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Micro-Structural, Magnetic and Electrical Studies of Nickel Doped $Y_3Ba_5Cu_8O_y$ (Y-358) Superconductors

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***Abstract*: The newly found $Y_3Ba_5Cu_8O_y$ (Y-358) superconductor was investigated for the replacement of copper by nickel. Samples of nominal composition of $Y_3Ba_5Cu_{(8-x)}Ni_xO_y$ (x = 0.00, 0.04, 0.08, 0.16, 0.80 and 8.00) were prepared by standard solid state reaction method. The structural and morphological characterization of the samples was carried out using X-ray diffraction (XRD), Scanning Electron Microscopy (SEM). The dielectric, magnetic and the transition temperature (T_c) measurements were carried out on the samples. The samples have a similar crystalline structure as that of very well-known Y-123 superconductor. The "c" axis values of the samples were found to be three times the "c" axis values of Y-123 superconductor. SEM studies on the samples indicated the presence some porosity. As the nickel amount increased, the T_c onset of the superconducting phase was found to be decreased.**

***Keywords*: High T_c superconductor, solid state reaction, XRD, SEM, Dielectric measurements, Magnetic measurements, T_c measurements, Nickel doping.**

I. INTRODUCTION

The discovery of superconductivity in $Y_1Ba_2Cu_3O_y$ in Y-123 [1] superconductors opened a new era to discover new materials with higher superconducting transition temperatures T_c. More recently, Aliabadi et al. [2] have reported a new superconductor $Y_3Ba_5Cu_8O_{18}$ (Y-358) of T_c above 100 K with the same crystalline structure as that of Y-123. Fisk et al. [3] and Hor et al. [4] have shown that in YBCO system superconductors, substitution at the rare earth site or alkaline-earth site would not change T_c considerably. But substitution at the oxygen site is very crucial in the formation of superconductors. According to the earlier studies done by Kistenmacher et al. [5] and Tarascon et al. [6] on the substitution, the effect on the plane sites reduce T_c more drastically. They found that the transition temperature T_c decreases with the increasing the substitution content due to a shift of Fermi – energy towards the mobility edges. One important fact to be noticed was that, whether the substitution ions are magnetic or non-magnetic does not make much difference in the depression in critical temperature T_c.

Over the last three decades, exhaustive study has been done on the substitution at the Cu site which has helped in understanding the mechanism of superconductivity (Iwasaki et al. [7], Akachi et al. [8], Nachumi et al. [9], A Rao [10]). Much

of literature appear to favour (Yamagata et al. [11], Maeda et al. [12], Shaked et al. [13]) that in Ni-doped YBCO systems, Ni ions have been generally thought to enter Cu (2) sites in the CuO_2 layers in the Y-123 materials. The results show that Ni ions enter Cu (2) sites at the low-doped concentration, as the concentration increases, a certain proportion of Ni ions begin to enter the Cu (1) sites until the appearance of the impurity phase. Even though Ni doping reduces T_c of the superconducting materials, but the colossal magnetism of the materials have got very important applications in the scientific field. In view of the above, Ni doped Y-358 system where Cu is replaced by Ni in a systematic manner are tried.

II. MATERIALS AND METHODS

Samples of nominal compositions of $Y_3Ba_5Cu_{(8-x)}Ni_xO_y$ (x = 0.0, 0.04, 0.08 0.16, 0.8 and 8.0) designated as (N1, N2, N3, N4, N5 and N6) were prepared by the standard solid state reaction method. Appropriate stoichiometric ratios of high purity (99.9%) powders of Y_2O_3, $BaCO_3$, CuO and $NiCO_3$ were mixed thoroughly and finely ground. The samples were taken in ceramic crucibles and calcined in air at 810°C for 17 hours in a tubular furnace. The powders were re-ground and pressed in the form of pellets of 10 mm diameter and about 2 mm thickness under the pressure of 5000 kg/cm². The pellets were then sintered at 920°C for 20 hours and then again for 21 hours with intermediate grinding and pelletisation.

