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THE STRUCTURAL STUDY OF Pr SUBSTITUTED Eu-123 HIGH T_C CUPRATE SUPERCONDUCTORS

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Abstract- We have investigated $Eu_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ high purity samples with compositions x = 0.0, 0.05, 0.15, 0.25, 0.35 and 0.45 by well known solid state reaction route. The XRD study of all samples is carried out. The XRD studies of the samples prepared by solid state reaction route show EuPr-123 system have an orthorhombic pervoskite structure. The lattice parameters, oxygen content, volume of unit cell, orthorhombicity, X-ray density and hole concentration were evaluated using XRD data. It is found that the lattice parameters and volume of unit cell increase while oxygen content orthorhombicity, X-ray density and hole concentration decrease as doping percentage of Pr increases. We report such qualitative results of Pr substituted Eu-123 in this paper quantitatively.

Keywords–HTSC, lattice parameters, oxygen content, orthorhombicity, hole concentration, X-ray density, superconductivity, transition temperature

INTRODUCTION

The suppression effect of superconductivity by the introduction of Pr in the rare earth site in R-123 high T_c cuprates have been widely investigated [1-3]. The substitution of rare earth element except (Ce, Pr, Pm and Tb) in the place of R in $RBa_2Cu_3O_{7-\delta}$ (R-123) gives rise to superconductivity with nearly equal to the critical temperature T_c = 90 K [4-5]. However, interesting exception is that the Pr-123 system which forms the same orthorhombic structure as Y-123 and other rare earth compounds but it is an insulator instead of superconductor [6-8]. There are some theoretical models such as hole filling [9-11], hole localization in CuO_2 plane [12-16], pair breaking [17-19] that have explained the suppression of T_c and superconducting insulator transition in these RPr-123 compounds. The experimental data shows that the occurrence of orthorhombic tetragonal transition at critical value is due to the enhancement of oxygen deficiency in the CuO chains [20]. To understand superconducting properties of Pr substituted Eu-123 system and oxygen deficiency, we have characterized the samples by X-ray diffractometer and explained qualitatively in this paper.

EXPERIMENTAL

The samples of $Eu_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ with compositions x = 0.0, 0.05, 0.15, 0.25, 0.35 and 0.45 were prepared by conventional solid state reaction route. The appropriate mixtures of high purity (Aldrich make 99.99%) Eu_2O_3, $BaCO_3$, Pr_6O_{11} and CuO were used. The powders were mixed thoroughly, ground for 3h and calcined twice at 915°C for 24h with intermediate grinding for 3h. The calcined samples are ground for 2 h for making pellets and finally sintered at 930°C for 24 h. These pellets were annealed in oxygen atmosphere for 24 h at 450°C followed by slow cooling at 1°C/min upto room temperature.

RESULTS AND DISCUSSION

X-ray diffraction patterns were recorded at room temperature by X-ray diffractometer (Model:PW-3710) using CuK_α radiation and only high intensity peaks are indexed as shown in fig. (**1**).

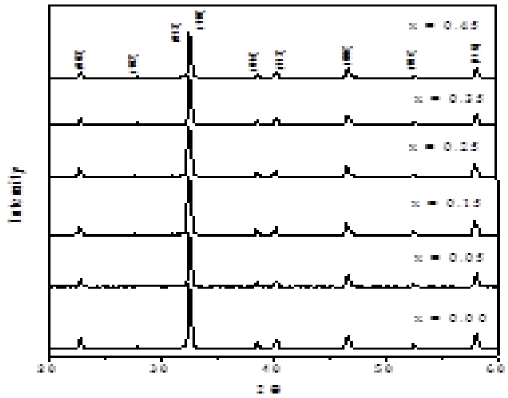

Fig. **1-** XRD patterns of $Eu_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ system.

The X-ray diffraction study of all samples reveals that they have the orthorhombic pervoskite structure with no evidence of impurity phase, suggesting that the preparation process was successful according to the stoichiometrical expectations and the material is in single phase form with homogeneous powder of finer particle size. The peak intensity goes on decreasing with increasing Pr concentration. The structural data obtained from XRD patterns are tabulated in table1. For all the Pr composition X-ray diffraction patterns correspond to that of the orthorhombic structure. The lattice parameters 'a'

& 'b' are increased while the lattice parameter 'c/3' increases gradually with increasing Pr content are shown in fig. (**2**).

