14 MeV Neutron Irradiation Effect on Superconducting Properties of Nb_3Sn Strand for Fusion Magnet

A. Nishimura 1), T. Takeuchi 2), S. Nishijima 3), N. Koizumi 4), G. Nishijima 2), K. Watanabe 5)

1) National Institute for Fusion Science, Toki, Japan
2) National Institute for Materials Science, Tsukuba, Japan
3) Graduate School of Osaka University, Suita, Japan
4) Japan Atomic Energy Agency, Naka, Japan
5) Tohoku University, Sendai, Japan

E-mail: nishi-a@nifs.ac.jp

Abstract. Using 14 MeV neutron source and fission reactors, neutron irradiation tests of Nb_3Sn strand were carried out. The maximum fluence was 2.65 x 10^{21} n/m^2 for 14 MeV neutron and 4.3 x 10^{24} n/m^2 for fission neutron. By the irradiation with 14 MeV neutron, increase of I_C (1.45 times at 14 T), no change of B_{C2} (under 100 mA) and degradation of T_C (0.2 K) were measured. Magnetization of irradiated samples at fission reactors suggested that 1.0 x 10^{21} n/m^2 irradiation would increase I_C but 1.1 x 10^{24} n/m^2 irradiation would degrade I_C. The irradiation of 4.3 x 10^{24} n/m^2 made Nb_3Sn strand non superconductivity any more. These facts were considered to be caused by strengthening of pinning force and disordering of A-15 crystal with knock-on effect of fast neutron. Higher neutron fluence would increase I_C but destroy the crystal at the same time.

1. Introduction

Large scale fusion devices such as ITER and JT-60SA being under construction [1,2] require large current superconductors working in high magnetic field to produce strong magnetic field to confine high performance plasma certainly. Since they will produce 14 MeV neutrons by deuterium-tritium (D-T) reaction or 2.45 MeV neutrons by deuterium-deuterium (D-D) reaction and the generated neutrons will go out of the plasma vacuum vessel and reach superconducting magnets, though neutron shielding systems like a blanket and a double-wall vacuum vessel with water will be installed, irradiation effects of fast neutrons on superconducting properties must be understood clearly.

Since the Nb_3Sn strands have been planned to apply to ITER magnets and other fusion applications, some important investigations on the irradiation effects have been performed based on individual and separated programs [3-6]. Therefore, the knowledge on the effect and the mechanisms on the change in superconductivity are fragmentary and limited. In order to promote better understanding on the neutron irradiation effect on superconducting properties of Nb_3Sn strands and to pile up the data base systematically for the design and the construction of the fusion magnets, a collaboration network has been established in Japan [7,8] and the neutron irradiation and post irradiation test facilities have been designed and installed step by step. One of the fruitful results of this network activity is an installation of a 15.5 T superconducting magnet with thermal conduction cooling at a radiation control area in Oarai branch of Institute for Materials Research (IMR), Tohoku University [9]. The high magnetic field tests of activated samples will start at the end of 2010 fiscal year.

This paper presents the changes in the superconducting properties of Nb_3Sn strands after 14 MeV neutron irradiation and magnetization property after fission neutron irradiation and the mechanisms for the property changes are discussed.

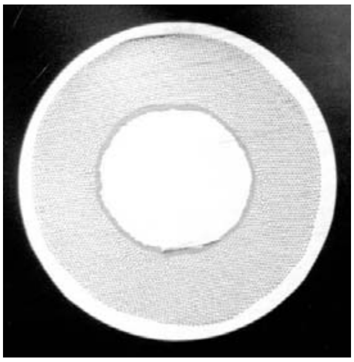

FIG.1. Cross section of Nb_3Sn strand used.

2. Test materials and procedures

Tested superconducting strand is a bronze route Nb_3Sn wire. The cross section is shown in Fig.1. The outer diameter of the strand is 0.7 mm and the matrix is niobium. The filament diameter is about 4.6 μm and number of filaments is about 4,900. The center part and the surrounding area are pure copper and the copper area ratio is about 20.5%. The residual resistance ratio (RRR) of copper is about 500. The straight strand was cut into about 45 mm long and heat treated to generate Nb_3Sn phase in vacuum.

