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Analysis of Electric Relaxation and Polaron Conduction in Nano-sized $SrFe_{12}O_{19}$

N. S. Krishna Kumar, T. S. Shahid, G. Govindaraj*

Department of Physics, School of Physical, Chemical and Applied Sciences
Pondicherry University, Puducherry, India

***Corres.author: ggraj_7@yahoo.com**

Abstract: Nanocrystalline Strontium hexaferrite was synthesized by co-precipitation method and characterized with X-Ray diffraction. Ac electrical studies of the sample were carried out by impedance spectroscopy for frequency range of 1Hz to 1MHz for various temperatures from 313K to 513K. The impedance data show anomalous electric relaxation and it was modelled with electrical equivalent circuit and Cole-Cole relaxation function. Hopping and conduction show a non-Arrhenius behaviour which is an indication of temperature dependence of activation energy. The power law behaviour of ac electric conductivity of the sample has been used to analyse polaron conduction. Summerfield scaling of ac conductivity did not follow at high frequency. The mechanism of electron hopping and polaron conduction is discussed.
Keywords: anomalous relaxation; ac conductivity; polaron conduction.

Introduction and Experimental:

Generally ferrite materials are used for magnetic applications. Many ferrites have applications on dc to high frequency electrical application. $SrFe_{12}O_{19}$ is used in microwave devices, telecommunication and electronic industry. In present work, the anomalous electrical conduction of $SrFe_{12}O_{19}$ is discussed.

Chemical co-precipitation method was used to prepare $SrFe_{12}O_{19}$ ferrite [1]. Analytical grade $Sr(NO_3)_2$ and $Fe(NO_3)_3.9H_2O$ used for synthesis. Sodium hydroxide (NaOH) was used as precipitating agent. Metal nitrates were dissolved in doubly distilled water in required molarities and mixed thoroughly with constant stirring to form a homogeneous solution. The aqueous mixture was slowly heated to 70 ^oC. NaOH of 2M concentration was added drop wise to the above solution and pH was adjusted to 11.5. To achieve good homogeneity and optimum crystal size, the solution was continuously stirred for 3 hr. The precipitated sample was washed several times with distilled water and centrifuged at 2000 rpm. It was kept in an oven overnight to dry out and later calcinated at 900 ^oC for 1 hr at a heating rate of 5 ^oC. Finally sample was pressed to form pellet and sintered at 900 ^oC and subjected to electrical studies. Material was characterized by powder X-ray diffraction at room temperature using X'Pert PANalytical X-Ray diffractometer (XRD). Complex impedance data were taken for a frequency range of 1Hz to 1MHz for temperature range 313K to 513K with Novocontrol broadband dielectric spectrometer.

Results and Discussion:

XRD spectrum of $SrFe_{12}O_{19}$ sintered at 900 °C is taken and confirmed the formation of pure magnetoplumbite structure. The crystallite size was calculated by Scherrer formula and was obtained as 38 nm. Impedance data of the sample show anomalous relaxation. Nyquist plot of the impedance is a depressed one which indicates the deviation from ideal resistance (R) and capacitance (C) parallel combination behaviour. The anomalous electrical relaxation is obtained due to deviation of distribution of relaxation time from ideal case. This anomalous nature is incorporated in empirically formulated Cole-Cole function with a parameter α, where $0<\alpha\leq1$. The parameter α=1 indicates Debye type relaxation (R-C parallel). The Cole-Cole relaxation function can be considered as constant phase element (CPE) parallel with R. CPE has impedance form $Z_{CPE}=1/Q(i\omega)^{\alpha}$ which behaves as a constant phase of $\alpha\pi/2$. Relaxation time in Cole-Cole function can be obtained by $\tau=(RQ)^{1/\alpha}$. Figure 1 shows Cole-Cole function fit of the impedance data and parameters are used to obtain the conductivity and relaxation time [2].

