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### Microstructure and Superconducting Properties of LaBaCaCu<sub>3</sub>O<sub>7-δ</sub>-Ba<sub>2</sub>HoNbO<sub>6</sub> Ceramic Composite

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## Microstructure and Superconducting Properties of $\text{LaBaCaCu}_3\text{O}_{7-\delta}$ - $\text{Ba}_2\text{HoNbO}_6$ Ceramic Composite

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In this work, we have synthesized and studied structural and microstructural characteristics of  $\text{LaBaCaCu}_3\text{O}_{7-\delta}$ - $\text{Ba}_2\text{HoNbO}_6$  composites.  $\text{Ba}_2\text{HoNbO}_6$  has an  $\text{A}_2\text{BB}'\text{O}_6$  complex cubic perovskite structure with lattice constant  $a = 8.439\text{\AA}$ .  $\text{Ba}_2\text{HoNbO}_6$  is chemically and physically compatible with  $\text{LaBaCaCu}_3\text{O}_{7-\delta}$  superconductor. So, we infer that  $\text{Ba}_2\text{HoNbO}_6$  could be a potential substrate material for the fabrication of the  $\text{LaBaCaCu}_3\text{O}_{7-\delta}$  superconducting films.

**Key words**  $\text{LaBaCaCu}_3\text{O}_{7-\delta}$ ,  $\text{Ba}_2\text{HoNbO}_6$ , superconducting composites,

### I. INTRODUCTION

Investigation on new substrate materials for high temperature superconducting films is a significant concern in materials research. Recently, complex perovskite oxides are being investigated for such applications [1-4]. In

the present work, we have synthesized and studied structural and microstructural characteristics of a complex cubic perovskite oxide  $\text{Ba}_2\text{HoNbO}_6$ , using x-diffractometry and scanning electron microscopy, respectively, for its use as a substrate material for the fabrication of  $\text{LaBaCaCu}_3\text{O}_{7.8}$  superconducting films.  $\text{LaBaCaCu}_3\text{O}_{7.8}$  is one of the important family of high temperature superconductors exhibiting superconductivity around 78K [5]. Chemical stability of  $\text{Ba}_2\text{HoNbO}_6$  with  $\text{LaBaCaCu}_3\text{O}_{7.8}$  was examined by x-ray diffractometry of  $\text{LaBaCaCu}_3\text{O}_{7.8}$ - $\text{Ba}_2\text{HoNbO}_6$  composites. Back-scattered scanning electron microscopy was used to study the interface interaction between  $\text{Ba}_2\text{HoNbO}_6$  and  $\text{LaBaCaCu}_3\text{O}_{7.8}$  grains in the  $\text{LaBaCaCu}_3\text{O}_{7.8}$ - $\text{Ba}_2\text{HoNbO}_6$  composites. Congruent melting aspect of the  $\text{Ba}_2\text{HoNbO}_6$  was studied to know whether this material could be grown as single crystal by melt growth processes. The effect of  $\text{Ba}_2\text{HoNbO}_6$  addition on the superconductivity of  $\text{LaBaCaCu}_3\text{O}_{7.8}$  was investigated by measuring magnetic susceptibility of  $\text{LaBaCaCu}_3\text{O}_{7.8}$ - $\text{Ba}_2\text{HoNbO}_6$  composites. These studies show that  $\text{Ba}_2\text{HoNbO}_6$  has favorable substrate characteristics and it could be a potential substrate material for the fabrication of high  $T_c$   $\text{LaBaCaCu}_3\text{O}_{7.8}$  superconducting films.

## II. EXPERIMENTAL DETAILS

$\text{Ba}_2\text{HoNbO}_6$  has been prepared by solid state reaction process. Stoichiometric mixture of high purity (99.99%) constituent chemicals  $\text{Ho}_2\text{O}_3$ ,  $\text{BaCO}_3$ ,  $\text{Nb}_2\text{O}_5$  were mixed thoroughly, pelletized and calcined at a temperature of  $1100^\circ\text{C}$  for 40h. The calcined material was reground, pressed as circular discs and sintered at  $1200^\circ\text{C}$  for 60h. Single phase  $\text{LaBaCaCu}_3\text{O}_{7.8}$  with nominal composition  $\text{La}_1\text{Ba}_1\text{Ca}_1\text{Cu}_3\text{O}_{7.8}$  was also prepared by solid state reaction process. Details of synthesis and characterization of  $\text{La}_1\text{Ba}_1\text{Ca}_1\text{Cu}_3\text{O}_{7.8}$  material are reported in our earlier publication [6].