III. RESULTS AND DISCUSSIONS

A. XRD Studies

The powder X-ray diffraction patterns of $Y_3Ba_5Cu_{8-x}Ni_xO\delta$ (x = 0.0, 0.04, 0.08, 0.16, 0.8, 8.0) samples are shown in Figure.1. The phase identification of the sample was performed by PHILIPS Pro analytical automated diffractometer with Cu-Kα radiation (λ = 1.5405Å) in angular steps of 0.033°. X-ray diffraction studies confirm the Orthorhombic phase with the space group Pmmm. The intensity diffraction peaks of samples have been found at (012), (015), (016), (0012), (113), (1012), (0017), (1019), (2017) etc. The orientations are mentioned in the Figure.1. The highest intensity diffraction peaks of samples have been found at 2θ = 32.807° for pure sample (N1) with

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(012) orientation. There is a minor shift in peak position with dopant concentration. However, small impurity peaks are observed in the spectrum, which may be due to impurities.
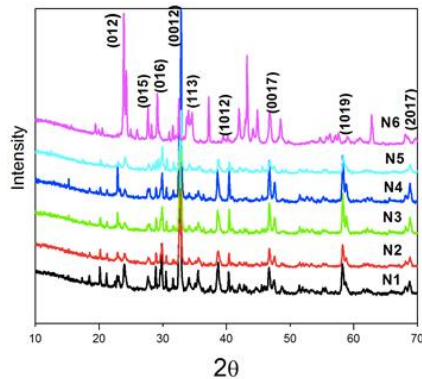

Fig. 1. X-ray diffraction patterns of $Y_3Ba_5Cu_{8-x}Ni_xO\delta$

The lattice parameters a, b, c, and unit cell volume V are obtained from the refinement process and shown in Table.1. The lattice parameters of Y-358 are compared with those for Y-123, which are reported earlier in the literature. The calculated values of a, b parameters for Y-358 are in agreement with the values of a, b parameters of Y-123. But the value of c parameter of Y358 is almost 3 times of the c parameter of Y-123. The c parameter shows linear relation with a number of Cu-atoms in the samples a.
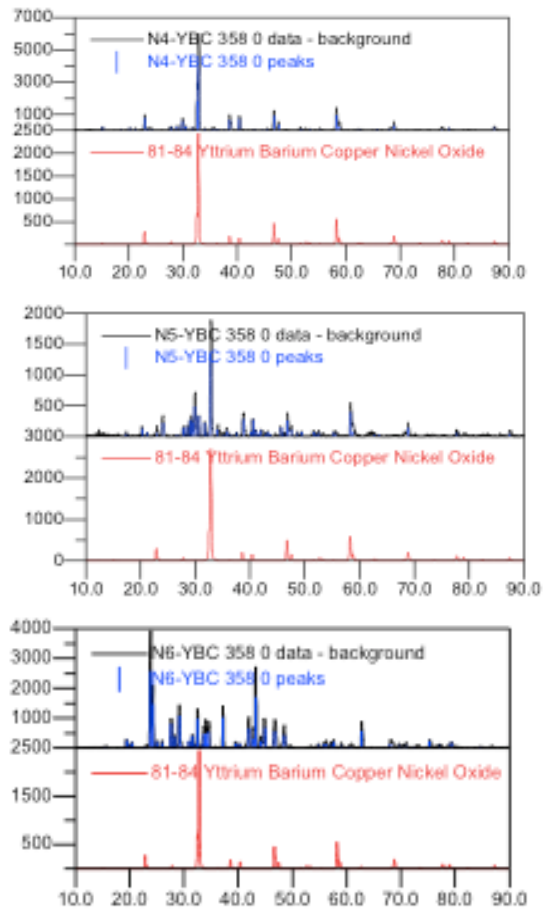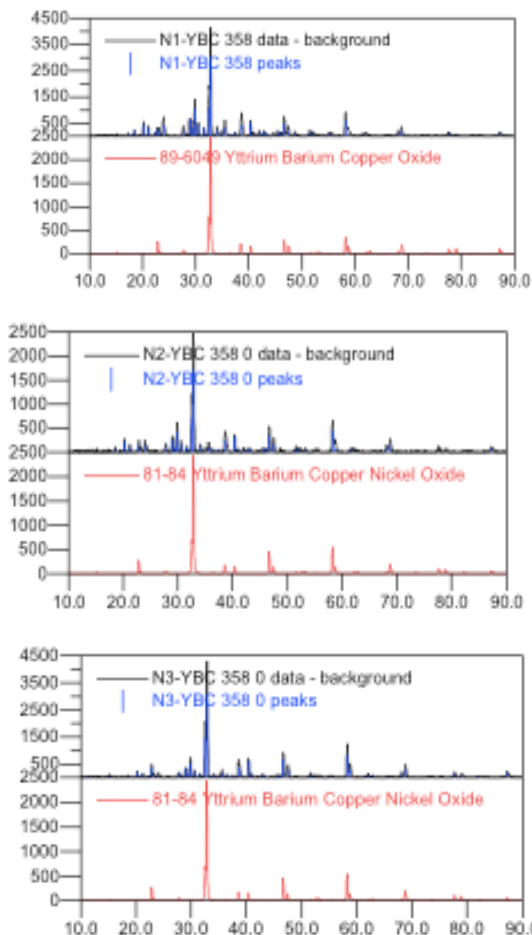

