



Study of Suppression of Superconducting Transition Temperature of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ System

KEYWORDS

$Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$; Praseodymium; X-ray diffraction; Transition temperature

Prachi Yadav

* Sandeep Kumar

Ravish Goutam

Department of Physics,
Kirori mal
College, University of Delhis, India

Department of Physics,
Bhaskaracharya College
of Applied Sciences,
University of Delhi, India
* Corresponding author

Reactor Physics Section, Nuclear
Power Corporation of India Limited,
Gujarat, India.

ABSTRACT The effect of Pr-doping on the transition temperature (T_c) and structural properties of $YBa_2Cu_3O_{7-\delta}$ (YBCO) superconductor have been investigated. The sol-gel method has been employed for the synthesis of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ (YPBCO) samples with the compositions ($x=0.0, 0.05, 0.10, 0.20$ and 0.30). The broadening in X-ray diffraction (XRD) peaks has been used to calculate the micro-strain by the Williamson-Hall (W-H) plot. The broadening in XRD peaks and micro-strain are found to increase with increase in Pr concentration x which could be representatives for some disorder in Cu-O planes. The resistance-temperature measurements of as prepared YPBCO samples show the monotonic suppression of T_c with increase in Pr concentration from $x=0$ to $x=0.30$. The experimental T_c values were in good agreement with theoretical T_c values calculated by the inclusion of disorder effects along with magnetic pair breaking and hole filling effects. It has been suggested that disorder in Cu-O planes might be strain-induced. The disorder effects along with magnetic pair breaking and hole filling effects are responsible for the suppression of T_c in YPBCO system.

Introduction

YBCO was the first superconductor to have transition temperature (T_c) above boiling temperature of Liquid Nitrogen¹. Its superconducting properties are not affected much by the substitution of Y with the rare earth (RE) elements like La, Nd, Sm, Eu and Gd etc. however, Praseodymium (Pr) is the exception^{2,3}. The Resistance-temperature measurements of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ system clearly indicate the influence of Pr-doping on electrical properties of YBCO⁴. The Pr-doped YBCO has potential applications as superconducting wires due to its enhance critical current density (J_c) in the magnetic field at liquid nitrogen temperature. The superconducting transition temperature (T_c) decreases monotonically with increase in Pr concentration in YPBCO system. Different mechanisms based on Pr valence have been suggested for the monotonic suppression of T_c in YPBCO system with increasing x . The magnetic property measurements⁵ revealed the tetravalent (+4) state of Pr, and suggested that hole filling effect is responsible for suppression of T_c in YPBCO system. The optical measurements^{7,8} indicate the trivalent state (+3) of Pr, and believed that superconducting pair breaking by the local moment of the Pr ion is responsible for suppression of T_c in YPBCO system⁹. In trivalent state (+3) of Pr, The degradation of T_c in YPBCO system with increasing Pr concentration follows the Abrikosov-Gorkov relation¹⁰. The variation of T_c (in Kelvin) with x and y in $(Y_{1-x}Ca_x)Pr_yBa_2Cu_3O_{6.95\pm 0.02}$ system can be represented by¹¹

$$T_c(x, y) = 97 - 425(0.1 - 0.95x + y)^2 - 96.5x \dots \dots \dots (1)$$

The equation (1) suggested that the T_c values depend on two terms (i) the linear term due to magnetic pair breaking mechanism and, (ii) the quadratic term due to hole filling effect. Further, the theoretical T_c (in Kelvin) of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ system for various values of x has been calculated by J. J. Neumeier et al¹¹ using Abrikosov-Gorkov¹⁰ (A-G) relation

$$T_c(x) = T_{c0} - Bx \dots \dots \dots (2)$$

Where T_{c0} and B . The equation (2) contains only linear term in x which is attributed to magnetic pair breaking mechanism.

