fig. 2 that with the increase in temperature, the resistivity decreases which is the normal semiconducting behavior of the prepared sample.

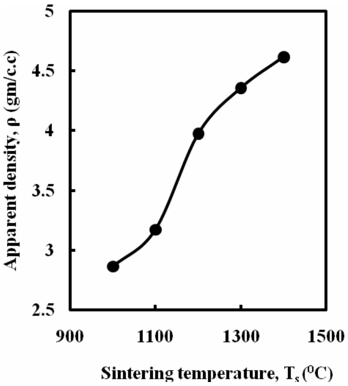

Fig. 1. Sintering temperature dependent apparent density.

In Fig. 3, the resistivity is seen to vary with sintering temperature. The resistivity was lowest for the samples sintered at 1400°C. Decrease in resistivity with increase in sintering temperature may be attributed to the micro structural factors such as grain size, porosity, grain boundary area as well as conversion of trivalent Fe^{3+} ions to the divalent Fe^{2+} state.

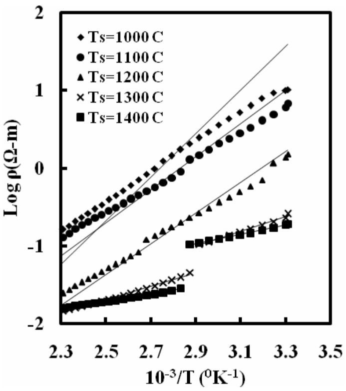

Fig. 2. Temperature dependent DC resistivity.

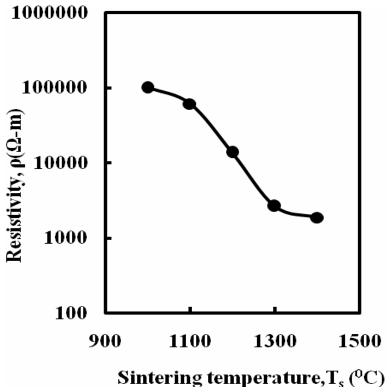

Fig. 3. Sintering temperature dependent resistivity.

Smaller the grain size more would be the number of grain boundaries. The ferrite grains being conducting, the bulk of the resistivity is contributed by the insulating grain boundaries. Increase in sintering temperature results in greater density and grain growth which decrease the porosity and the number of grain boundaries (Verma and Chatterjee 2006). And since the pores are non-conductive, the charge carriers will face less pores on their way for higher sintering temperature and thereby leading to decrease in resistivity.

In addition to this, with increasing sintering temperature partial reduction of trivalent Fe^{3+} ions to the divalent Fe^{2+} takes place. The Fe ions at the A sites contribute little to conduction due to the larger distances between them but the formation of Fe^{2+} ions gives rise to conduction of ferrite due to electron hopping between Fe^{3+} and Fe^{2+} ions co-existing at the closer spaced B sites in the spinel lattice and thereby decrease the resistivity (Amarendra *et al*. 2002).

The activation energy has been calculated and is shown in Table 1. It is observed that the activation energy decreases with increasing sintering temperature. Decrease of activation energy with the increase of sintering temperature may be attributed to the fact that at a high sintering temperature, partial reduction of Fe^{3+} to Fe^{2+} takes place and these places act as donor centre and are responsible for this decrease in activation energy (Shamima Choudhury *et al.* 2011). The conduction mechanism is due to hopping of electron of the types $Fe^{2+} \leftrightarrow Fe^{3+}$. It can also be seen that samples having low resistivity have low activation energies and *vice versa* (Shabasy 1997).

Table 1. Variation of activation energy with the variation of sintering temperature.

