IOP Conf. Series: Materials Science and Engineering **1345** (2026) 012018 doi:10.1088/1757-899X/1345/1/012018

Nuclear transmutation effect by thermal neutron on degradation in superconductivity of ReBCO tapes

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Abstract. Rare earth barium copper oxide (ReBCO) tapes of GdBCO, EuBCO and YBCO have been neutron-irradiated in Japan Research Reactor #3 and isotopes produced during the irradiation were investigated by a Ge detector along with superconductivity tests at high magnetic field. The maximum thermal and fast neutron was 8.29 x 10^{22} n/m^2 and 1.46 x 10^{21} n/m^2, respectively. As the results, ^{153}Gd, ^{152}Eu and ^{155}Eu were detected and nuclear transmutation was confirmed. No isotopes of Y were found. Since the cross section of Gd for {n,γ} reaction is very large in thermal neutron region, the nuclear transmutation of Gd occurs by the capture of thermal neutrons. Eu transmutation occurs by the same mechanism. These facts suggest that the thermal neutrons have dominant effect on the degradation in superconductivity of GdBCO and EuBCO tapes. The possible mechanisms on the degradation, a Gd/Eu recoil model, an oxygen deficiency model and an electron exchange model, are discussed.

1. Introduction

Fusion energy is one of new energy sources and many countries are trying to realize the fusion energy. Since Deuterium-Tritium reaction occurs at relatively lower temperature, a D-T plasma device is a target of most projects in the world. The D-T reaction yields 14 MeV neutrons and one of the key challenges is to keep the D-T plasma going for as long as possible. To produce the high magnetic field and to keep the high-performance plasma, the superconducting (SC) magnets are applied and operated. It must be noted that high temperature SC (HTS) tapes have brought the possibility of high temperature operation, such as 20 K [1], and re-mountable joint [2]. The high temperature operation enables significant savings in on-site electricity, and the re-mountable joint makes the short time replacement of inner components in a plasma vacuum vessel (VV) possible. These benefits will improve the fusion reactor operation technology.

The high energy neutron can penetrate a blanket and a VV and reach the SC magnet. Some neutrons stream out of the VV and hit the SC magnet. Although the neutron fluence will be not so high, but the neutron irradiation effect on the SC wires and tapes must be understood and the fundamental knowledge must be reflected on the design of the D-T plasma devices.

With the Japanese Research Reactor #3 (JRR-3) of a fission reactor restarted in 2022 after shutdown due to the Great East Japan Earthquake in 2011, neutron irradiation tests of rare earth barium copper oxide (ReBCO) tape have resumed immediately in FY2022. After the irradiation, the irradiated ReBCO layer was investigated by a Ge detector and SC properties were investigated. Based on the experimental results and data of cross section to thermal neutrons, a mechanism of

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degradation in superconductivity of ReBCO tapes. Three models, a recoil model, oxygen deficiency model and electron exchange model, are presented and the possible mechanisms are discussed.

2. Test material and test procedures

2.1 Test materials

The ReBCO tapes tested in this study were GdBCO, EuBCO with artificial pinning center and YBCO tapes provided by Shanghai Superconductor Technology Co., Ltd. The width was 4 mm and thickness was about 0.1 mm. According to the web site of the company [3], the layer structure is as follows: The right surface is Cu layer, with an Ag layer underneath that, and a ReBCO layer underneath Ag layer. The ReBCO layer is 1-2 μm thick. Below that is an insulating layer of CeO_2, and between that and Hastelloy layer are $LaMnO_3$, MgO, Y_2O_3, and Al_2O_3 layers. From the Al_2O_3 layer to the CeO_2 layer is so-called buffer layer. There is an Ag layer underneath Hastelloy and a Cu layer on the backside. Hastelloy layer is 50 μm thick. The ReBCO layer was grown epitaxially.

The tapes were cut into about 35 mm to fit a sample holder of a variable temperature insert (VTI) and bundled with fine stainless wire. Three bundles were prepared. The first bundle was wrapped with only an aluminium foil and designated as Cd00. The second was done three times with 25 μm thick Cd foil, resulting in a minimum shielding thickness of 75 μm, which was called as Cd20. The third was wrapped five times with the Cd foil, resulting in a minimum shielding thickness of 125 μm, which was called as Cd50. All bundles were wrapped with aluminium foil and set in the aluminium alloy capsule. The aluminium foil was expected to carry the nuclear heat in the samples to the capsule and to cooling water in a fission reactor. The capsule was purged with helium gas and welded. In addition, non-irrad. samples were prepared for reference data.

