UNIVERSITI PUTRA MALAYSIA

SYNTHESIS AND CHARACTERIZATION OF SAMARIUM DOPED Bi1.6Pbo.4Sr2Ca2Cu3O10 SUPERCONDUCTOR PREPARED VIA COPRECIPITATION METHOD

IMAD MOH'D KHAIR RASHID HAMADNEH

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By

IMAD MOH'D KHAIR RASHID HAMADNEH

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfillment of the Requirement for the Degree of Doctor of Philosophy

September 2002

DEDICATIONS

To my wife, Lama and my son, Yazan,
for their love, support and understanding....

To my late father, my mother and the family,
for their love and concern....

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the Requirements for the degree of Doctor of Philosophy

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IMAD "MOH'D KHAIR" RASHID HAMADNEH

September 2002

Chairman: Professor Abdul Halim bin Shaari, Ph.D

Faculty : Faculty of Science and Environmental Studies

The coprecipitation technique was used in the preparation of samarium doped $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_{10}$ polycrystalline ceramic superconductor. In this study, four sites (calcium, bismuth, strontium and copper) were separately subjected to Sm doping with nominal composition ranging from x=0 to x=0.3 at different sintering times. The superconducting properties of the samples have been investigated. The pure sample, which exhibits $T_{C(R=0)}$ around 102 – 103 K and $T_{C\text{-onset}}$ around 108 K, showed large flaky grains of ~7 μm in size which are randomly distributed. However at longer sintering time the pure superconductor showed a better orientation as compared to that of the short sintering time sample. The calculated value of Josephson current I_o , obtained from the ac susceptibility data showed a much higher value (I_o =138.7μA) as compared to the conventional prepared to that of the sample prepared by conventional method

(I_o =55.9 μA). This indicates better grain connectivity and higher 2223 phase content, which was confirmed by SEM photographs. In addition, the nature of the ultra fine particles of the oxalate powders produced by coprecipitation method have increased the diffusion reaction and shortened the heat treatment procedure for the sample preparation, this leads to better superconducting properties as compared to the samples prepared by conventional solid state technique where its diffusion reaction requires high sintering temperatures for long duration and sometimes several grindings.

The resistivity measurements showed the normal metallic behaviour followed by shifts in $T_{C(R=0)}$ towards lower temperature as the samarium concentration increased due to the decrease in the 2223 phase and an increase in the formation of 2212 phase. However, the material lost its superconductivity at Sm concentration x >0.20. When samarium was doped in Ca^{2+}, Sr^{2+}, and Cu^{2+} sites, it probably brings about changes in the hole carrier concentration which in turn alters $T_{C(R=0)}$. Hence it could be deduced that the valency of the dopant has some influence on the electron pairing mechanism.

The temperature dependence of ac susceptibility data χ' shows the shifting of the onset diamagnetism towards lower temperature as the Sm concentration increased due to the presence of low T_c phase. The imaginary component, χ", shows a shift in the intergranular coupling peak, Tp, towards lower temperature as the Sm concentration increased. Hence it can be deduced that the dynamic magnetic response of the samples are not only phase dependent but also dependant on the intergranular coupling. The calculated I_0 which revealed the quality of the coupling of the grains, showed a decrease

in its value as the samarium concentration increased. For highest doping percentage of samarium the values of I_0 decreased in the following order; $I_{0\,(in\,Ca)} < I_{0\,(in\,Cu)} < I_{0\,(in\,Sr)} < I_{0\,(in\,Bi)}$.

The results of x-ray diffraction (XRD) pattern show that all samples with Sm concentration above x=0.02 contain unknown peaks which correspond to the non-superconducting phase. The intensity of these peaks increases toward higher value, as the Sm concentration increases. The volume of 2223 phase decreased drastically as the Sm concentration increases, whereas the amount of decrease varied due to the doping at different sites. In addition, there is a possibility that either Sm^{2+} or Sm^{3+} might have occupied other sites at the same sample.

When long sintering time was applied, the improvement in superconducting properties was obvious at low doping concentrations x< 0.06 where the sample was still dominated by 2223 phase. Above that concentration, the grain size decreased and became short and thick, randomly distributed as compared to the pure phase. It is also observed that the superconducting properties and the microstructure improved when the sample was sintered for 48 hours and 100 hours, the high T_C phase dominates, indicating that the optimum time must be above 48 hours.

Abstrak disertasi yang dikemukakan kepada senat Universiti Putra Malaysia bagi memenuhi keperluan untuk ijazah Doktor Falsafah

SINTESIS DAN PENCIRIAN SUPERKONDUKTOR $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_{10}$ YANG DIDOPKAN DENGAN SAMARIUM MELALUI KAEDAH PEMENDAKAN BERSAMA

Oleh

IMAD "MOH'D KHAIR" RASHID HAMADNEH

Ogos 2002

Pengerusi : Profesor Dr. Abdul Halim Shaari, Ph.D

Fakulti : Sains dan Pengajian Alam Sekitar

Teknik pemendakan bersama telah digunakan bagi menyediakan superkonduktor seramik polihablur $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_{10}$ yang didopkan dengan samarium. Dalam kajian yang dijalankan ini, empat tapak (kalsium, bismut, strontium dan kuprum) telah didopkan dengan Sm secara berasingan dengan komposisi nominal di antara julat x=0 hingga x=0.3 pada masa pensinteran yang berbeza. Sifat kesuperkonduksian bagi sampel-sampel ini telah dikaji. Di dapati sampel tulen menunjukan $Tc_{(R=0)}$ di antara 102 –103 K dan Tc_{onset} adalah sekitar 108 K. Sampel ini menunjukkan butiran besar dan berkeping yang bertaburan secara rawak dengan saiz butiran >7 μm. Pada masa pensinteran yang tinggi, superkonduktor tulen akan menunjukkan orientasi yang lebih baik berbanding orientasi pada sampel denganmasa persinteran yang lebih pendek.