The T_c (in Kelvin) for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ system according to Dole¹² is given by

$$T_c(x) = 97 - 425(0.1 - 0.95x)^2 - 96.5x \dots \dots \dots (3)$$

The equation (3) also suggested the degradation of T_c due to magnetic pair-breaking mechanism and hole filling effects. The Gill et al.¹³ reported the variation of T_c (in Kelvin) with x in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ system by the relation

$$T_c(x) = 97 - 425(0.1 - 0.95x)^2 - 96.5x - 15.4\sqrt{x} \dots \dots \dots (4)$$

Where initial quadratic and linear terms have same dependence as given by equation (3) but third square root term is due to disorder in Cu-O planes after doping with Pr.

In the present work, we synthesized the series samples of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ superconductor with various values of x . The samples were characterized by various techniques and the results are presented in this paper.

Experimental

The samples of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ ($x=0, x=0.05, x=0.10, x=0.20$ and $x=0.30$) were prepared by using sol-gel method. The appropriate amounts of $Y(NO_3)_3$, $Ba(NO_3)_2$, $Cu(NO_3)_2$ and $Pr(NO_3)_3$ were mixed in the solution of water and ethyl alcohol. The mixture was stirred at 50-70°C until it became gel. It was dried in oven, then grinded and put in the furnace at 500 °C to eliminate moisture and nitrates for 5 h. The material was then regrinded and calcined in air at 900 °C for about 20 h. It was cooled and pressed in to pellets. Subsequently, the pellets were sintered at 930 °C for 20 hours in the presence of oxygen. The transition temperature T_c of as prepared samples was determined from resistance-temperature measurements by means of standard four probe method. The structure of the samples was characterized using XRD technique. The size and surface morphology of grains were determined from SEM measurements.

Results and Discussions

Fig. 1 shows the variation of relative resistance with temperature in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ ($x=0, x=0.05, x=0.10, x=0.20$ and $x=0.30$) samples. The transition temperature T_c for various

concentrations of Pr is calculated using fig.1, eq. (2), 3 and 4 are shown in table-1 and found in good agreement with reported values^{12,14}. The variation of experimental T_c with Pr concentration x is shown the fig. 2. The monotonic suppression of T_c with increase in Pr concentration was observed. Further, the various values of transition temperature calculated from equations (2), (3) & (4) along with experimental transition temperature is plotted against Pr concentration x in fig. 3. The transition temperatures calculated from eq. (4) are found in good agreement with experimental T_c values. From fig. 3, this is evident that there is some disorder present in the system.

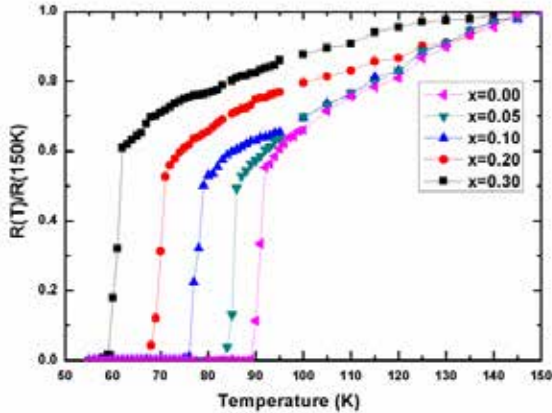


Fig. 1. Resistance-Temperature variation in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples for various values of x .

Table 1 Comparison between experimental T_c and theoretical T_c calculated from Eq. (2), (3) & (4);

Pr Conc. x	Experimental T_c (K)		Theoretical T_c (K)		
	$T_{c\ onset}(K)$	$T_{c\ zero}(K)$	Eq. (2)	Eq. (3)	Eq. (4)
0.00	92	87	97	92.75	92.75
0.05	86	82	92.18	88.12	84.68
0.10	79	74	87.35	83.87	79.00
0.20	71	67	77.70	76.07	69.18
0.30	62	57	68.05	67.96	59.52

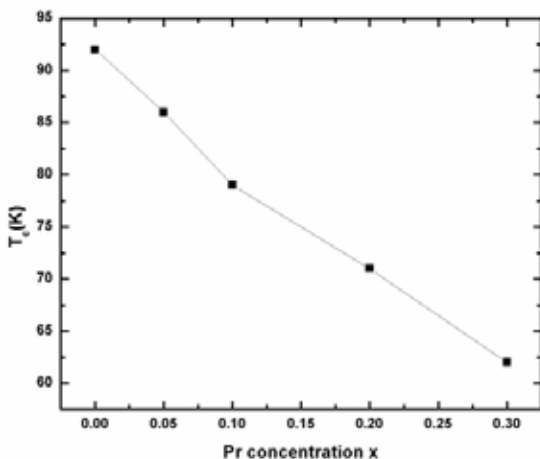


Fig. 2. Variation of experimental T_c values in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples for various values of x .

