Reactor Irradiation Effects on Superconductivity in Y-Ba-Cu Oxides at Low and Pile Temperature

Kozo ATOBE, Hiroyuki YOSHIDA, Kiyomi MIYATA and Hisao KODAKA

(Research Reactor Institute, Kyoto University, Kumatori-cho, Sennan-gun, Osaka 590-04)

Y-Ba-Cu 酸化物超伝導特性に及ぼす低温と常温の原子炉照射効果

跡部 紘三・吉田 博行・宮田 清美・小高 久男

(京都大学原子炉実験所, 590-04 大阪府泉南郡熊取町)

Reactor irradiation effects on the superconducting transition temperature (T_c) have been studied by the in-pile measurements of electrical resistance and inductance of the $YBa_2Cu_3O_x$ before and after as well as during neutron irradiation. The reactor irradiations were performed for the periods of one and two weeks at 25 and/or 360 K using the low temperature irradiation loop facility at KUR. The resistance and the inductance have been measured continuously during the cooling down and the warming up of the loop before and after the irradiation. It was found that the irradiation at the ambient (360 K) temperature enhances broadening of the transition in the superconductivity in Y-Ba-Cu oxides, compared with the irradiation at the low temperature (25 K), up to the fluence of 10^{15} - 10^{16} fast neutron/cm². [Received November 19, 1987; Accepted January 22, 1988]

Key-words : In-pile measurement, Y-Ba-Cu oxides, Reactor irradiation effects, Irradiation damage, Oxygen vacancy, Aggregation

1. Introduction

Since Bednorz and Müller¹⁾ reported the oxide compounds with high superconducting transition temperature (T_c), extensive works have been done on high T_c oxides. Now, the oxygen poor layer structure with oxygen vacancy is considered to be related to the superconductivity of the oxide compounds.²⁾ The study of neutron irradiation effects on these oxides are essential in order to know the relation between the superconducting transition and the oxygen vacancies, and are also important to understand the sensitivity of the oxides against radiation, if we expect the oxides as high T_c superconductors for high field magnet of future fusion reactors. These superconducting magnet materials will be exposed to fast neutron irradiation at low temperature, i. e. in superconducting condition.

For the above reason, we tried in situ measurements of resistance and inductance for superconducting $YBa_2Cu_3O_x$ during reactor irradiation in order to obtain the informations of the damage process. Further, the superconducting characteristic of these oxides before and after the irradiation at 25 K and 360 K were compared by in-pile measurements using the low temperature irradiation loop (LTL) of Kyoto University Research Reactor (KUR). We have partly reported the effects of the low temperature irradiation on $YBa_2Cu_3O_x$ superconductor.³⁾

2. Experimentals

2.1 Specimens

All specimens used in this experiment were prepared by solid reaction. Raw materials of powder were weighed in cation ratio, Y : Ba : Cu = 1 : 2 : 3. Powder of Y_2O_3 , $BaCO_3$ and CuO were mixed automatically for 4 hours and it was calcined for 10 hours at 800°C in an open furnace for 8 hours. The typical shape of the specimen is plate and the specimens; Y-1, Y-2 and Y-3 were prepared from the sintered compacts to the size of $15 \times 4 \times 2.9$, $22 \times 4 \times 2.3$ and $15 \times 3 \times 1.5$ mm³, respectively for the measurements. The orthorhombic structure of the specimens was determined by X-ray powder diffraction method before the irradiations.

2.2 Measurements

All the experiments have been done by in-pile measurements. The specimens connected with four leading Cu wires to measure the resistance by standard four point method were set in the irradiation loop at KUR.⁴⁾ Silver paint was used to improve the contact between the specimen and Cu wires. Some of specimens were set simultaneously in the inductance coil for the measurement of Meissner effect in the magnetization. Their resistance and inductance were continuously measured during the reactor irradiation and also measured during cooling down and warming up. These in-pile measurements have been done

Table 1 The condition of reactor irradiations.

Irradiation patterns	P-1	P-2	P-3
Irradiation specimens	Y-1	Y-2	Y-3
Irradiation conditions \ Irradiation week	One week	First week	Second week
Fast neutron dose (n/cm ²)	5.41×10^{15}	3.74×10^{15}	3.69×10^{15}
Thermal neutron dose (n/cm ²)	8.32×10^{16}	6.09×10^{16}	6.01×10^{16}
Gamma-ray dose (R)	3.00×10^8	2.31×10^8	2.28×10^8
Irradiation time (hr)	77	77	76
Irradiation temperature (K)	25	25	360
Irradiation atmosphere	helium gas	helium gas	helium gas

Fast neutron: $E > 0.1$ MeV

in three irradiation patterns (P-1, P-2 and P-3), i. e. P-1 : 25 K irradiation for 77 hours, P-2 : 25 and 360 K irradiations for 77 and 76 hours, respectively. P-3 : 360 K irradiation for 76 hours as seen Table 1.

The in-pile measurements of resistance and inductance were continuously performed by a computer control system. Three specimens $\text{YBa}_2\text{Cu}_3\text{O}_x$, i. e. Y-1, Y-2 and Y-3 were used in the reactor irradiation patterns, i. e. P-1, P-2 and P-3, respectively. The resistivity at room temperature of the specimen is $1.1 \times 10^{-3} \Omega \cdot \text{cm}$.