Irradiation tests were carried out at three facilities, i.e. Fusion Neutronics Source (FNS) in Japan Atomic Energy Agency (JAEA), Japan Research Reactor No.3 (JRR-3) in JAEA, and Belgian Reactor No.2 (BR2) in Belgium. FNS equips a rotating target to produce 14 MeV pure neutrons by D-T reaction. The flux is about 7.2×10^{12} n/s. The temperatures of the samples during the irradiation were 4.5 K using GM refrigerator and RT on the cryostat surface. The samples were set in a certain distance from the D-T point and the maximum neutron fluence of 2.65 x 10^{21} n/m^2 was achieved in total at RT. The JRR-3 has a Rabbit system with cooling water to irradiate materials. Samples are set in an aluminum alloy capsule with aluminum foil and sent to a suitable position in the JRR-3 core. The system allows taking out the samples even when the reactor is under operation. The water temperature is controlled at 30 C and the sample temperature during irradiation is around 100 C. The fast neutron flux of over 0.1 MeV is 1.4 x 10^{16} or 1.7 x 10^{16} n/m^2/s., depending on the location in the core. The BR2 has larger nuclear power than the JRR-3 and the higher neutron flux is possible. The maximum neutron fluence achieved in this study is 4.3 x 10^{24} n/m^2. The samples cut out of the same wire were delivered to these facilities and the neutron irradiation was conducted.

The irradiated samples at FNS were sent to High Field Laboratory for Superconducting Materials (HFLSM) in IMR, and the critical current (I_C) and the critical magnetic field (B_{C2}) were measured using 28 T hybrid magnet. The criterion of the I_C determination was 1 μV/cm and the B_{C2} was evaluated by sweeping the magnetic field at the rate of 3.2 T/min under 100 mA of transfer current. The critical temperature (T_C) was measured using GM refrigeration system installed at FNS by raising the temperature to 20 K at the rate of 0.1 K/min and recording the voltage with a span of about 10 mm under +/- 50 mA. The magnetization of the samples irradiated at the JRR-3 and the BR2 was evaluated using Magnetic Property Measurement System (MPMS-5, Quantum Design Inc.) installed at Oarai branch of IMR. Diagrams of magnetization against magnetic field (+/-5 T) were obtained at 4.5 K and diagrams of magnetization against temperature were done under 0.005 T. A sample tested

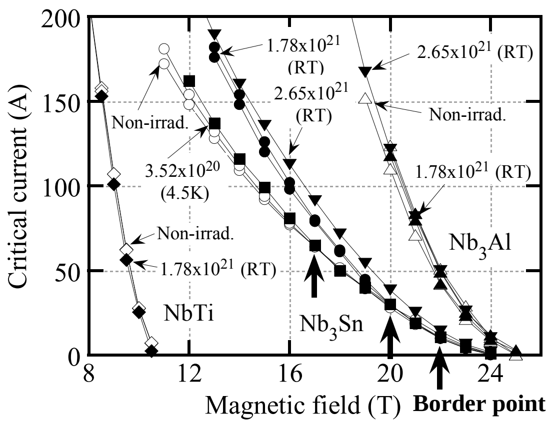

FIG.2. Change in I_C of Nb_3Sn strand by neutron irradiation.

was a few millimeters long and cut out of the about 45 mm long strand. To avoid an electrical circuit, both ends of the sample were polished with emery papers. All tests with the activated samples were carried out according to the rules on radioactive isotopes.

3. Results and discussion

3.1 Critical current, critical field and critical temperature

The dependence of I_C of the Nb_3Sn strand on the magnetic field is shown in Fig.2. The results of the NbTi and Nb_3Al strands are also presented for a comparison. The Nb_3Sn strand showed significant increase in the I_C after the 14 MeV neutron irradiation. The increase occurred in the lower magnetic field first and then the change expands to the higher magnetic field. When the border point is defined as the field where the I_C of the irradiated sample got away from the I_C curve of the non-irradiated one, the border point moves to the higher magnetic field as the neutron fluence increased. It means that the irradiation damage density would have the effect on the pinning force which affects the increase of I_C discussed later. The results of B_{C2} measurement are shown in Fig.3. At the neutron fluence of 2.65 x 10^{21} n/m^2, the B_{C2} under 100 mA did not change. The B_{C2} is related to three parameters, normal state resistivity (ρ_n), electronic specific heat coefficient (γ) and critical temperature (T_C). As mentioned later. T_C drops a little, but there are no clear information on ρ_n and γ. So, the detailed discussion on the B_{C2} is very hard at present. To discuss the effect of the neutron fluence on the I_C, the ratio of the I_C of irradiated sample to the I_C of non irradiated one (I_{C0}) is calculated and plotted

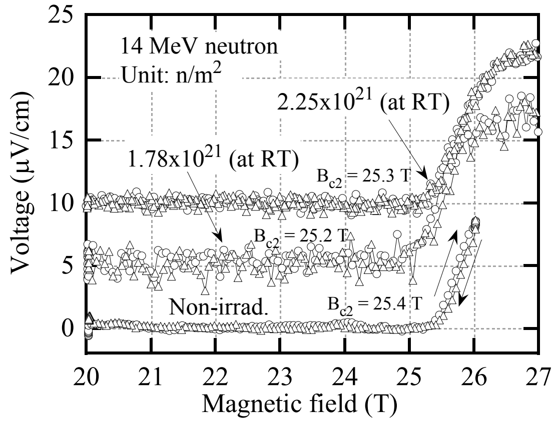

FIG.3. Results of B_{C2} measurements of Nb_3Sn strand under 100 mA.

