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Synthesis and Characterization of Fluorinated Superconducting $Y_3Ba_5Cu_8O_y$ Compound

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Abstract:
The effect of fluorine addition on morphological, structural and electrical properties of superconducting samples of composition $Y_3Ba_5Cu_8O_{y-x}F_x$ (x = 0.0, 0.2, 0.4 & 0.6) were investigated by X-ray diffraction (XRD), scanning electron microscopy (SEM) and transition temperature (T_c) measurements. The samples were synthesized by solid state reaction method. The XRD of the samples revealed a structure similar to that of Y-123 compound with about three time larger 'c' axis. The grain size calculated was found to increase with fluorine doping as indicated by SEM micrograph. As the fluorine amount increased, the T_c onset of the superconducting phase was found to be increased. Over all, the fluorine addition had improved the superconducting property of the samples considerably.

Keywords: High T_c superconductor, solid state reaction, XRD, SEM, T_c measurements, fluorine doping.

I. Introduction

The discovery of superconductivity in La-Ba-Cu-O system with a critical temperature (T_c) of 30 K by Bednorz and Müller [1] in the year 1986 led to a tremendous amount of research activity in the field of high temperature superconductors. Basically this was to understand the mechanism as well as to initiate search for new superconducting materials at still higher temperatures. The T_c of around 92 K was reported by Chu *et al*., [2] in a well known compound $Y_1Ba_2Cu_3O_7$ (Y-123) in 1987. The research in oxide superconductors was continued to bring out new aspects arising out of their structure and synthesis process. These ideas were soon adopted by many researchers to find out higher T_c materials in Y-Ba-Cu-O family. In the year 1988, Marsh *et al*., [3] reported that $Y_1Ba_2Cu_4O_{16}$ (Y-124) superconducts at 80 K. Bordet *et al*., [4] reported superconductivity in $Y_2Ba_4Cu_7O_{15}$ (Y-247) with T_c of 40 K. Fisk et al., [5] and Hor et al., [6] have shown that in Y-123 system superconductors, the substitution at rare earth site or alkaline-earth site will not change T_c considerably. But Ovshinsky *et al*., and Gupta *et al*., [7, 8] have reported that a substantial increase of T_c in Y-123 system by fluorine substitution at the oxygen site. More recently Aliabadi *et al*., [9] and Tavana *et al*., [10] have reported a new yttrium based high T_c superconductor $Y_3Ba_5Cu_8O_{15}$ (Y-358) with a T_c above 100 K. This prompted us to investigate the new superconductor for further enhancement of T_c by substituting fluorine for oxygen. In this paper, we report the results of our attempts to synthesize and characterize the pure and the fluorinated Y-358 superconductors.

II. Experimental

Samples of the nominal composition $Y_3Ba_5Cu_8O_{y-x}F_x$ (x = 0.0, 0.2, 0.4 & 0.6), designated as Y0, Y2, Y4 and Y6 were prepared by standard solid-state reaction technique. Appropriate stoichiometric ratios of high purity powders of Y_2O_3, $BaCO_3$, CuO and CuF_2 were mixed thoroughly and finely ground. The samples were taken in ceramic crucibles and calcined in open atmosphere at 810°C for 21 hours using 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 a pressure of 5000 kg/cm^2. The samples Y0 and Y2 were then sintered at 950°C for 20 hours and furnace cooled to room temperature. A slight melting was noticed on the surface of the sample Y2 and therefore all further sintering was carried out at lower temperatures. The pellets of all the samples were then sintered at 940°C for 24 hours and again at 930ºC for 24 hours with one intermediate grinding and pelletisation. The XRD data was recorded using a compact PHILIPS Pro analytical automated diffractometer with copper Kα source (λ = 1.5405 Å) in the two theta ranges from 10º to 100º. The grain morphology of the fractured surface of the samples was analyzed using scanning electron microscope (Quanta ASM 840A). The T_c of the samples was determined by self inductance method using a Colpitt's oscillator and a frequency counter. The temperature of the sample was recorded using a calibrated chromel – alumel thermocouple with an accuracy of ±1ºC.