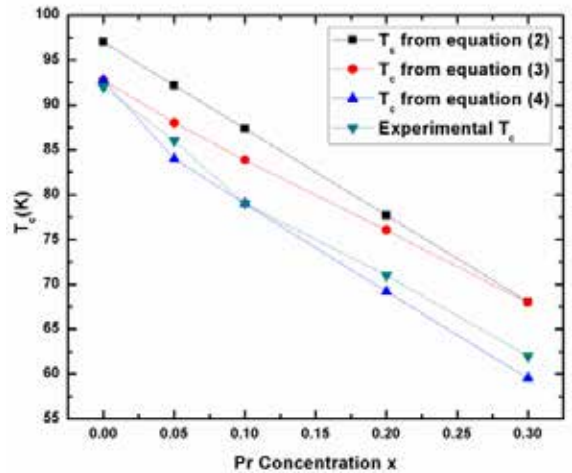


Fig. 3. Plot of T_c values from experiment and equations (2), (3) & (4) in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples for various values of x .

Fig. 4 shows the XRD measurements of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples for various values of x . It shows that all the samples have the orthorhombic Perovskite structure and contain no extra peaks due to impurity phases within the experimental error. The lattice parameters calculated for each sample from the diffraction peak positions (2θ) are listed in table-2. The variation of lattice parameters with the Pr concentration x is shown in the fig. 5. From XRD pattern, it is evident that lattice parameters a and b first decreases and then increases with x . On the other hand, lattice parameter c increases monotonically with x . The volume of the unit cell first decreases for $x=0.05$ and then monotonically increases up to $x=0.30$ ¹². The results clearly show that the distance between the two Cu-O planes increases and Cu-O planes bend towards the Ba-O planes with increase in Pr concentration¹⁵.

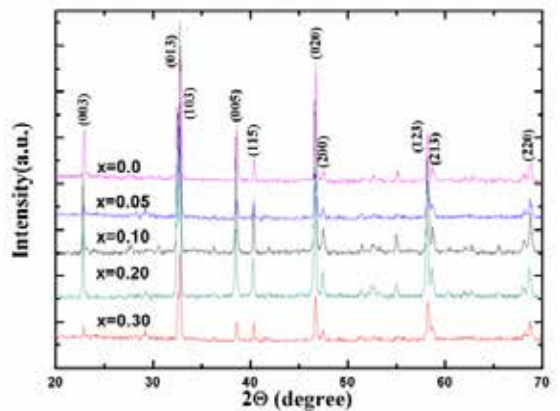


Fig. 4. XRD patterns of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples for various values of x .

TABLE 2 Lattice parameters for various values of x in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples;

Pr Conc. (x)	$a(\text{Å})$	$b(\text{Å})$	$c(\text{Å})$	Unit cell volume (Å^3)
0.00	3.822	3.885	11.645	172.91
0.05	3.827	3.876	11.654	172.87
0.10	3.826	3.885	11.679	173.60
0.20	3.830	3.898	11.686	174.46
0.30	3.830	3.893	11.705	174.52

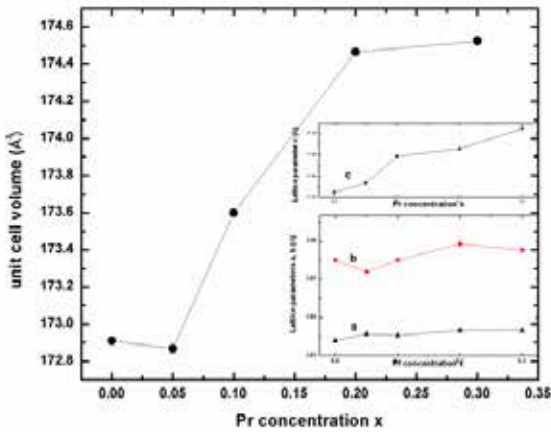


Fig. 5. The variation unit cell volume in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples for various values of x. Inset: variation of lattice parameters a, b and c with x.