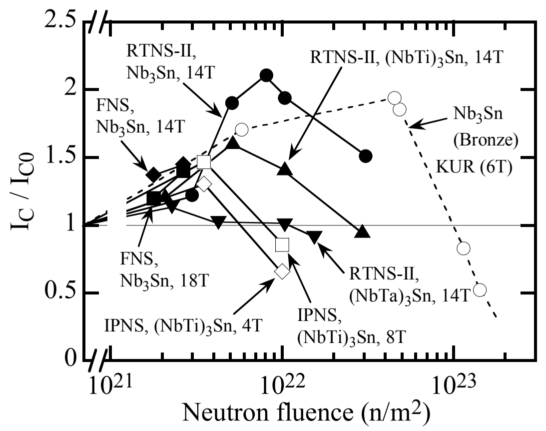

FIG.4. I_C/I_{C0} of Nb_3Sn strand against neutron fluence (> 0.1 MeV) [3-5].

against the neutron fluence as shown in Fig.4 [3-5]. As the general tendency, the I_C increases once and decreases to below I_{C0} when the neutron fluence increased. As discussed above (Fig.2), the degree of the I_C increment depends on the magnetic field and there is a tendency that the ratio becomes large in the lower field. The present results are plotted on a little bit higher ratio region than those reported before.

The results of T_C measurements are shown in Fig.5. After the 2.65 x 10^{21} n/m^2 irradiation, it was found that T_C decreased by 0.2 K. It means that the disturbance of long range ordering starts already even though the I_C is in an increasing phase. The ratio of T_C/T_{C0} presented in

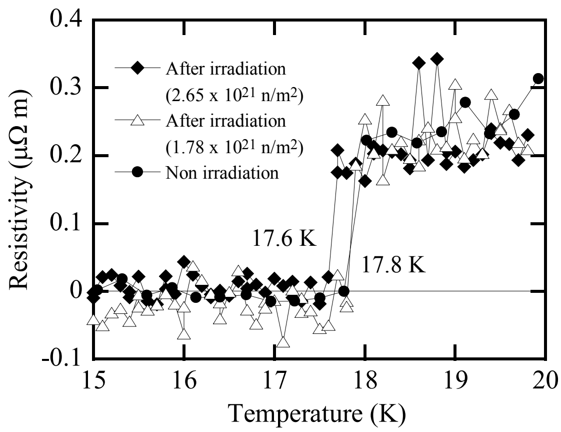

FIG.5. Results of T_C measurement of Nb_3Sn strand.

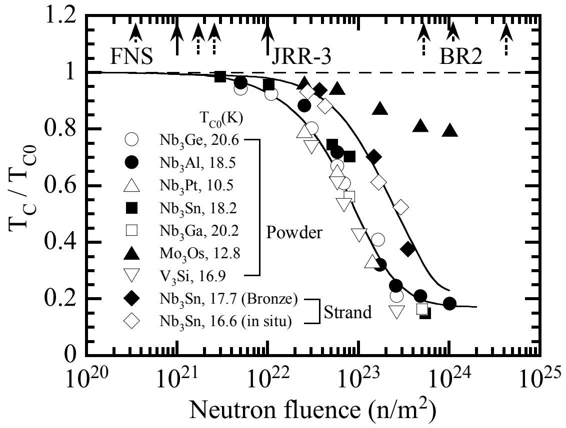

FIG.6. T_C/T_{C0} of A-15 powders and Nb_3Sn strands against neutron fluence (> 0.1 MeV) [3,6].

