Abstract

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The $Nd_{1-x}Sr_xFeO_3$ ($0 \leq x \leq 0.5$) and $NdFe_{1-y}Ni_yO_3$ ($0.1 \leq y \leq 0.7$) orthoferrite systems are successfully synthesized by solid state reaction method. In the present study the structural, electrical and magnetic properties of these materials are investigated. The orthorhombic structure is observed in all systems. With the increase in Sr and Ni doping, increase in tolerance factor and decrease in cell volume is observed. Orthorhombic distortion decreases with increase in the Sr and Ni contents up to 50% while above 50% Ni doping it increases. SEM examination of the samples, indicate increase in grain size with increase in the Sr and Ni contents. Comparison of bulk and theoretical densities indicate that in all samples porosity is less than 3%. Room temperature Mössbauer spectroscopic investigations are performed to explain local structure, Fe oxidation states and collapse of the magnetic ordering with increase in the Sr and Ni doping level. These results show an increase in the Fe^{4+}/Fe^{3+} ratio, sagging and local octahedral distortions with increase in the strontium content; the origin behind the anomalous octahedral distortions in this system can be attributed to the increase in the concentration of Fe^{4+}, mismatch in the cationic radii and decrease in the oxidation state of Sr in comparison to Nd. In Ni doped samples the Fe oxidation state remains +3. The major factors behind the collapse of magnetic ordering in the Sr and Ni doped systems are weakening of super-exchange interactions, decrease in the Neel temperature, increase in the spin-spin relaxation frequency and high spin to low spin transition.

Impedance spectroscopic studies are carried out for the investigation of ac electrical properties of different electroactive regions, conduction mechanisms and dielectric properties. It is observed that grains, grain boundaries and electrode-semiconductor contacts are the three electro-active regions present and the conduction and relaxation processes are thermally activated in all systems. In order to correlate the electrical properties with the microstructure of these materials, different equivalent circuit models are proposed. In $NdFeO_3$ small polaronic hole hopping (SPH) conduction mechanism is present in the measured temperature range while in $Nd_{1-x}Sr_xFeO_3$ and $NdFe_{1-x}Ni_xO_3$ systems, conduction mechanism changes from Mott variable range hopping (VRH) to adiabatic SPH with the increase in temperature. The temperature at which change of conduction mechanism is observed decreases with increase in the Sr and Ni concentrations. The activation energy of conduction decreases with increase in the dopants concentration. The positive values of Hall coefficient indicate that holes are majority charge carriers in $Nd_{1-x}Sr_xFeO_3$ ($0 \leq x \leq 0.5$) systems. The increase in

localization length is also observed with increase in the Sr concentration. For both $Nd_{1-x}Sr_xFeO_3$ and $NdFe_{1-y}Ni_yO_3$ systems, the permittivity of grains is from 8-10 while the high permittivity observed at higher temperatures is extrinsic, due to the formation of different schottky barriers as a consequence of grain boundaries, ceramic surfaces and electrode-semiconductor contacts effects. With increase in the dopants concentration the dielectric constant first increases with increase in temperature and then decreases which may be due to inability of polarization process to complete as a result of thermal effects, increase of the delocalization of charge carriers and presence of inductive effects. The positive values of the $d\rho/dT$ indicate that the charge carriers are localized but the delocalization increases with increase in dopant concentration and temperature.

The magnetic data of $NdFeO_3$ and $Nd_{0.9}Sr_{0.1}FeO_3$ indicates that the Fe sublattices dominate at higher temperatures while at lower temperatures Nd sublattices play a significant role. The coercivity and remanence decrease while maximum magnetization at higher applied magnetic field increases with the decrease in temperature. The overall magnetic behavior changes from weakly ferromagnetic to paramagnetic with decrease in temperature.