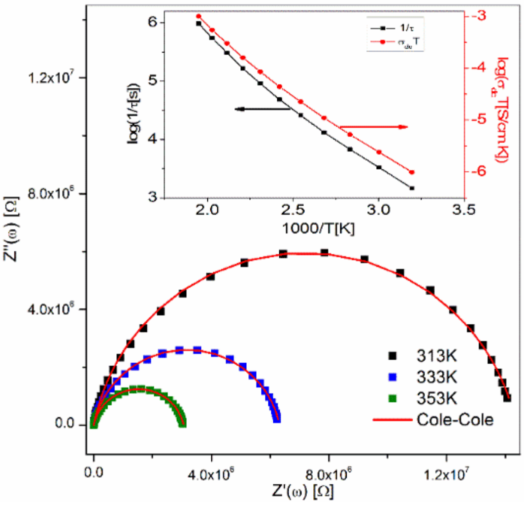

Figure 1. Ac impedance data and Cole-Cole function fit. Inset shows Arrhenius plot of conduction and hopping processes

The conductivity expected to follow Arrhenius equation, $\sigma_{dc}T=\sigma_0 \exp(-E_a/k_BT)$ where Ea is the activation energy. Hopping process expected to follow $1/\tau=1/\tau_0 \exp(-E_h/k_BT)$ where E_h is the activation energy for hopping and $1/\tau_0$ is the attempt jump [3]. The inset of Figure 1 shows the conduction and hopping which deviates from the Arrhenius behaviour due to the variation of activation energy with temperature. Slope of the Arrhenius plot gives the activation energy for each temperature and it is obtained as it varies from 0.26eV to 0.63eV with increase of temperature. It indicates the increase of number carriers with temperature in addition to increase of mobility with temperature.

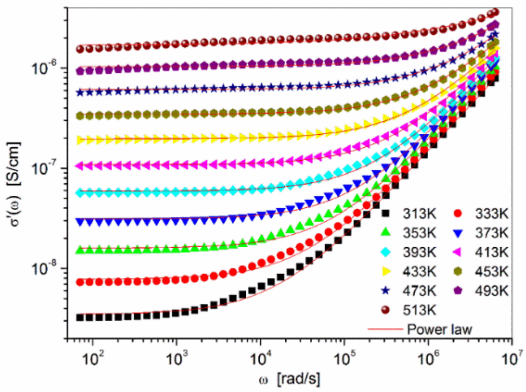

Figure 2. Ac conductivity data and power law fit

Ac conductivity data show a frequency independent dc region and dispersive high frequency region as shown in Figure 2. Most of the dielectric materials follow the Jonscher's power law $\sigma'(\omega)\propto\omega^n$ for the ac conductivity. The ac conductivity is analysed with Almond-West formulation $\sigma'(\omega)=\sigma_{dc}(1+(\omega/\omega_p)^n)$ where ω_p is the hopping frequency [4]. Polaron is a quasi-particle of electron and lattice deformation produced due to motion of electron through the dielectric. The parameter n increases with increase of temperature and then decreases with increase of temperature as shown in inset of Figure 3. Variation of parameter n with temperature indicate the mechanism of conduction, n increases with increase of temperature indicates that the small polaron conduction is predominant and n decreases with increase of temperature is an indication of predominance of large polaron conduction [5]. This studies indicate that the conduction mechanism in the present material is a combination of small and large polaron. The anomalous temperature dependence of conductivity occurs due to the variation of number of polaron with temperature in addition to the variation of mobility. The polaron may be originating from the Sr^{2+}-Sr^{3+} valences and oxygen vacancy assisted polaron from Fe^{3+} [6]. The Summerfield scaling of ac conductivity deviates at high frequency as shown in Figure 3 which indicates the variation of mechanism of dc and ac conduction.

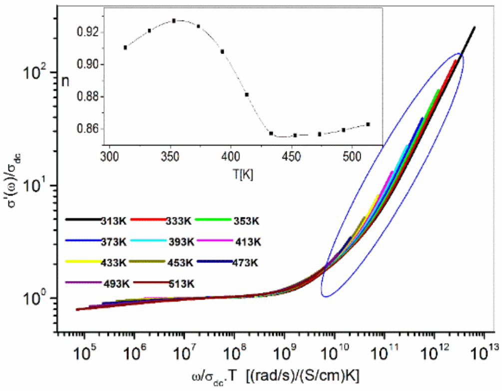

Figure 3. Summerfield scaling with the dc conductivity. Inset shows variation of power law parameter with temperature.

Strontium hexaferrite was synthesised by chemical co-precipitation method. Anomalous electric relaxation and conduction were investigated and variation of activation energy with temperature was observed. Ac conductivity studies indicate a mixed small and large polaron conduction.

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