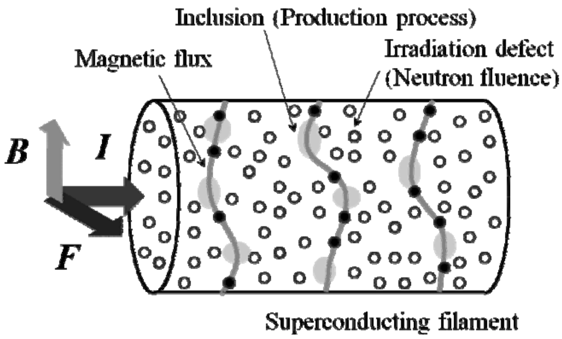

FIG.7. Schematic drawing of pinning sites in irradiated Nb_3Sn filament.

published papers was plotted against neutron fluence as shown in Fig.6 [3,6]. T_C is T_C of the irradiated sample and T_{C0} is T_C of the non-irradiated one. The T_C begins to decrease at the fluence range of 10^{21} to 10^{22} n/m^2. In the reference papers, the T_C of A-15 powders were evaluated by magnetization method [6] and the strands were studied by transferring current method [3]. Therefore, there might be some differences in T_C/T_{C0}. In the present study, the T_C is 17.6 K and the T_{C0} is 17.8 K, and the ratio becomes 0.988 at 2.65 x 10^{21} n/m^2 irradiation. This result is consistent with the data in Fig.6. The arrows in Fig. 6 show the performed neutron fluence in each neutron source and some magnetization results will be shown later.

A schematic image of the strengthening mechanism of pinning sites is shown in Fig. 7. The I_C is strongly related to pinning condition of the magnetic flux. In the case of Nb_3Sn strand, the magnetic flux passes through inclusions on grain boundaries and pinned there. When the current runs into Nb_3Sn filament, electro-magnetic force acts on the flux so that it would be moved to the vertical direction of the current. Therefore, the I_C is considered as a current when the pinned fluxes start to move gradually. When the strand is irradiated with neutrons, irradiation damage such as interstitials and vacancies installed by knock-on effect of the fast neutron would be generated, and when the irradiation defects are installed on the flux or near the flux, they would support and strengthen the flux pinning force resulting in the increase of the I_C. Since the irradiation defects would have small ability to pin the flux, the large density of the irradiation damage will be needed to increase the I_C. However, when the large density of the damage is installed, the disordering of the A-15 crystal would occur and the degradation of the T_C will start. Since the inclusions are initiated during the fabrication process of the strand, the effect of the irradiation damage (change in the pinning force) will be different depending on the production process. In addition, there will be effects of the neutron spectrum and the irradiation temperature.

3.2 Magnetization property

Diagrams of the magnetization against the magnetic field are shown in Fig.8. There are three diagrams, non-irradiated one, after the irradiation of 1.0 x 10^{22} n/m^2, and after the irradiation of 1.1 x 10^{24} n/m^2. The irradiation of 1.0 x 10^{22} n/m^2 increased the hysteresis and makes the slope at around 2 to 5 T gentle. It suggests that the I_C and the B_{C2} would increase. However, the irradiation of 1.1 x 10^{24} n/m^2 made the hysteresis very small and it means the very small I_C. From the change in T_C/T_{C0} shown in Fig.6, it is noticed that the 1.1 x 10^{24} n/m^2 irradiation would make T_C/T_{C0} around 0.2.

The enlarged diagram of 1.1 x 10^{24} n/m^2 irradiated sample is shown in Fig.9 together with the results of 4.3 x 10^{24} n/m^2 irradiation. The magnetization is very small comparing with Fig. 8

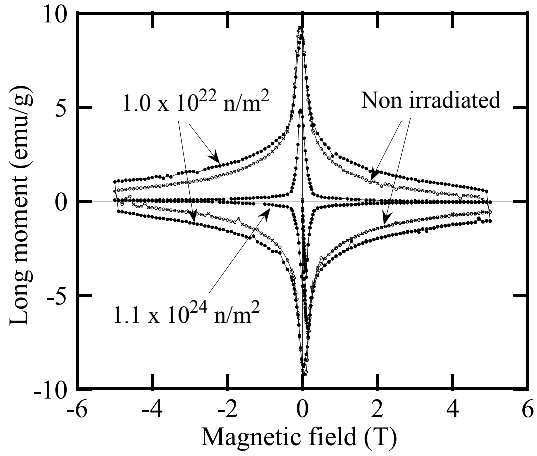

FIG.8. Magnetization of non-irradiated and irradiated Nb_3Sn strands.