Nilai perkiraan arus Josephson, I_o , yang diperolah daripada data kerentanan arus ulang alik, menunjukkannilai yang lebih tinggi (I_o =138.7μA) berbandingkan sampel yang di sediakan secara konvensional (I_o =55.9 μA). Ini disebabkan oleh penyambungan butir yang lebih baik dan kandungan fasa 2223 yang tinggi dan disahkan oleh gambar foto SEM. Tambahan pula, sifat semula jadi zarah halus serbuk oksalat yang dihasilkan melalui kaedah pemendakan bersama telah meningkatkan kadar tindakbalas resapan dan memendekkan tempoh proses rawatan haba dalam penyediaan sampel. Ini menghasilkan sifat kesuperkonduksian yang lebih baik berbanding dengan sampel yang disediakan menggunakan kaedah lazim iaitu teknik tindakbalas keadaan pepejal, di mana tindakbalas resapannya memerlukan suhu persinteran yang tinggi dan lama serta turut memerlukan beberapa proses pengisaran.

Pengukuran kerintangan menunjukkan sifat logam normal dan diikuti dengan anjakan $Tc_{(R=0)}$ kepada suhu rendah apabila kepekatan samarium meningkat. Ini disebabkan oleh pengurangan pada fasa 2223 dan peningkatan dalam pembentukan fasa 2212. Walau bagaimanapun, bahan ini akan kehilangan sifat kesuperkonduktsian pada kepekatan x>0.2. Perubahan pada kepekatan pembawa lubang mungkin akan berlaku jika semarium didopkan pada tapak Ca^{2+}, Sr^{2+} dan Cu^{2+} seterusnya akan mengubah $Tc_{(R=0)}$. Oleh itu bolehlah disimpulkan bahawa valensi bagi pendopan akan mempengaruhi mekanisme pasangan elektron.

Data-data kerentanan arus ulang-alik, χ', yang bersandarkan suhu menunjukkan berlakunya suatu anjakan pada onset diamagnet akibat suhu yang lebih rendah dan pertambahan kepekatan Sm akibat kewujudan fasa T_C yang rendah. Komponen khayal, χ'', menunjukkan anjakan pada puncak gandingan antara butiran T_P ke suhu yang lebih rendah apabila kepekatan Sm bertambah. Dengan yang demikian, bolehlah disimpulkan bahawa tindakbalas dinamik magnet sampel-sampel bukan hanya bersandarkan fasa tetapi juga bersandar pada gandingan antara butiran. Nilai kiraan I_0, menunjukkan kualiti gandingan butiran telah menunjukkan pengurangan nilainya apabila kepekatan semarium bertambah. Untuk kepekatan samarium yang paling tinggi, nilai bagi I_0 berkurangan dalam tertib berikut $I_{0(\text{ dalam Ca})} < I_{0(\text{ dalam Cu})} < I_{0(\text{ dalam Sr})} < I_{0(\text{ dalam Bi})}$.

Keputusan pembelauan corak sinar-X atau XRD bagi semua sampel dengan kepekatan dopan Sm melebihi x=0.02, mengandungi puncak-puncak yang tidak diketahui yang berkaitan dengan fasa bukan superkonduktor. Keamatan puncak-puncak berikut didapati meningkat kepada nilai yang lebih tinggi sejajar dengan peningkatan komposisi dopan Sm. Isipadu fasa 2223 berkurang secara mendadak apabila komposisi dopan ditingkatkan, manakala kadar pengurangan berbeza-beza disebabkan oleh pendopanan pada tapak-tapak(site) yang berlainan. Tambahan lagi, terdapat kemungkinan Sm^{2+} atau Sm^{3+} menduduki oleh tapak-tapak yang lain di dalam sampel yang sama.

Apabila masa pensinteran yang panjang dilakukan, didapati terdapat peningkatan bagi sifat-sifat kesuperkonduksian pada kepekatan dopan yang rendah iaitu pada x=0.06, di mana sampel masih di dominasi oleh fasa 2223. Pada kepekatan yang lebih tinggi,

saiz butiran akan berkurangan dan bentuknya menjadi pendek dan tebal serta bertaburan secara rawak berbanding fasa tulen. Turut dapat diperhatikan, sifat-sifat kesuperkonduksian dan mikrostrukturnya akan meningkat apabila sampel disinter selama 48 jam dan 100 jam, di mana fasa T_C yang tinggi akan berlaku dan ini menunjukkan bahawa masa optimum bagi pensinteran perlu dilakukan melebihi 48 jam.

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I certify that an Examination Committee met on 25th September 2002 to conduct the final examination of Imad Moh'D Khair Rashid Hamadneh on his Doctor of Philosophy thesis entitled "Synthesis and Characterization of Samarium Doped $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_{10}$ Superconductor Prepared via Coprecipitation Method" in accordance Universiti Pertanian Malaysia (Higher Degree) act 1980 and Universiti Pertanian Malaysia (Higher Degree)Regulations 1981. the committee recommends that candidate be awarded the relevant degree. Members of the Examination Committee are as follows:

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I hereby declare that the thesis is based on my original work except for quotations and citations, which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at UPM or other institutions.

_____

IMAD "MOH'D KHAIR" RASHID HAMADNEH

Date:

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