EFFECTS OF Mn SUBSTITUTION IN YBa$_2$Cu$_3$O$_{7-\delta}$ SUPERCONDUCTING CERAMIC

By

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Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Master of Science

March 2004
This thesis is dedicated to:

My late brother: Khaled
Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirements for the degree of Master of Science

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Chairman: Professor Abdul Halim Shaari, Ph. D.

Faculty: Science and Environmental Studies

The influence of manganese (Mn) substitution in copper (Cu), barium (Ba) and yttrium (Y) sites in the Y-Ba-Cu-O system namely: YBa$_{2-x}$Mn$_x$Cu$_3$O$_{7-\delta}$ (0.00 $\leq$ x $\leq$ 0.5), YBa$_{2-x}$Mn$_x$Cu$_3$O$_{7-\delta}$ (0.00 $\geq$ x $\geq$ 0.5) and Y$_{1-x}$Mn$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ (0.00 $\geq$ x $\geq$ 0.5) were studied. The samples were prepared using the conventional solid state sintering technique. The sintered temperature and soaking time were 920ºC and 24 hours respectively. The transport properties of the samples were measured using the four-point probe electrical resistance measurement and the magnetic properties were measured by using ac susceptibility. Scanning electron microscope (SEM) was used to identify the surface morphology while the phase of the samples was determined using x-ray diffraction (XRD) technique.

The pure sample, which exhibits $T_{c,(R=0)}$ around 90 K and $T_{conset}$ around 94 K showed large flaky grains of $\sim$ 25 $\mu$m in size with some gaps and voids. The effect of Mn substitution on the superconducting behaviour of YBCO shows that the transition...
temperature, $T_C$ was sharply reduced when the substitution site is at Ba. For the sample in which $x=0.30$ the values of $T_C$ are 80K, 72K and 60K for substitution in the Y, Cu and Ba sites, respectively.

The dynamic magnetic properties of the samples show that the effect of Mn substitution is markedly decreased in the Ba substituted samples. The value of Josephson current $I_o$ for $x=0.3$ are 13.6µA, 14.1µA and 6.9µA for the substitution in Y, Ba and Cu sites, respectively. The XRD patterns showed that the orthorhombic structure was retained for all the samples. However, at higher concentration of Mn substitution, some unknown peaks were observed. There is a general trend of an increase of (102) peaks as Mn concentration increases at all sites, while the (003) and (006) peaks decreased.

The morphology of the samples varies as Mn was substituted at different sites. At lower concentration, the shapes of the grains are sharper in Mn substituted the Y samples. When $x=0.3$, the grains are loosely packed and more rounded when Mn substituted the Ba sites. When manganese was incorporated in $Y^{3+}$, $Ba^{2+}$ and $Cu^{2+}$ sites, the resistivity result showed the shifting in $T_{C(R=0)}$ towards low temperature as the Mn increases.

The temperature dependence of ac susceptibility data $\chi'$ shows the shifting of the onset diamagnetism towards lower temperature as the Mn concentration increased
due to the presence of low $T_C$ phase. The imaginary component, $\chi''$ shows a decrease in the intergranular coupling peak, $T_P$ towards lower temperature as the Mn concentration increased. XRD patterns showed the existence of the unknown peaks, which belong to the impurities. SEM micrographs showed the decrease in the grain size as the concentration increases.
Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