Fig. 2. XRD search matches

The XRD search matches for all samples are shown in Figure.2. From the figure it is evident that the main peaks of the Y-358 spectrum are the same, which also exist in Y-123. The average crystallite size of the powders were measured by X-ray line-broadening technique employing the Scherer formula,

$$D = K \lambda / (\beta_{1/2} \cos \theta)$$

Where D is average size of the crystallites, K is Scherer constant (K=0.9), λ is wavelength of radiation (1.5405A°), $\beta_{1/2}$ is full width at half maximum height (FWHM) measured in radians and θ corresponds to the peak.

B. SEM Studies

The morphology and compositional analysis was studied using SEM and Energy Dispersive Spectroscopy (EDS) attached with SEM. Figure.3 shows SEM images of all samples. SEM pictures confirm that the particles in all samples (Y-358 and Ni doped Y-358) are in nano size. However, nano order grain size did not alter T_C significantly, which may be due to the sintering. In the sintering process grains may be agglomerated and formed as bulk.
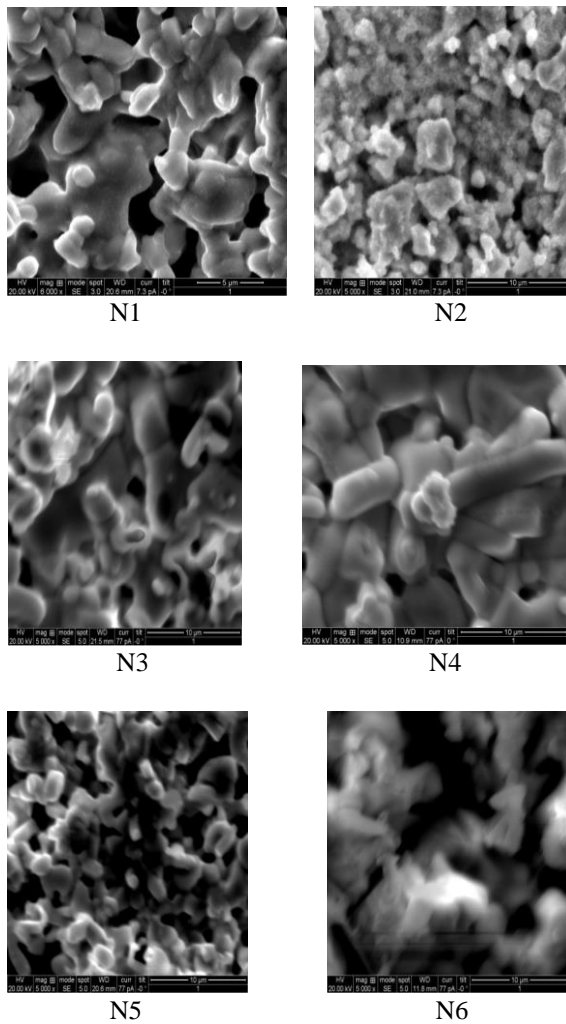
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Fig. 3. SEM images of the samples