2.2 Neutron irradiation

Neutron irradiation was carried out at JRR-3 in Tokai, Japan. It equips a neutron irradiation system named Rabbit. The capsule can be sent to HR-1 hole and return to the operation hall after a scheduled time. It works with light water and the Rabbit system can be operated even when the JRR-3 is under operation at 20 MW. In this study, the irradiation was carried out for 24 hours. Since the maximum fast and thermal neutron flux are 1.7×10^{16} and 9.6×10^{17} $n/m^2/s$, respectively [4], the fast and thermal neutron fluences for 24 hours irradiation are 1.46×10^{21} and 8.29×10^{22} n/m^2, respectively. After the irradiation, the capsule was sent back to the radiation control area at Oarai center in Tohoku University when the samples were cooled down.

Researchers at Atominstitut in Vienna, Austria, have carried out many irradiation tests with TRIGA MARK II, and published many papers. Since the neutron flux envelope against neutron energy of JRR-3 and TRIGA are different, both envelopes are compared. The results are shown in Figure 1. The envelope in the core of TRIGA (250 kW operation, solid line) was traced from Ref [5] and that in HR-1 hole of JRR-3 (20 MW operation, dotted line) was copied from Ref [6]. At 0.1 MeV neutron flux, TRIGA is almost twice as large as JRR-3, and at thermal neutrons, JRR-3 is about one order of magnitude larger than TRIGA. Therefore, for the same fast neutron flux, the thermal neutron flux in JRR-3 is about 50 times larger than that in TRIGA. Also, it is noticed that the flux at 0.1 MeV is almost the same as that at thermal neutrons for TRIGA.

Previous analytical studies of neutrons in magnets have focused on neutrons of 0.1 MeV or higher, and there are no reports on thermal neutrons. However, it is easy to imagine that the maximum energy is 14 MeV and the minimum energy is around a few meV. Therefore, it could be said that the energy range in Figure 1 covers almost all the energies of neutrons that reach the magnets of a fusion reactor.

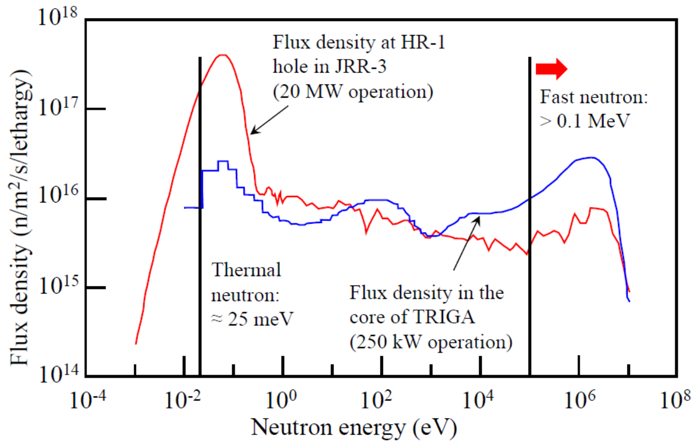

Figure 1. Comparison of neutron irradiation sources. Solid line: Envelope in the core of TRIGA MARK II. Dotted line: Envelope in irradiation hole of HR-1 in JRR-3.

2.3 Ge detector analysis and superconductivity tests

To investigate the isotopes in the ReBCO tapes, a Ge detector analysis was attempted. The tape has a 50 μm thick Hastelloy layer and ^{58}Ni in Hastelloy is transmuted to ^{60}Co during the irradiation. Gamma ray peak of ^{60}Co is very strong and other peaks cannot be seen. So, a trial to take out only the ReBCO layer from the irradiated tape was conducted. The irradiated tape was cut into about 6 to 7 mm long and at least 10 thermal cycles between about 340 degrees (iron) and about 24 degrees (water) were given to the sample. Then the sample was plastically bended many times and the coated layer was peeled and scratched out of the Hastelloy substrate. The flakes were collected and sent to the Ge detector analysis. The total weight of the collected flakes was about 1 mg. The analysis for GdBCO and EuBCO tapes were investigated after 154 and 277 days after the irradiation. The YBCO tape was tested twice, 277 and 410 days after the irradiation.