Sintering temperature, T_s (^{0}C)	Activation energy (eV)	
	Low temperature	High temperature
1000	0.5546	0.5546
1100	0.4202	0.4202
1200	0.392	0.392
1300	0.1865	0.1701
1400	0.1172	0.0975

The variations of dielectric constant as a function of frequency for samples $NiFe_2O_4$ are shown in Fig. 4. It is clear that the dielectric constant decreases rapidly with increasing frequency at the lower frequency region and become asymptotic to lower values at high frequencies. The decrease of dielectric constant with increasing frequency is a normal dielectric behavior of spinel ferrites. The dielectric dispersion curve can be explained on the basis of Koop’s two layer model and Maxwell-Wagner polarization theory. To interpret the frequency response of dielectric constant in ferrite materials, Koop’s suggested a theory in which relatively good conducting grains and insulating grain boundary layers of ferrite material can be represented with the behavior of an inhomogeneous dielectric structure as described by Maxwell (Maxwell 1982). Since an assembly of space charge carries in the inhomogeneous dielectric structure described requires finite time to line up their axes parallel to an alternating electric field, the dielectric constant naturally decrease, if the frequency of the reversal field increases. This is in agreement with the observed dielectric dispersion.

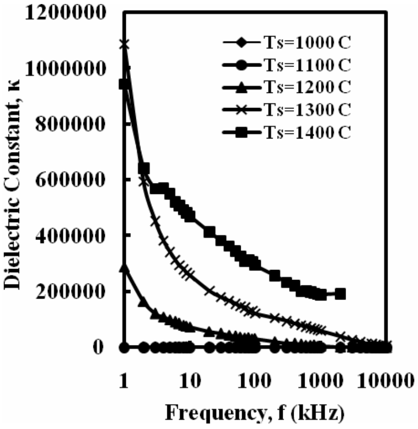

Fig. 4. Frequency dependent dielectric constant.

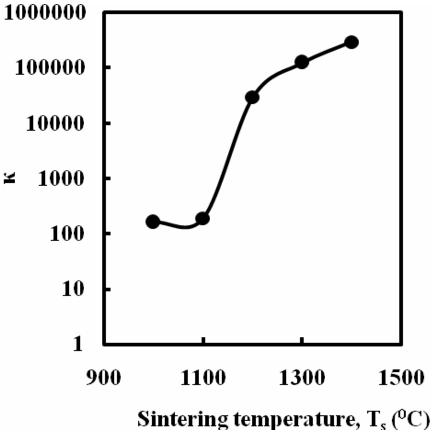

Fig. 5. Sintering temperature dependent dielectric constant at f = 100kHz.

The variation of dielectric constant with sintering temperature at frequency f=100 kHz is shown in Fig. 5. It is observed that the dielectric constant (κ) increases with increasing sintering temperature. Dielectric constant in ferrites is contributed by several structural and micro structural factors. The space charge polarization resulting from electron displacement on application of electric field and the subsequent charge build up at the insulating grain boundary is a major contributor to the dielectric constant in ferrites. Therefore, more the number of Fe^{2+} ions in the ferrite more would be the space charge polarization expected due to the ease of electron transfer between Fe^{3+} and Fe^{2+} ions and consequently higher the dielectric constant. Now, with increasing sintering temperature, partial reduction of Fe^{3+} to Fe^{2+} takes place. So the value of dielectric constant (κ) increases with increasing sintering temperature.

CONCLUSIONS

The present study confirms the influences of sintering temperature on the apparent density, electrical resistivity and dielectric constant of $NiFe_2O_4$. With the increase in sintering temperature, enhancement of apparent density and decrease in porosity has been observed which are due to the diffusion kinetics. Decrease in DC resistivity has been found with the increase in sintering temperature which is due to the microstructural factors such as grain size, porosity, grain boundary area as well as conversion of trivalent Fe^{3+} ions to the divalent Fe^{2+} state. The decrease in activation energy with the increase in sintering temperature may be attributed due to the conversion of trivalent Fe^{3+} ions to the divalent Fe^{2+} state which is also in agreement with the resistivity measurement. Increased values of dielectric constant have been observed because with the increase in sintering temperature, conversion of trivalent Fe^{3+} ions to the divalent Fe^{2+} ions took place and thereby increased in dielectric constant.

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