Fig. 6 shows the variation of the full-width half maximum (FWHM) of the (005) peak in YPBCO system and, confirms that the FWHM monotonically increases with Pr concentration x¹⁶. From the results, it can be said that Pr-doping influences the structure of entire unit cell. The FWHM of XRD peaks can be expressed as linear sum of FWHM of size, strain and Instrumental¹⁷.

$$\beta = \beta_{size} + \beta_{strain} + \beta_{Instrumental} \dots\dots\dots (5)$$

Here, in order to find the strain broadening each XRD peak fitted with Lorentzian profile. The micro-strain, ϵ and the average grain size, D can be calculated from Williamson-Hall (W-H) plot for the X-ray diffraction peak broadenings¹⁸,

$$\beta \cos\theta = 4\epsilon \sin\theta + \lambda/D \dots\dots\dots (6)$$

Where λ is wavelength of incident X-rays. In fig. 7, plotted against $4\epsilon \sin\theta$ and linearly fitted to find micro-strain ϵ . The micro-strain ϵ increases with increase in Pr concentration x as shown the fig. 8. Further, fig. 9 shows that transition temperature T_c of YPBCO system decreases monotonically with increase in micro-strain ϵ . The variation of T_c with micro-strain ϵ has not been reported elsewhere and, suggested that micro-strain ϵ is a good variable to define the suppression of T_c along with magnetic pair-breaking and hole filling effects. A closer correlation can be found between micro-strain ϵ and transition temperature T_c compared with the correlation between the disorder present in Cu-O planes and transition temperature T_c .

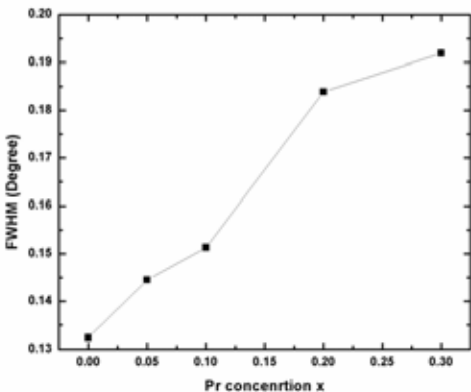


Fig. 6. The variation of FWHM for (005) peak in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples for various values of x.

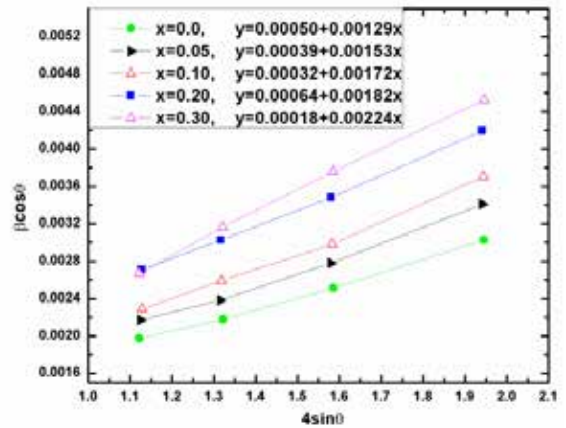


Fig. 7. Plot of versus of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples for various values of x.

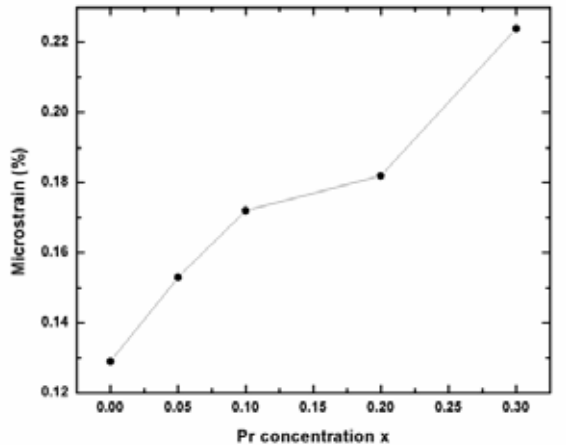


Fig. 8. Plot of microstrain (ϵ) versus Pr concentration x.