2.4 Tests for superconducting properties

A 15.5T SC magnet and VTI were installed in the radiation control area at Oarai center in Tohoku University in 2012 [7], and the test system has been advanced [8]. The ReBCO sample is set on a sample holder with a low melting point solder. It takes about 13 hours to cool down to around 5 K. The sample current is provided by a 500 A DC power supply. The critical temperature (T_C), and the upper critical magnetic field (B_{C2}) were measured as follows:

Voltage taps were soldered at the center of the sample with the distance of several mm. A sample current of 1 or 2 A was put into the sample and the temperature of the sample holder was controlled and lowered gradually. The output of voltage taps and temperatures of positive and negative electrodes were recorded. The average temperature was adopted as the sample temperature and a diagram between the voltage output and the sample temperature was drawn. The offset point was determined by linear approximation of the data in the transit region. And the offset temperature was adopted as T_C. The B_{C2} was defined as the magnetic field when the T_C was measured. Sampling rate was 50 Hz.

3. Test results and discussion

The results of the Ge detector analysis are shown in Figure 2. ^{144}Ce was detected in all tapes which means the ReBCO layer was included in the collected flakes. ^{52}Mn is also evidence that shows the ReBCO layer was collected. ^{153}Gd was found for the tape GdBCO tape and ^{152}Eu and ^{155}Eu were

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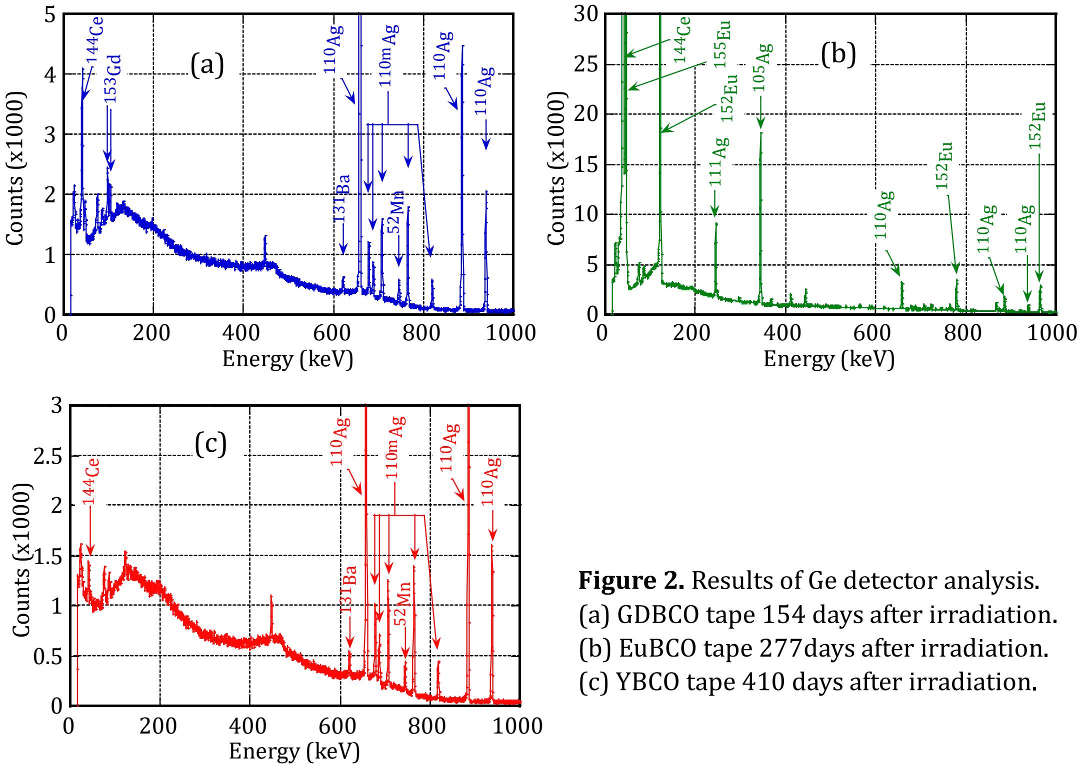

Figure 2. Results of Ge detector analysis.
(a) GDBCO tape 154 days after irradiation.
(b) EuBCO tape 277days after irradiation.
(c) YBCO tape 410 days after irradiation.

confirmed for EuBCO tape. The YBCO tape did not show any isotopes of Y. ^{153}Gd, ^{152}Eu and ^{155}Eu were produced as the result of nuclear transmutation and this fact supports the consideration that the thermal neutron was captured by Gd and Eu and {n,γ} reaction occurred.