III. Results and discussion

The X-ray diffractograms of the samples Y0 and Y2 along with that of standard Y-123 are shown in Fig.1, and that of the samples Y4 and Y6 are

shown in Fig. 2. As seen from the figure, most of the peaks of Y0, Y2, Y4 and Y6 are identical with that of Y-123. This observation is similar to that of Udomsamuthirun *et al.*, [11]. The prominent peaks of the samples were indexed in an orthorhombic system. The 'c' value of our samples is almost 3 times the 'c' value of Y-123 sample [Table-1]. This result is similar to the one reported by Aliabadi *et al*., [9]. But we observed a prominent peak at $2\theta \approx 15^{o}$ which was not accounted by Aliabadi *et al.* The scanning electron microscope (SEM) images of the samples are projected in Fig. 3. The surface images of the samples by SEM revealed the presence of pores. Also it is observed that the average grain size, calculated from XRD data using Scherrer formula: $t=0.89*\lambda/B*\cos\theta B$ (where t is the grain size, λ is the wavelength of the X-ray, θB is the Bragg angle and B is the FWHM), increases as the fluorine content increases and reaches a saturation value at x = 0.2 (Fig.4). Lattice parameter 'b' versus fluorine composition graph is shown in Fig.5. The plots of frequency versus temperature of the samples are shown in Fig.6. As seen, the $T_{c\ onset}$ of the samples have increased with increase in fluorine amount and a saturation value is reached at about x = 0.6 (Fig. 7). This can be explained as due to optimization of the oxygen content in the sample by fluorine substitution. The T_c of Y-Ba-Cu-O superconductor is known to vary with oxygen stoichiometry and is found to be a maximum for a particular value of oxygen content (Rao et al., [12] Namgung et al., [13] and Narottam P.Bansal et al., [14]). The T_c value decreases, for oxygen content above or below this optimum value. By the fluorine substitution, the optimum value of oxygen content is attained at which $T_{c\ onset}$ is maximum. Increase in fluorine amount beyond x = 0.6 may not further increase the $T_{c\ onset}$ value since that will disturb the optimum oxygen stoichiometry.