KESAN PENGGANTIAN Mn DALAM BAHAN SERAMIK SUPERKONDUKTOR YBa$_2$Cu$_3$O$_{7-\delta}$

Oleh

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Pengaruh penggantian mangan (Mn) ke dalam kuprum (Cu), barium (Ba) dan yitrium dalam sistem Y-Ba-Cu-O, iaitu YBa$_2$Cu$_{3-x}$Mn$_x$O$_{7-\delta}$ (0.00 $\geq$ x $\geq$ 0.5), YBa$_{2-x}$Mn$_x$Cu$_3$O$_{7-\delta}$ (0.00 $\geq$ x $\geq$ 0.5) dan Y$_{1-x}$Mn$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ (0.00 $\geq$ x $\geq$ 0.5) telah dikaji. Sampel-sampel disediakan dengan menggunakan teknik lazim keadaan pepejal. Suhu sinter dan masa meresap adalah masing-masing 920ºC dan 24 jam. Ciri-ciri pengangkutan bagi sampel telah diukur dengan menggunakan penguku ran rintangan elektrik 4 titik dan ciri-ciri magnetik telah diukur dengan menggunakan kaedah kerentanan au. Mikroskop pengimbas elektron (SEM) telah digunakan untuk mengenalpasti permukaan morfologi sementara fasa bagi sampel tela h ditentukan menggunakan teknik belauan sinar-x (XRD).

Sampel tulen mempamerkan $T_c(R=0)$ dalam 90K dan $T_c$ mula dalam 94K menunjukkan saiz butiran besar lebih kurang 25µm dengan sedikit liang.

Penggantian Mn terhadap YBCO menunjukkan suhu peralihan $T_c$ mengurang dengan mendadak apabila penggantian di tapak Ba dilakukan. Untuk sampel di mana x=0.3,
nilai $T_c$ adalah 80K, 72K dan 60K untuk penggantian dalam bahagian Y, Cu dan Ba masing-masing.

Ciri-ciri magnetik yang dinamik bagi semua sampel menunjukkan bahawa penggantian mangan (Mn) menyebabkan $T_c$ menurun secara mendadak dengan penggantian Ba. Nilai arus Josephson, $I_o$ pada $x=0.3$ adalah 13.6µA, 14.0µA dan 6.9µA untuk penggantian dalam Y, Ba dan Cu masing-masing.

Bentuk corak pembelauan sinar-X menunjukkan struktur ortorombik telah diperolehi untuk semua sampel. Walau bagaimanapun, bagi pehingkatan Mn yang berkepekatan tinggi, beberapa puncak tidak dikenali dapat dilihat. Terdapat arah peningkatan umum puncak-puncak (102) seperti peningkatan kepekatan Mn pada setiap sudut, sementara puncak (003) dan (006) berkurangan.

Morfologi sampel berbeza sebagaimana penggantian Mn pada kekisi yang berlainan. Bentuk butiran lebih tajam di dalam sampel dimana Mn menggantikan Y pada kepekatan yang rendah. Apabila $x=0.3$, butiran adalah longgar dan lebih bulat bagi penggantian Mn kepada kekisi Ba.

Apabila mangan di masukkan ke dalam tapak $Y^{3+}$, $Ba^{2+}$ and $Cu^{2+}$, keputusan kerintangan menunjukkan peralihan $T_C$ kepada suhu rendah dengan pertambahan Mn. Pergantungan data kerentanan ac $\chi'$ menunjukkan peralihan diamagnet mula kepada suhu rendah dengan pertambahan kepekatan Mn disebabkan kehadiran fasa $T_C$ rendah. Komponen khayalan, $\chi''$ menunjukkan pengurangan dalam puncak gandingan antara butir, $T_P$ kepada suhu rendah dengan petambahan kepekatan Mn. Corak XRD
menunjukkan kewujudan puncak-puncak tidak dikenali, yang dipunyai oleh bendasing. Mikrograf SEM menunjukkan pengurangan saiz butir dengan pertambahan kepekatan.
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I certify that an Examination Committee met on 9th March 2004 to conduct the final examination of Mustafa Musa Ali Dihom on his Master of Science thesis entitled “Effects Of Mn Substitution In YBa$_2$Cu$_3$O$_{7-\delta}$ Superconducting Ceramic” in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulations 1981. The committee recommends that the candidate be awarded the relevant degree. Members of the Examination Committee are as follows:

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This thesis submitted to the Senate of Universiti Putra Malaysia has been accepted as fulfillment of the requirement for the degree of Master of Science. The members of the Supervisory Committee are as follows:

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DECLARATION

I hereby declare that the thesis is based on my original work except for quotation 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.

Mustafa Musa Ali Dihom
Date:
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