Since B_{C2} has a following relation with T_C[9], the parameter of $T_C^{1.5}$ was adopted in this study.

$$B_{C2}(T_C) = B_{C2}(0) \text{ x } (1 - (T_C/T_C(0))^{\nu}) \qquad \text{----------} \ (1)$$

where ν is a universal parameter of 3/2. $B_{C2}(0)$ is B_{C2} at 0 K and $T_C(0)$ is T_C at 0 T.

Relation between B_{C2} and $T_C^{1.5}$ is shown in Figure 3. (a), (b) and (c) are the results of GdBCO, EuBCO and YBCO tapes, respectively. For the GdBCO tape, GdBCO Cd00 did not show the superconductivity even at 5 K. So, there is no data in the Figure. The GdBCO tape with Cd foil shielding showed superconductivity, with thicker shielding better protecting the sample from irradiation, resulting in less T_C degradation. Since Cd foil absorb thermal neutrons, the thermal neutron fluence will become an effective parameter instead of the fast neutron fluence. D. Fischer et al. reported that the irradiation with fast neutron of 6 x 10^{21} n/m^2 degraded T_C of GdBCO tape by several K using the data obtained in TRIGA [10]. The degradation of several K is very small compared to the result in this study. As shown in Figure 1, the fast neutron flux is approximately the same as the thermal neutron flux for TRIGA. So, if the neutron fluence of 6 x 10^{21} n/m^2 is considered to the thermal neutron fluence, it would be match with the results obtained here. And it suggests that the primary parameter is thermal neutron fluence, not fast neutron fluence.

For the EuBCO tape, only EuBCO Cd00 was degraded by the irradiation. This fact also means the effectiveness of neutron shielding with Cd foil. The YBCO tape did not show any degradation in superconductivity. The same results have already been reported by J. Emhofer et al. [11] as part of experiments carried out under the fast neutron (>0.1 MeV) irradiation of 1 x 10^{22} n/m^2.

The cross section of Gd, Eu and Y were investigated along with Cd using JENDL-5 database [12]. When an atom captures a thermal neutron, {n,γ} reaction happens and gamma ray emits. The results are shown in Figure 4. The unit of the cross section of the vertical axis is barn, with one