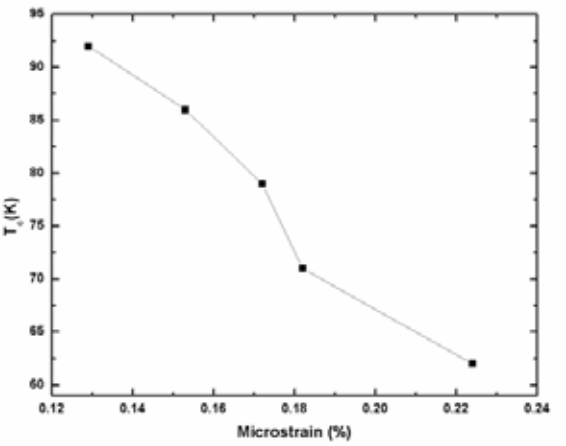


Fig. 9. Plot of transition temperature T_c versus microstrain ϵ (%).

From these results, it can be said that the disorder in Cu-O planes might be strain-induced. Fig. 10 shows the SEM of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples with (a) x=0, (b) x=0.05, (c) x=0.10, (d) x=0.20 and (e) x=0.30. The average size of the grains calculated from SEM images ranges from 4 to 15 μm . The EDAX

analysis of grains in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples for the range $0 \leq x \leq 0.30$ revealed that the various elements were present in the appropriate proportion.

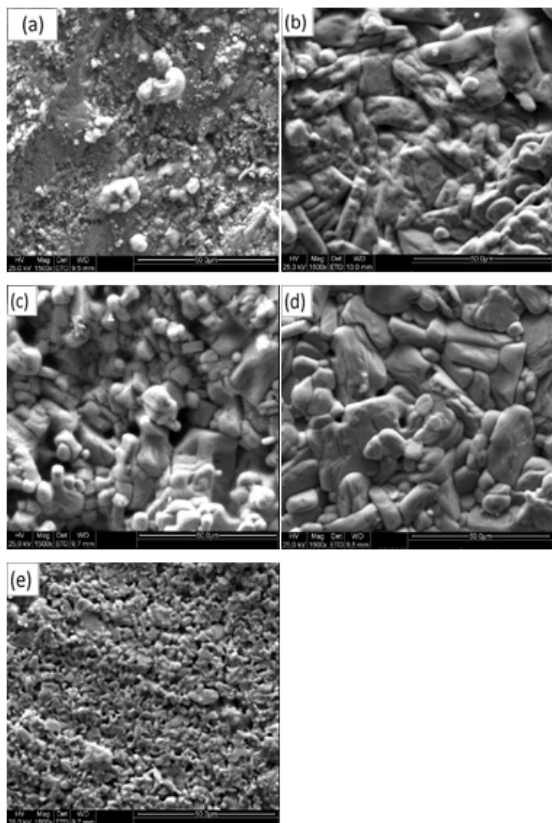


Fig. 10. SEM of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples; (a) $x=0.0$, (b) $x=0.05$, (c) $x=0.10$, (d) $x=0.20$ and (e) $x=0.30$.

Conclusions:

The effects of Pr-doping on the superconducting transition temperature T_c and XRD patterns of YBCO samples have been investigated. The monotonic suppression of T_c was observed with increase in Pr-doping. The broadening in XRD peaks was found to increase with increase in Pr-doping and, has been used to calculate the micro-strain ϵ . We have observed an increase of ϵ and a decrease of T_c with increase in Pr-doping. Our experimental T_c values were in good agreement with theoretical T_c values calculated from equation (4) which includes the disorder term along with the magnetic-pair breaking and hole filling terms. Probably, the theoretically predicted disorder in Cu-O planes was strain-induced. The more experimental studies will be required to correlate the micro-strain and disorder effects in YPBCO system. The present work emphasizes on the strain-induced disorder effect, along with the other effects, to explain the suppression of T_c in Pr-doped YBCO superconductor.

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