X-ray diffraction (XRD) spectra of the materials were recorded by a Siemens D5000 x-ray diffractometer, using  $\text{Cu-K}\alpha$  radiation ( $\lambda = 1.5406 \text{ \AA}$ ). SEM micrographs were recorded by a Leico-Cambridge model stereoscan 440 electron microscope. For the study of chemical compatibility,  $\text{Ba}_2\text{HoNbO}_6$  -  $\text{LaBaCaCu}_3\text{O}_{7.8}$  composites, with 0 to 30wt% of  $\text{Ba}_2\text{HoNbO}_6$  component, were synthesized. For the synthesis of composites, component materials were mixed in desired wt% ratios and the mixture was pelletized as circular discs at a pressure of  $2\text{ton/cm}^2$ . These discs were heat treated at  $950^\circ\text{C}$  for 24h in flowing

oxygen and cooled down slowly at a rate of  $2^\circ\text{C}/\text{min}$  to room temperature for proper oxygenation.. Chemical stability of  $\text{Ba}_2\text{HoNbO}_6$  with  $\text{LaBaCaCu}_3\text{O}_{7-\delta}$  was examined by x-ray diffraction. Back-scattered electron microscopy was used to examine the interface interaction between  $\text{Ba}_2\text{HoNbO}_6$  and  $\text{LaBaCaCu}_3\text{O}_{7-\delta}$  grains. Effect of  $\text{Ba}_2\text{HoNbO}_6$  addition on superconductivity of  $\text{LaBaCaCu}_3\text{O}_{7-\delta}$  superconductors was investigated by a. c. magnetization measurements of  $\text{LaBaCaCu}_3\text{O}_{7-\delta}$ - $\text{Ba}_2\text{HoNbO}_6$  composites in the temperature range 5 to 300K, using a Quantum Design SQUID magnetometer.

### III. RESULTS AND DISCUSSION

X-ray diffraction (XRD) spectrum of single phase  $\text{Ba}_2\text{HoNbO}_6$  is shown in Figure 1. The XRD spectrum of  $\text{Ba}_2\text{HoNbO}_6$  is similar to that expected for  $\text{A}_2\text{BB}'\text{O}_6$ -type ordered complex cubic perovskites, reported in JCPDS files. As for many perovskite of the general formula  $\text{A}_2\text{BB}'\text{O}_6$ , an ordered arrangement of B and B' cations is most probable when large differences exist in either their charges or their ionic radii [3]. This is due to the fact that in a substitutional solid solution  $\text{BB}'$ , there is random arrangement of B and B' cations on equivalent position in the crystal structure. If upon suitable heat treatment the random solid solution rearranges into a structure in which B and B' occupy the same set of positions but in a regular way, such structure is described as superstructure [6]. In the superstructure the positions occupied by B and B' is no longer equivalent and this is exhibited in the XRD spectrum by the presence of superstructure reflection lines.

As seen from Figure1, the XRD pattern of  $\text{Ba}_2\text{HoNbO}_6$  consists of strong peak, characteristics of primitive cubic perovskite plus few weak lines arising from the superlattice. The significant presence of superstructure reflection lines (111) and (311) clearly reveal the  $\text{Ho}^{3+}$  and  $\text{Nb}^{3+}$  cations ordering on B and B' positions in  $\text{A}_2\text{BB}'\text{O}_6$  structure of  $\text{Ba}_2\text{HoNbO}_6$ . In  $\text{Ba}_2\text{HoNbO}_6$ ,  $\text{Ba}^{2+}$  cation (ionic radius 1.34 Å) with the largest ionic radius in this composition, occupies A position and  $\text{Ho}^{3+}$  (ionic radius 0.89 Å) and  $\text{Nb}^{3+}$  (ionic radius 0.69 Å) cations occupy B position due to their smaller ionic radii compared to that of  $\text{Ba}^{2+}$  cation. Due to the ordering of B and B' cations on octahedral site of the primitive  $\text{ABO}_3$  unit cell, there is doubling in the lattice parameter of the basic cubic perovskite unit cell.