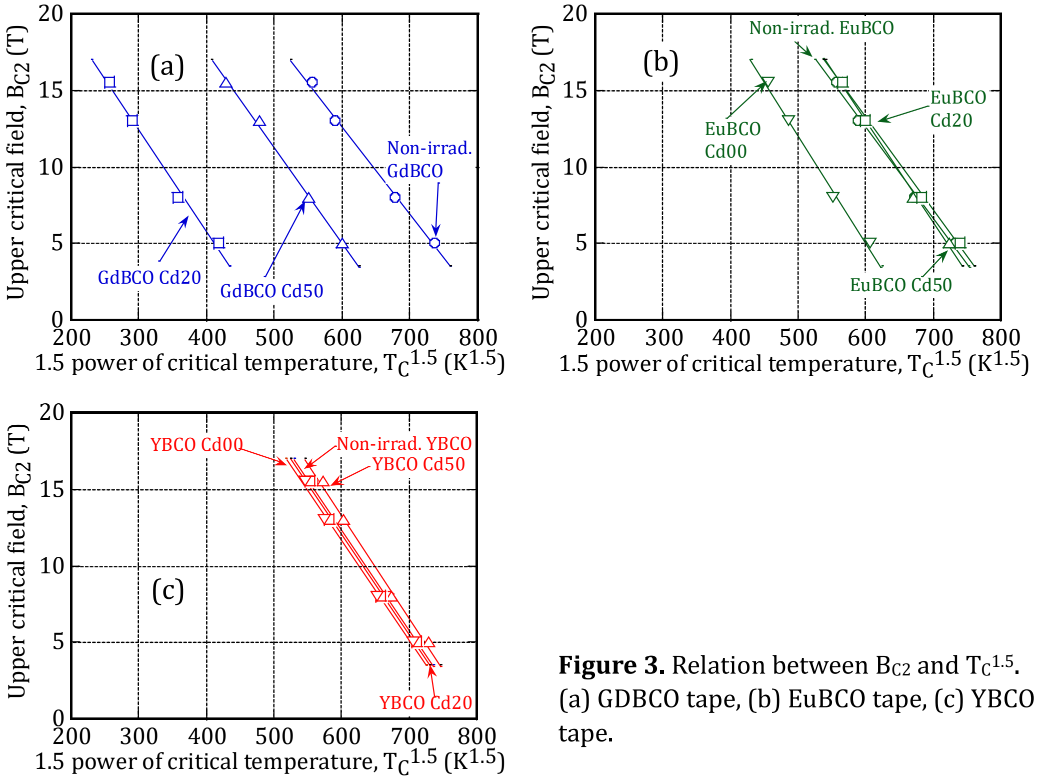

Figure 3. Relation between B_{C2} and $T_C^{1.5}$. (a) GDBCO tape, (b) EuBCO tape, (c) YBCO tape.

barn being 10^{-28} m^2. Vertical lines are drawn at 10^5 eV for the fast neutron and 2.5 x 10^{-2} eV for the thermal neutron in each diagram. In the case of Gd, following 6 isotopes occur naturally, ^{154}Gd (2.18%), ^{155}Gd (14.80%), ^{156}Gd (20.47%), ^{157}Gd (15.65%), ^{158}Gd (24.84%) and ^{160}Gd (21.86%). And for Eu, ^{151}Eu (47.81%) and ^{153}Eu (52.19%) exist. The percentage in parentheses shows natural abundance ratio. The cross sections of these isotopes were researched except for ^{154}Gd which has the smallest abundance ratio.

The cross section for fast neutron is less than 10^0 barn for Gd and Eu. Y has a very small cross section of around 10^{-3} barns. However, there is a big difference in the cross sections for the thermal neutrons. ^{157}Gd and ^{155}Gd have huge cross section on order of 10^5 barns and ^{151}Eu has that on order of 10^4 barns. ^{157}Gd and ^{155}Gd can capture the thermal neutrons and transmuted to another isotope emitting gamma ray. Though the cross section is smaller than ^{157}Gd and ^{155}Gd, ^{151}Eu and ^{153}Eu also can absorb the thermal neutrons. On the contrary, ^{89}Y has cross section of ca 10^0 barn and almost no degradation by the irradiation. Therefore, it would be a strong thinking that the thermal neutron capture would cause the degradation of GdBCO and EuBCO tapes.

For Cd isotopes, ^{113}Cd has the largest cross section of on order of 10^4 barns, which captures the thermal neutron and protects from the irradiation for GdBCO and EuBCO tapes.

4. Mechanism of superconducting degradation in GdBCO and EuBCO tapes

As presented above, the {n,γ} reaction would play a key role for the degradation of SC properties. The mechanism for the degradation was considered based on the nuclear transmutation. The image of $ReB_2C_3O_7$ crystal is shown in Figure 5. Re atom is in the center of the crystal and two Ba atoms locate above and below the Re atom. Cu atoms form planes and O atoms sit the space between Cu atoms. The Cu planes near Re atom make perovskite structure of CuO6 and SC current runs in the perovskite structures.