3. Results and discussion

All the irradiation conditions are shown in Table 1. Figure 1 shows the changes of electrical resistance of Y-1 during the continuous measurement for the low temperature irradiation (P-1). The temperature of the specimen before the irradiation was 20 K and then it became to 25 K when KUR power reached at 5 MW. The superconducting transition of the specimen occurs at 91 K before the irradiation and the superconduct-

Fig. 1 Resistance in normal state and superconducting state vs. measuring time curve of Y-1 specimen obtained by in-pile measurement at 25 K, and after subsequent short-annealing.

ing state was kept during reactor irradiation (fast neutron flux = $1.1 \times 10^{11} \text{ n/cm}^2 \cdot \text{s}$). After the irradiation for 77 hours, Y-1 was warmed up to 300 K and then kept at the temperature for 0.5 hour to get the information for annealing effect of irradiation in defect induced specimen at 25 K. After annealing, it was cooled down again for the measurement. The resistance change during this thermal process is also shown in Fig. 1. The resistance showed same level before and after the annealing at 300 K. Figure 2 shows the partial figure of the resistance vs. temperature curves for the same specimen. The temperature dependence of the resistance and the superconducting transition temperature, i. e. T_c (onset) and T_c (final), in Y-1 was compared before and after the 25 K irradiation. There is no significant change in the transition temperature and the resistance level above T_c . As seen in Fig. 3, the change of inductance in Y-2 is clear near at 90 K from 0.97 to 0.92 mH, however, no discernible difference can be detected in the transition temperature before and after the 25 K irradiation (P-2) as in the resistance measurement (Fig. 2). In oxides, it is well known that vacancies at oxygen lattice

Fig. 2 Resistance vs. temperature curve for Y-1 specimen measured before and after the 25 K irradiation.

Fig. 3 Inductance vs. temperature curve for Y-2 specimen measured before and after the 25 K irradiation.

Fig. 4 Inductance vs. temperature curve for Y-2 specimen measured after irradiation at 25 K and after irradiation at 360 K.

sites are produced through collision sequence by irradiation with fast neutrons ($E > 0.1$ MeV).⁽⁵⁻⁷⁾ The induced point defects are normally detected as F^+ and F centers (oxygen vacancy traps one or two electrons, respectively) by optical absorption and ESR methods. In Y-Ba-Cu oxides, it can be also expected that this type of oxygen vacancies might be produced at lattice sites through displacement collisions by fast neutron irradiation. However, the production of oxygen vacancies through the irradiation process does not give any detectable effect on either the transition temperature or the resistance value as seen in Figs. 1 and 2. While, as seen in Fig. 4, the broadening of transition temperature width appeared after the 360 K irradiation in contrast with the case of the 25 K irradiation. These observations indicate that the irradiation temperature of the specimen is an important factor on the damage process in this oxide by the reactor radiation probably because the radiation-induced point defects can migrate and aggregate in this

Fig. 5 a) Resistance vs. temperature curve before irradiation for Y-3 specimen.
b) Resistance vs. temperature curve after irradiation at 360 K for Y-3 specimen.

structure at 360 K. Figure 5 shows the temperature dependence of the resistance in Y-3 before (Fig. 5 a)) and after (Fig. 5 b)) the 360 K irradiation (P-3) to a dose of 6.02×10^{16} fast neutron/cm². It clearly shows the presence of the irradiation effect on Y-3, i.e. the disappearance of the superconductive transition. Such a drastic change in the electrical property is observed only in the result of the 360 K irradiation. It can be understood that the electrical resistance measurement during the irradiation damage is more sensitive as compared with the inductance measurement. Thus, it may detect the destruction of superconductive path in the local region and it does not mean the transition of super- to normal-conductor in the bulk of this oxide by the 360 K irradiation. This interpretation is supported by the inductance measurement after the 360 K irradiation (Fig. 4).

From the above results, the 25 K irradiation gives no clear change to Y-Ba-Cu oxides superconductor in the transition temperature and the superconductivity in this oxides is retained under reactor neutron irradiation (fast neutron flux = 1×10^{11} n/cm² · s) at 25 K up to a 5×10^{15} neutrons/cm². However, the 360 K irradiation shows clear difference comparing with the 25 K irradiation,

i.e., the broadening of the transitions occurs after the irradiation.

It can be interpreted that radiation induced point defect, such as oxygen vacancy is "frozen" at the low temperature as a pinning point defect and gives little effect on the superconductivity, but the production of oxygen vacancies and their migration and the aggregation of each vacancies during the 360 K irradiation affect strongly on the superconducting local phase in this oxide.

Acknowledgement The authors would like to express their thanks to Mr. M. Okada and members of the LTL Joint Study Group, and to KUR members for their support to the present experiment.

References

- 1) J.G. Bednorz and K.A. Müller, *Z. Phys.*, **B 63**, 189 (1986).
- 2) S.B. Qadri, L.E. Toth, M. Okfsky, S. Lawrence, D.U. Gubser and S.A. Wolf, *Phys. Rev.*, **B 35**, 7235 (1987).
- 3) K. Atobe and H. Yoshida, *Phys. Rev. B*, **36**, 7194 (1987).
- 4) K. Atobe et al., *Annual. Repts. Res. Reactor Inst., Kyoto Univ.*, **9**, 139 (1976).
- 5) Y. Chan, J.L. Lolopus and W.A. Sibley, *Phys. Rev.*, **186**, 865 (1969).
- 6) K.H. Lee and J.H. Crawford, Jr., *Phys. Rev.*, **B 15**, 4665 (1977).
- 7) G.S. White, K.H. Lee and J.H. Crawford, *Phys. Rev.*, **B 21**, 2578 (1980).