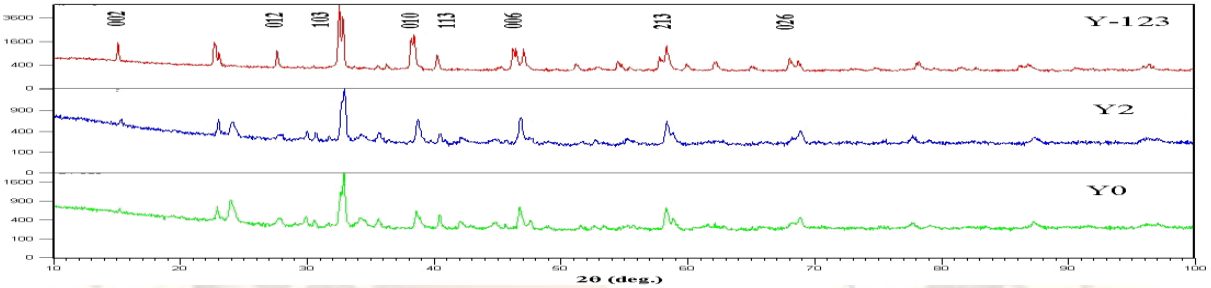

Fig. 1, XRD spectra of samples Y-123, Y2 and Y0

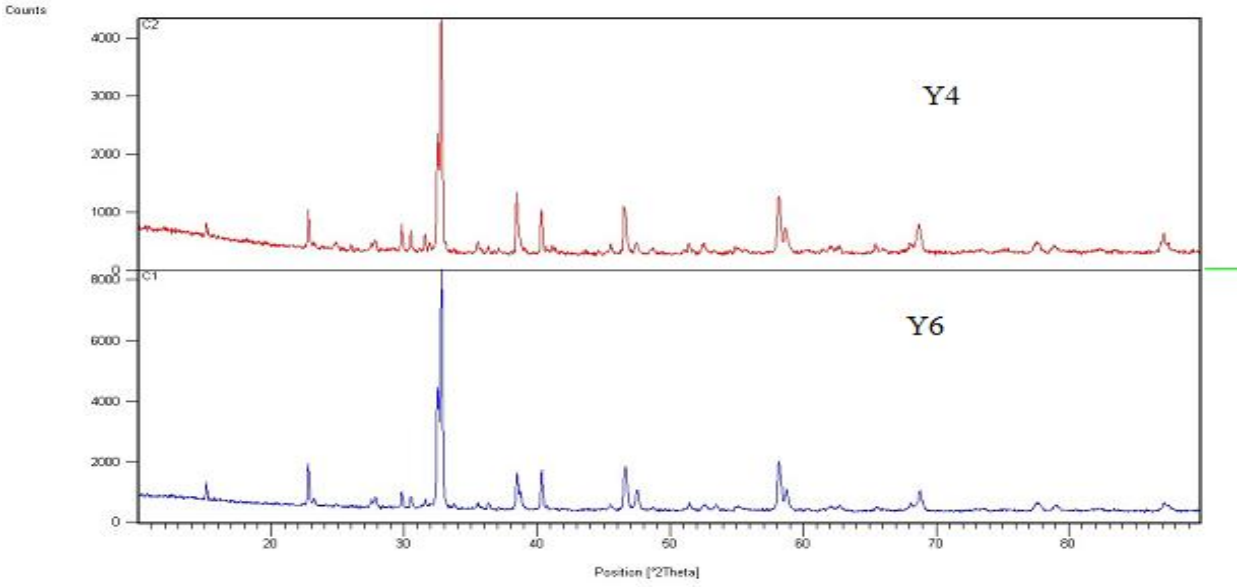

Fig. 2, XRD spectra of samples Y4 and Y6

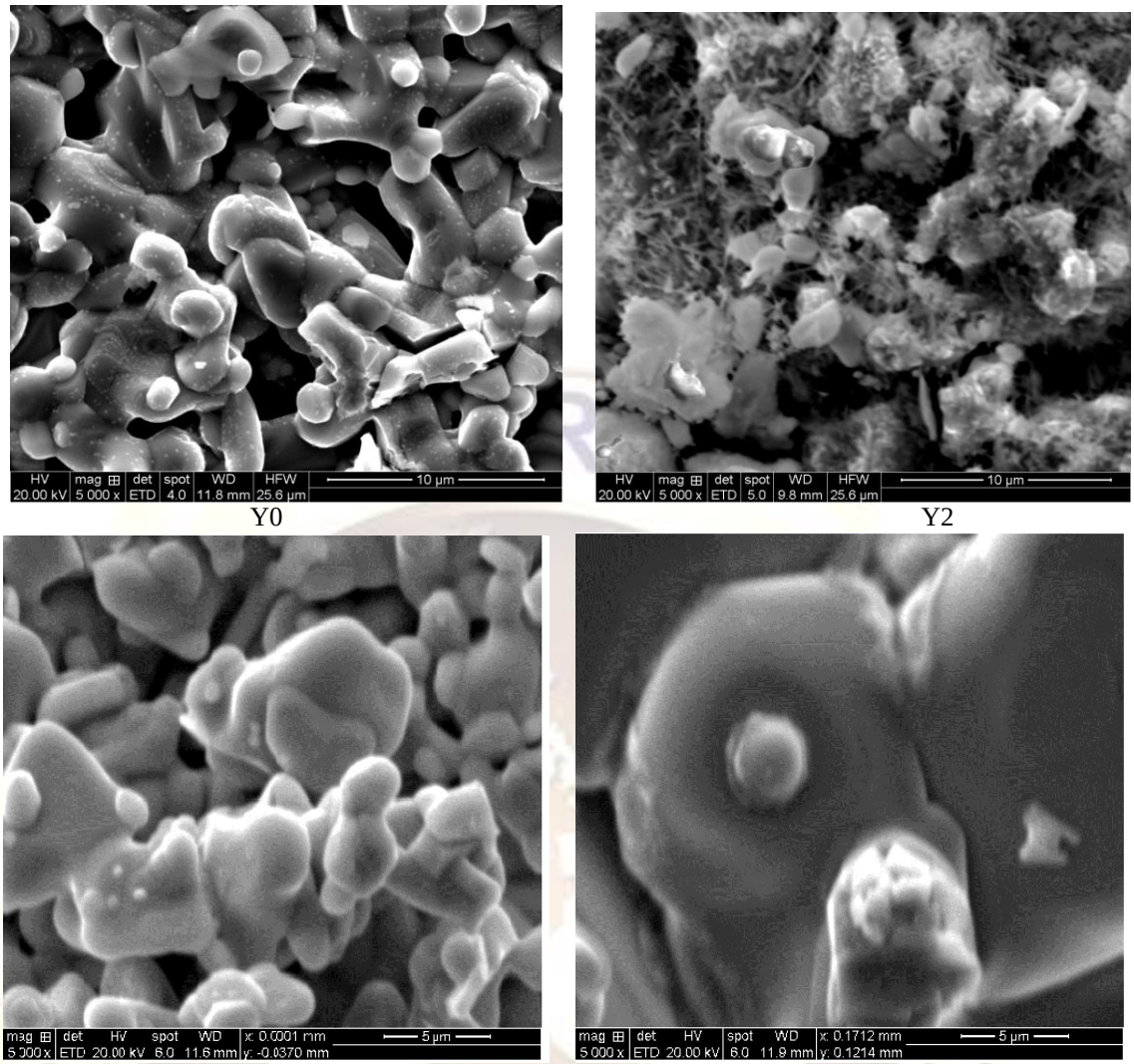

Fig. 3, SEM micrographs of the samples (Magnification of 5000 and marker length 10, 5 µm)