4.1 Gd/Eu recoil model

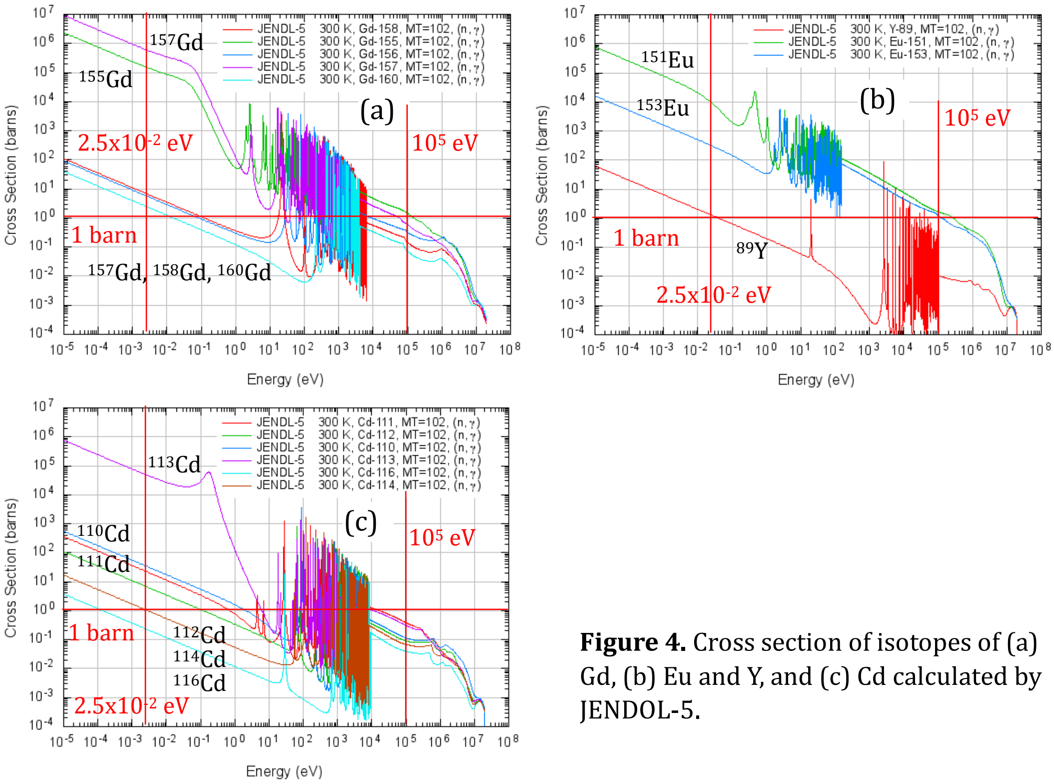

Figure 4. Cross section of isotopes of (a) Gd, (b) Eu and Y, and (c) Cd calculated by JENDOL-5.

The {n,γ} reaction emits a strong gamma ray and the gamma ray would push out Gd or Eu atom which is called as recoil motion. If Gd or Eu jumps out, vacancy is left and the balance of perovskite structure would be collapsed and SC current would stop. It is difficult to calculate how much energy is needed to cause the recoil in the presence of Ba, Cu, and O atoms. In addition, there are 33 radioisotopes [13] and half-life of most of the isotopes is very short. The transmutation path is very comprehensive, but all isotopes are transmuted to Eu or Tb. As mentioned above, only about 30% of the Gd has potential to undergo nuclear transmutation, and the other 70% are not damaged. The mechanism by which this 30% of isotopes degrades the properties of the entire tape must be considered.

4.2 Oxygen deficiency model

When the oxygen near Gd or Eu is blown away by the gamma ray emitted by the {n,γ} reaction, the perovskite structure is destroyed and SC current would stop flowing. This model is called as an oxygen deficiency model. If the additional heat treatment with oxygen gas should recover the SC properties, it would be good evidence. But a question on 30% of the natural abundance ratio exists.

4.3 Electron exchange model

The last model is electron capture or emission between the Gd or Eu atom and the free electrons in the perovskite structure. In the case of Gd, the isotopes with atomic masses below 157 are transmuted to Eu by the electron capture ($p^+ + e^- \rightarrow n + \nu^+$) and the isotopes with atomic masses greater than 158 are transmuted to Tb by the beta decay ($n \rightarrow p^+ + e^- + \nu^-$), where p^+ is proton, e^- is electron and ν is neutrino. So, the electron exchange continues in the GdBCO crystal during the nuclear transmutation. The electron exchange disturbs the electron potential and the resistance occurs in the perovskite structure. This model can be proved by the re-test of the irradiated tapes after the transmutation is complete.