Based on above considerations, we have indexed the XRD peaks of  $\text{Ba}_2\text{HoNbO}_6$  as an ordered complex cubic  $\text{A}_2\text{B}'\text{B}'\text{O}_6$  crystal structure. The lattice parameter of  $\text{Ba}_2\text{HoNbO}_6$ , calculated from XRD data, is  $a = 8.439 \text{ \AA}$ . Lattice matching of the superconductor with the substrate is an important aspect for the fabrication of good quality superconducting films.  $\text{Ba}_2\text{HoNbO}_6$  has a double cubic perovskite structure. As discussed earlier,  $(\frac{1}{2})a$  of  $\text{Ba}_2\text{HoNbO}_6 = 4.219 \text{ \AA}$ .  $\text{LaBaCaCu}_3\text{O}_{7.8}$  has a tetragonal crystal structure with lattice parameters  $a = 3.869 \text{ \AA}$  and  $c = 11.617 \text{ \AA}$ .

Therefore,  $\text{Ba}_2\text{HoNbO}_6$  has  $\sim 9\%$  lattice mismatch with  $\text{LaBaCaCu}_3\text{O}_{7.8}$  superconductor.

It may be noted that currently MgO is most widely used substrate for microwave applications. It has a cubic crystal structure (lattice constant  $a = 4.208 \text{ \AA}$ ) and has a comparable lattice mismatch with  $\text{LaBaCaCu}_3\text{O}_{7.8}$  superconductor..

The SEM micrographs of  $\text{Ba}_2\text{HoNbO}_6$  and  $\text{LaBaCaCu}_3\text{O}_{7.8}$  show that materials present homogenous surface morphology. Average particle sizes of the  $\text{Ba}_2\text{HoNbO}_6$  and  $\text{LaBaCaCu}_3\text{O}_{7.8}$  materials are estimated to be 3 - 5 microns and 3 - 10 micron, respectively. In the fabrication and processing of high  $T_c$  superconducting films, grain interface interaction between the grains of the substrate and superconductor material is an undesirable factor. Even MgO, most widely used substrate for high  $T_c$  superconducting films, does form an interlayer of barium salt at the superconductor-substrate interface, if the temperature of processing is higher than  $700^\circ\text{C}$  [7].

In present work, back scattered electron micrographs of the  $\text{Ba}_2\text{HoNbO}_6$ ,  $\text{LaBaCaCu}_3\text{O}_{7.8}$  single-phase materials and the  $\text{LaBaCaCu}_3\text{O}_{7.8}$ - $\text{Ba}_2\text{HoNbO}_6$  composite materials were recorded using quarterback scattering detector. The back-scattered electron SEM micrographs of  $\text{LaBaCaCu}_3\text{O}_{7.8}$ - $\text{Ba}_2\text{HoNbO}_6$  composite, presented in Figure 2, shows that there is no detectable interface interaction between  $\text{Ba}_2\text{HoNbO}_6$ , and  $\text{LaBaCaCu}_3\text{O}_{7.8}$  grain and  $\text{Ba}_2\text{HoNbO}_6$  grains are distinguishably distributed in the  $\text{LaBaCaCu}_3\text{O}_{7.8}$  matrix

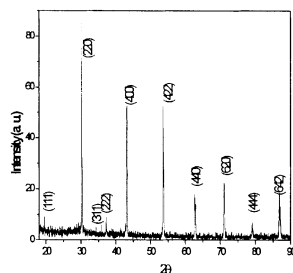


FIGURE 1 XRD spectrum of  $\text{Ba}_2\text{HoNbO}_6$ .