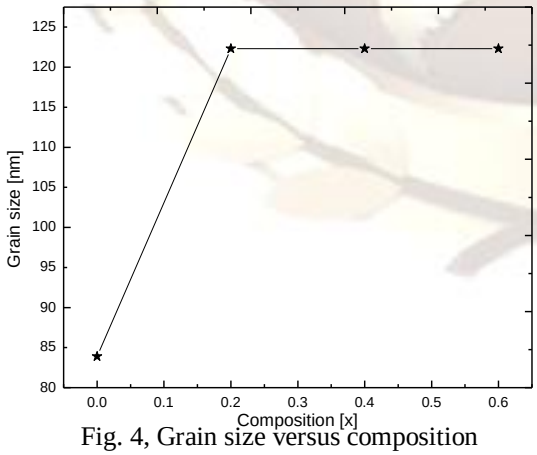

Fig. 4, Grain size versus composition

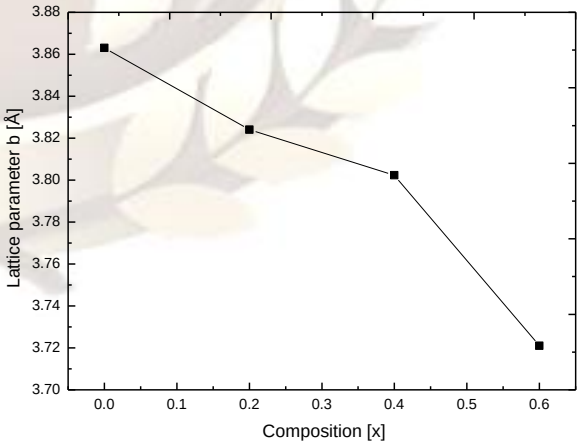

Fig. 5, Lattice parameter versus composition

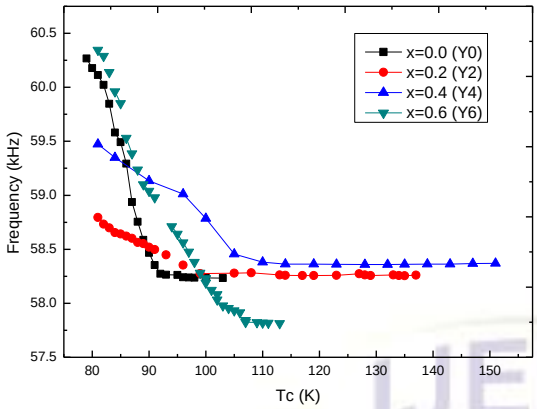

Fig. 6, T_c curves of samples Y0, Y2, Y4 and Y6

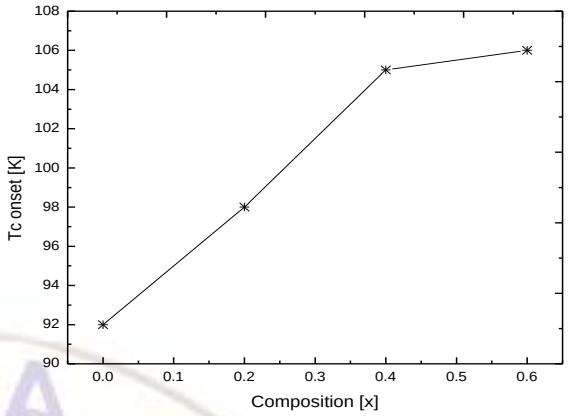

Fig. 7, T_c onset versus composition

Table.1, T_c and Lattice parameters details of the samples

Compounds	Y0 (x = 0.0)	Y2 (x = 0.2)	Y4 (x = 0.4)	Y6 (x = 0.6)
$T_{c\ onset}$ (K)	92	98	105	106
a (Å)	3.902	3.942	3.863	3.871
b (Å)	3.824	3.863	3.802	3.721
c (Å)	30.690	31.382	31.330	32.499
Cell volume $(Å)^3$	457.933	477.956	460.188	468.115

IV. Conclusions

This study confirms the formation of Y-358 superconducting phase. The crystalline structures of all the samples are similar to that of Y-123 but for the 'c' axis which is almost three times the 'c' axis of Y-123. Fluorine Doping can be used to optimize the oxygen stoichiometry and thereby enhancing the $T_{c\ onset}$ value. It is noteworthy that the fluorine doping not only improved superconducting behavior of the Y-358 phase but also decreased the melting point of the sample.

V. Acknowledgements

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