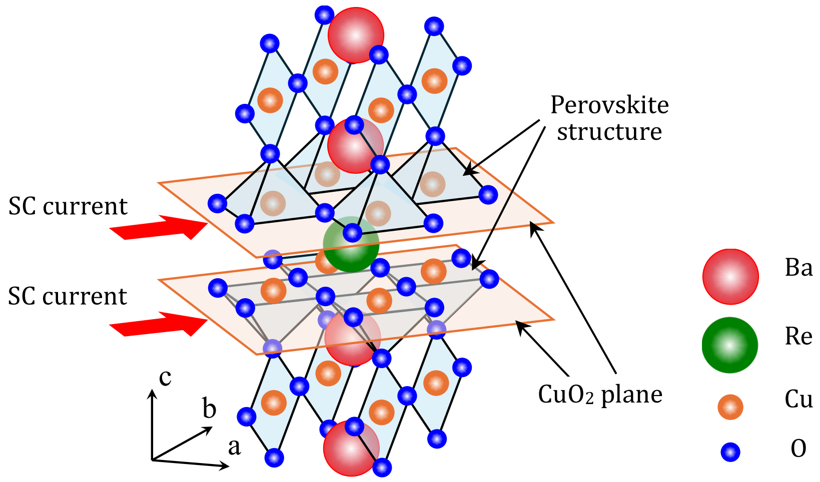

Figure 5. Image of $ReBa_2Cu_3O_7$ crystal. Rare earth atom locates at the center of the crystal sandwiched by Ba atoms. Cu atoms are on the planes between atom planes of Ba and Re.

All models discussed above include the 30% abundance ratio problem. The recoil motion, the oxygen deficiency and the electron exchange will occur only on the ^{157}Gd and ^{155}Gd or ^{151}Eu. In the case of Nb_3Sn wires, the fast neutron could make irradiation damage and pinning sites were increased. Also, it could release the internal strain resulting in some improvement of the critical current. But the thermal neutron does not have such energy. At present, it is difficult to provide a satisfactory explanation for why the remaining 70% sound region of the GdBCO tape loses superconductivity by relatively small neutron fluence. Further investigation is required to discover the mechanism on the change in the superconductivity of the GdBCO and EuBCO tapes.

5. Summary

GdBCO, EuBCO and YBCO tapes were neutron irradiated in JRR-3 and radioisotopes of these tapes were investigated by Ge detector analysis. The superconductivity was evaluated using 15.5 T superconducting magnet and the variable temperature insert. The main results are summarized as follows:

(1) The neutron energy envelope at HR-1 hole in JRR-3 in Tokai is very different from that in the core of TRIGA in Vienna. For a given fast neutron fluence, the thermal neutron fluence in HR-1 is about 50 times higher than that in TRIGA. The maximum fast and thermal neutron for 24 hours irradiation at HR-1 was 1.46 x 10^{21} n/m^2 and 8.29 x 10^{22} n/m^2, respectively.

(2) The GdBCO tape without Cd shielding did not show superconductivity even at 5 K. The GdBCO sample with Cd shielding showed superconductivity, and the thicker the shielding, the more suppressed the degradation of T_C. The Cd shielding is very effective for the GdBCO tape. For EuBCO tapes, only the sample without Cd shielding showed degradation in T_C, while the other Cd shielded samples showed almost the same T_C as the unirradiated EuBCO tape. No degradation was observed in the YBCO tape.

(3) The clear evidence of the nuclear transmutation was obtained by the Ge detector analysis for GdBCO and EuBCO tapes. It is easily expected that a lot of isotopes which do not emit gamma ray would be generated by the thermal neutron irradiation.

(4) ^{157}Gd and ^{155}Gd have cross section on order of 10^5 barns and ^{151}Eu has that on order of 10^4 barns. The huge cross section causes much nuclear transmutation resulting in the degradation of the T_C. The thermal neutron will be a proper parameter to present the degree of the degradation.

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(5) There is a big difference between the JRR-3 data and the TRIGA data when considering fast neutrons as a parameter. The data in this study showed the strong effect of the thermal neutron and almost no effect of the fast neutron was seen. If it is assumed for TRIGA that the thermal neutron fluence is roughly the same as the fast neutron fluence, the JRR-3 data and the TRIGA data are almost identical.

(6) There are three models to explain the degradation, i.e. Gd/Eu recoil model, Oxygen deficiency model and Electron exchange model. These models are considered based on the {n,γ} reaction. For GdBCO tape, about 30% Gd will be nuclear transmuted and the remaining 70% are no damaged and can carry SC current. This is not match with the fact. Further experiments and considerations on the results are needed.

Acknowledgement

The authors would like to express sincere thanks to Mr. Masanori Yamazaki at Tohoku University and Prof. Naoko Oono at Yokohama National University for their continuous contributions and discussions, and Mr. Makoto Watanabe at Tohoku University for Ge detection analysis, and Shanghai Superconductor Technology Co., Ltd for the supply of ReBCO tapes. Part of this work was supported by the NIFS Mission Realization Strategy Project 2024 (MIS013) and NIFS Collaboration Research Program (NIFS24KIGT007). This work was performed at the International Research Center for Nuclear Materials Science (Oarai center) in the Institute for Materials Research, Tohoku University.

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