Fig. 4. Energy Dispersive Spectra of the samples

C. EDS Studies

The compositional analysis was studied using Energy Dispersive Spectroscopy (EDS) attached to SEM made by Oxford instruments Inca, model Penta FET x3. Figure.4 shows the peaks corresponding to elements Y, Ba, Cu, Ni and O. EDS analysis shows that the samples contain only the target elements.
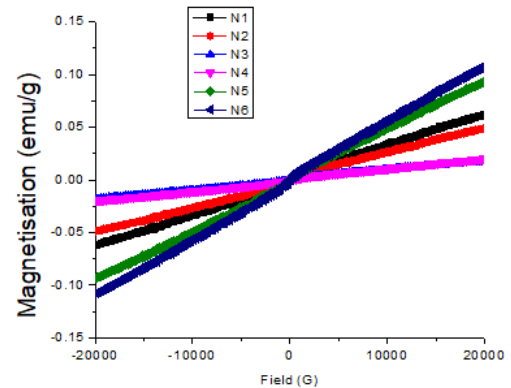

D. Magnetic Studies

Fig. 5. Graph of magnetisation versus applied magnetic field

Since the samples are Ni doped Y358, we studied magnetic properties also. M-H graph is shown in Figure.5. M-H graph clearly shows that N1, N2, N3, N4 are paramagnetic at room temperature, whereas N5, N6 show a very little ferrimagnetism at room temperature. That may be due to the effect more of dopant (Ni), which is ferromagnetic material.

E. Dielectric Studies

The variation of real and imaginary parts permittivity (dielectric constant and dielectric loss) as a function of frequency are shown in Figure 6 (a,b). At low frequencies the value of dielectric constant (ε') rapidly decreases with increase in frequency. At high frequencies it is almost constant. The sample N2 show the deviation, which may be due to impurities. The Dielectric loss (ε'') decreases exponentially at low frequencies but remains constant and attains almost same value for all the samples at high frequencies.
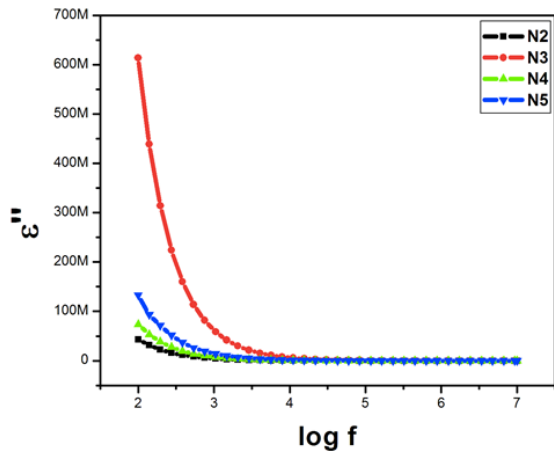
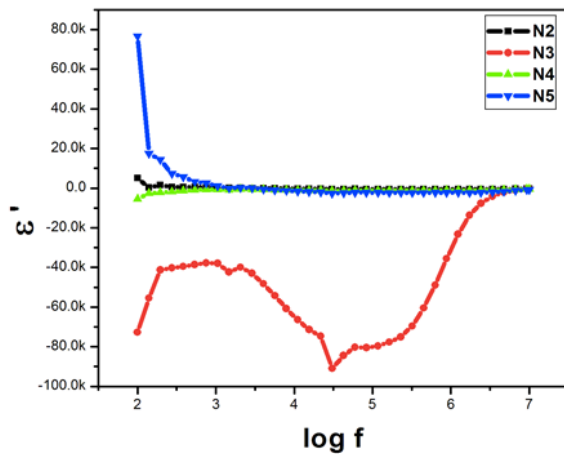Fig. 6a. Graph of real part versus frequency