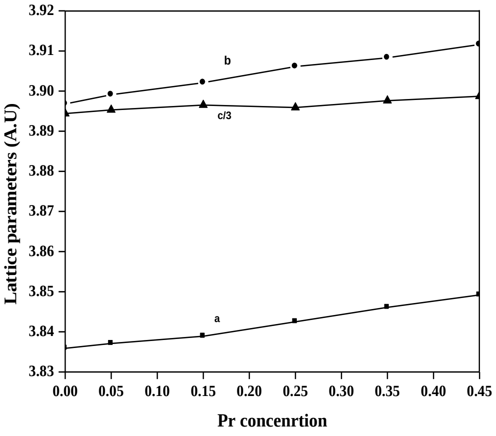

Fig**. 2-**Lattice parameters vs Pr concentration

The oxygen content plays an important role in assigning the orthorhombic to tetragonal structure in the HTSC. An δ = 0.00 stoichiometry is only reached if samples are slowly cooled in oxygen atmosphere [21-22]. The oxygen deficiency of the samples is calculated by X-ray diffraction data using the formula [23]. The values of oxygen deficiency (δ) for each composition have been obtained and amount of oxygen content of each sample is listed in table 1.

It is observed that the average oxygen content is found ≈ 6.86 for all samples with minimum value 6.82 and maximum value 6.91. It is observed that the oxygen content depends on the lattice parameter 'c' and almost independent for Pr content in the samples The values of oxygen content for each composition have been obtained as shown in fig. (**3**).

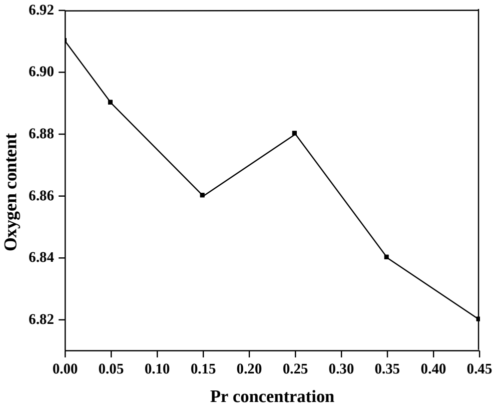

Fig**. 3-** Oxygen content vs Pr concentration.

From table 1, it is observed that the volume cell goes on increasing with increasing Pr concentration. It is shown in fig. (**4**).

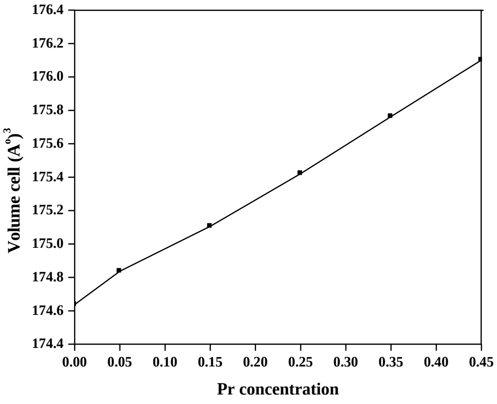

Fig. **4-** Volume cell vs Pr concentration.

From fig. (**4**), it is observed that this may be related to the ionic radii of Pr ion and Eu ion. The enhancement of volume cell with increasing Pr content shows that the Pr^{3+} goes to Eu^{3+} site. This may be due to the ionic radius of Pr^{3+} which is greater than that of Eu^{3+}. The orthorhombicity against Pr content is shown in fig. (**5**).

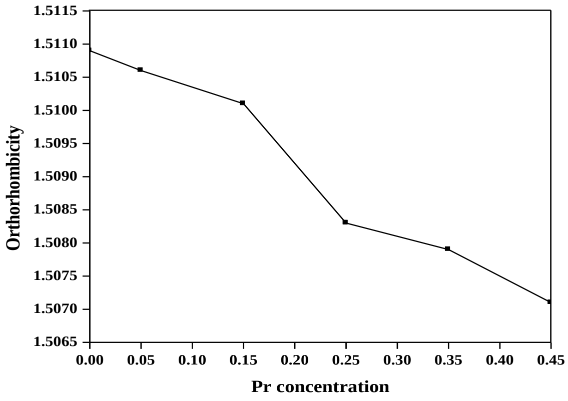

Fig**. 5-** Orthorhombicity vs Pr concentration.

It is observed that the orthorhombicity decreases with increasing Pr content. The orthorhombic distortion slowly decreases from x = 0.00 to x = 0.45 with increasing x.

The X-ray density is calculated from X-ray diffraction data by the formula [24] and listed in table1. The X- ray density plotted against Pr concentration is shown in fig. (**6**).

X-ray density goes on decreasing with increasing Pr concentration. This may be due to the change in molecular weight which reflects the change in X-ray density. The molecular weight of given system goes on decreasing with increasing Pr concentration resulting the reduction in X-ray density. This may be due to the ionic radii of Pr and Eu.