Chemical stability of  $\text{Ba}_2\text{HoNbO}_6$  with  $\text{LaBaCaCu}_3\text{O}_{7-\delta}$  superconductor was investigated by x-ray diffractometry on  $\text{LaBaCaCu}_3\text{O}_{7-\delta}$ - $\text{Ba}_2\text{HoNbO}_6$  composites. Figure 3 shows the XRD spectra of these composites. As seen from the XRD results, all the XRD peaks correspond either to  $\text{Ba}_2\text{HoNbO}_6$  or  $\text{LaBaCaCu}_3\text{O}_{7-\delta}$  and there is no extra peak corresponding to any impurity phase. These results show that there is no chemical interaction between these materials and  $\text{Ba}_2\text{HoNbO}_6$  is chemically compatible with  $\text{LaBaCaCu}_3\text{O}_{7-\delta}$  superconductors.

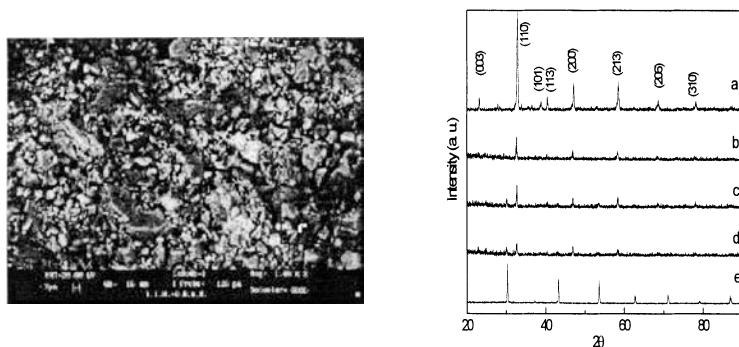


FIGURE 2 Back-scattered SEM micrographs of  $\text{LaBaCaCu}_3\text{O}_{7-\delta}$ - $\text{Ba}_2\text{HoNbO}_6$  composite containing 30wt%  $\text{Ba}_2\text{HoNbO}_6$  component

FIGURE 3 XRD spectra of the  $\text{LaBaCaCu}_3\text{O}_{7-\delta}$ - $\text{Ba}_2\text{HoNbO}_6$  composites containing (a) 0wt%, (b) 5wt%, (c) 20wt%, (d) 30wt%, and (e) 100wt%  $\text{Ba}_2\text{HoNbO}_6$

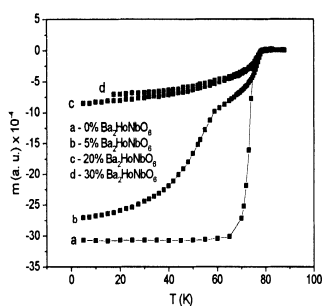


FIGURE 4 a.c. magnetization versus temperature curves of  $\text{LaBaCaCu}_3\text{O}_{7-\delta}$ - $\text{Ba}_2\text{HoNbO}_6$  composites

Figure 4 shows the temperature dependence of the real part of the ac magnetization for  $\text{LaBaCaCu}_3\text{O}_{7.8}\text{-Ba}_2\text{HoNbO}_6$  composites. All the composites gave a  $T_c$  of 78K, same as of pure  $\text{LaBaCaCu}_3\text{O}_{7.8}$  superconductor. However, with decreasing superconductor volume fraction the magnitude of magnetization decreases in all the composite samples. Accordingly, we infer that addition of the  $\text{Ba}_2\text{HoNbO}_6$ , an insulating ceramic material, has no deteriorating effect on the superconducting properties of the  $\text{LaBaCaCu}_3\text{O}_{7.8}$  superconductors.

#### IV. CONCLUSIONS

In conclusion, we have studied structural and microstructural characteristics of  $\text{LaBaCaCu}_3\text{O}_{7.8}\text{-Ba}_2\text{HoNbO}_6$  composite.  $\text{Ba}_2\text{HoNbO}_6$  has fairly good lattice matching (lattice mismatch  $\sim 9\%$ ) with this superconductor. X-ray diffractometry, scanning electron microscopy and magnetic measurements made on  $\text{LaBaCaCu}_3\text{O}_{7.8}\text{-Ba}_2\text{HoNbO}_6$  composites show that  $\text{Ba}_2\text{HoNbO}_6$  is chemically compatible with  $\text{LaBaCaCu}_3\text{O}_{7.8}$ . These favorable characteristics show that  $\text{Ba}_2\text{HoNbO}_6$  could be used as a potential substrate material for the fabrication of the  $\text{LaBaCaCu}_3\text{O}_{7.8}$  superconducting films.

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