Fig. 6b. Graph of imaginary part versus frequency
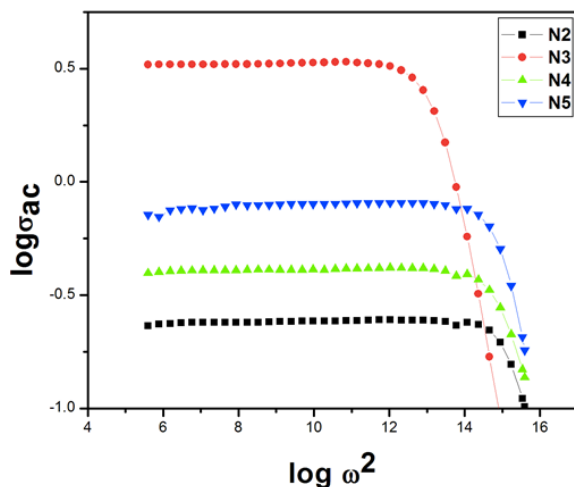
F. AC Conductivity Studies

Fig. 7. variation of a.c. resistivity at room temperature

The variation of a.c. resistivity at room temperature is shown in Figure 7. It is observed that, at low frequencies conductivity is almost constant. At high frequencies the conductivity almost drops to zero.

G. T_c Studies

The T_c of the samples was measured by self-inductance method using a frequency counter. Fig.8 shows the normalised frequency with T_c for $Y_3Ba_5Cu_{(8-x)}Ni_xO_y$ (x = 0.0, 0.04, 0.08 0.16, 0.8 and 8.0). It is noticed that the transition temperature T_c decreases nearly linearly with increasing Ni content indicates a homogeneous distribution of Ni in the sample. For x > 0.08, the superconducting signal given by the samples are very feeble and hence the T_c measurements were not able to be carried out. The above result is attributed to the fact that the magnetic moment of Ni, which suppresses the superconductivity through the cooper - pair breaking mechanism and change in the oxidation state of copper in the CuO_2 planes.
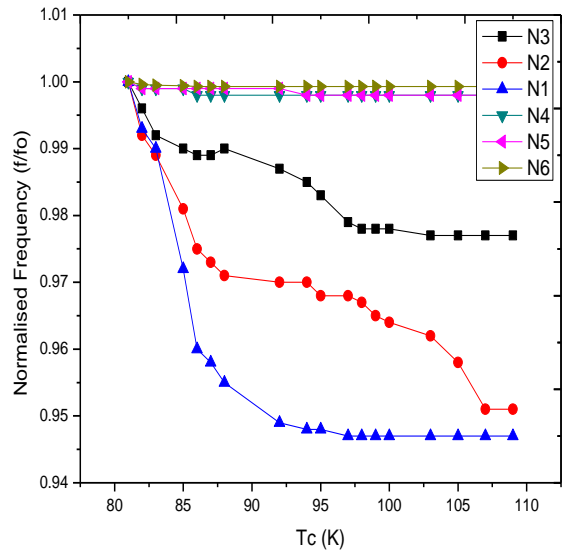

Fig. 8. Graph of normalized frequency versus T_c

IV. CONCLUSIONS

The effect of Nickel substitution for copper in Y-358 sample is attempted and samples are characterized by XRD, SEM, EDS, magnetic, dielectric and Tc measurements. All the samples showed sharp X-ray diffraction patterns with no impurity phases but sample N6 showed some unidentified peaks. X-ray emission spectroscopy results show that Nickel atoms enter the lattice of Y-358. The transition temperature Tc decreases with increasing dopant concentration when compared with the undoped sample. Samples N1-N5 showed paramagnetic properties and sample N6 showed soft ferrites nature at room temperature.

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