It is well known that in the family of 123 HTSCs, the oxygen deficiency affects the electron transport, crystal structure and other superconducting properties [25]. It is also well known that the 123 family of HTSCs has been related to the decrease of hole concentration in the CuO_2 planes due to the removal of oxygen from the charge reservoir chains [26]. The hole concentration in the CuO_2 plane of EuPr-123 samples is calculated using the relation [27], by assuming the charge neutrality of the unit cell.

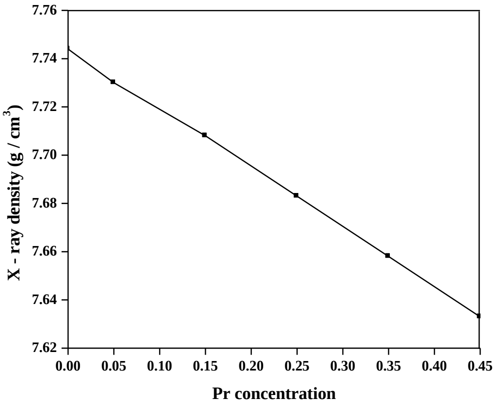

Fig. **6-**X-ray density vs Pr concentration.

$$P_{sh} = \left[\frac{0.5-2\delta-(Q_{\mathrm{Pr}}-3)x}{2}\right] \quad \ldots\ldots\ldots\ldots\ldots(1)$$

The hole concentration is calculated using formula (1) and values of hole concentration are listed in table 1. The hole concentration against 2x+δ for $Q_{Pr} = 4$ is shown in fig. (**7**).

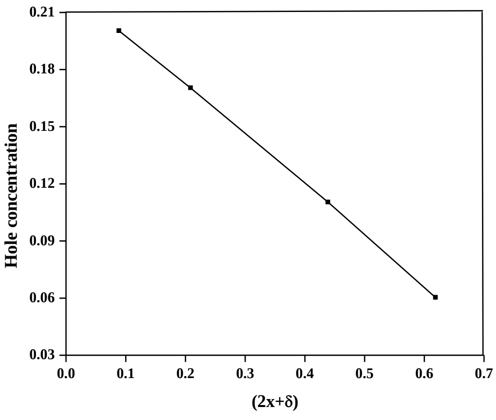

Fig. **7-** Hole concentration vs (2x + δ).

From this figure, it is observed that, the hole concentration has a linear dependence to (2x+δ) for each value of x. The linear behavior indicates the correlation between oxygen deficiency and the Pr doping value for EuPr – 123. The oxygen deficiency goes on decreasing with x which affects the hole concentration. It may be a possible cause for changing the superconducting properties of the compound.

CONCLUSION

The samples of $Eu_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ system were synthesized by solid state reaction route. From XRD data it is confirmed that all samples are in orthorhombic pervoskite structure. It is found that the lattice parameters and volume of unit cell increases linearly. This may be due to the oxygen deficiency and ionic radii of Pr^{3+} and Eu^{3+}. The orthorhombicity, X-ray density and hole concentration decrease as doping percentage of Pr increases. It is also observed that, the oxygen content depends on the lattice parameter 'c' and almost independent for Pr content in the samples. The hole concentration has a linear dependence to (2x+δ) for each value of Pr content. It may be a possible cause for changing the superconducting properties of the compound.

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Table 1-The lattice parameters, oxygen content, oxygen deficiency, volume cell, orthorhombicity, X-ray density and hole concentration of the $Eu_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ *samples.*

Pr Conc.	**Lattice parameters in A°**			**Oxygen content (7 - δ)**	**Oxygen Deficiency (δ)**	**Volume Cell (A°)^3**	**Orthorombi-city e = c / (a + b)**	**X - ray density in (g /cm^3)**	**Holes in CuO**
	A	**B**	**c**						
0.00	3.8359	3.8968	11.6832	6.91	0.09	174.6374	1.5109	7.744	0.20
0.05	3.8371	3.8991	11.6860	6.89	0.11	174.8370	1.5106	7.730	0.17
0.15	3.8389	3.9021	11.6895	6.86	0.14	175.1060	1.5101	7.708	0.11
0.25	3.8425	3.9061	11.6876	6.88	0.12	175.4214	1.5083	7.683	0.06
0.35	3.8461	3.9083	11.6928	6.84	0.16	175.7628	1.5079	7.658	-
0.45	3.8492	3.9116	11.6960	6.82	0.18	176.1012	1.5071	7.633	-