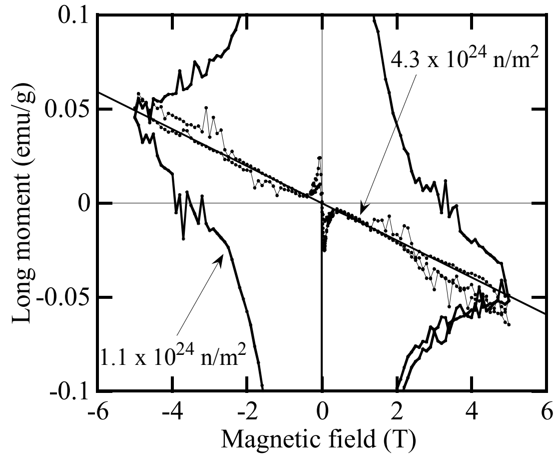

FIG.9. Magnetization diagrams of irradiated Nb_3Sn strands to 1.1 and 4.3 x 10^{24} n/m^2.

and the vertical axis is enlarged by 100 times. It is clear that the 1.1 x 10^{24} n/m^2 irradiated sample still shows weak superconducting and diamagnetism. The same diamagnetism is observed in the results of 4.3 x 10^{24} n/m^2 irradiation and it is considered that the copper caused this diamagnetism which was used for a stabilizer in the strand. The sample irradiated to 4.3 x 10^{24} n/m^2 shows no superconducting, but very small indication of the magnetization is seen in the region of +/- about 0.3 T. This would be caused by pure niobium used for producing Nb_3Sn phase. It is recognized that niobium remained still in superconducting even when Nb_3Sn already became normal.

The diagrams of the magnetization against the temperature are shown in Fig.10. Non irradiated sample shows the T_C of 17.7 K and it is a good agreement with the results shown in Fig.5 (17.8 K). In the case of the resistivity-temperature relation (Fig.5), the resistivity changed sharply and it was easy to define the T_C. However, the magnetization changes gradually against temperature, so there is small uncertainty to determine the T_C. The magnetization profile of the non irradiated one presents three critical temperatures. The lowest is about 9.2 T which is the T_C of pure niobium. The second is about 15 K and it may be the T_C of other Nb-Sn phase. The third is the T_C of Nb_3Sn phase. The irradiation to 1.0 x 10^{22} n/m^2 drops the T_C to 17.3 K and the profile is almost same as the non irradiated one. The ratio of 17.3/17.7 (T_C/T_{C0}) becomes 0.977 and this figure is good agreement with Fig.6 when it is plotted against the fluence of 1.0 x 10^{22} n/m^2. The degradation of T_C at this fluence also suggests the disordering of the crystal even though the I_C is still increasing. After the

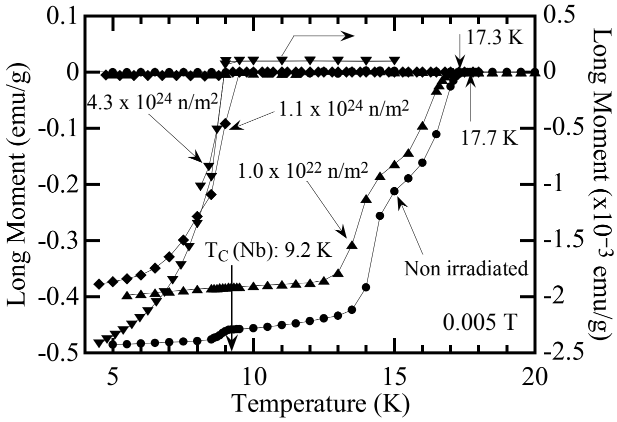

FIG.10. T_C measurement results of irradiated Nb_3Sn strand by SQUID.

irradiation to 1.1 x 10^{24} n/m^2, the T_C dropped to about 9 K which was the T_C of niobium. In the engineering sense, this situation is no use for operation and it is very difficult to detect the change in T_C during the operation under the neutron irradiation. The evaluation method for the degradation of the superconductivity under the operation must be investigated, otherwise the quench should occur suddenly and it would not be recovered. The irradiation to 4.3 x 10^{24} n/m^2 makes the strand isolator and very weak superconductivity of niobium remains.

4. Summary

The irradiation effect of the 14 MeV and the fission neutrons on the superconductivity of the Nb_3Sn strand was studied. The maximum fluence achieved was 2.65 x 10^{21} n/m^2 for 14 MeV neutron and 4.3 x 10^{24} n/m^2 for fission neutron. The main results are summarized as follows:

1. By the irradiation tests with 14 MeV neutron, the increase of I_C by 1.45 times at 14 T, no change of B_{C2} under 100 mA transport current and the degradation of T_C by 0.2 K were measured. Also, it is clarified that the increase of I_C occurs at the lower magnetic field first and the border point, where the I_C gets away from the I_C - B curve of the non irradiated one, moves to the higher magnetic field side as an increase of the fast neutron fluence. Therefore, it is expected that the B_{C2} will increase when the neutron fluence increases more and the border point passes the original B_{C2}. Since the irradiation tests with 14 MeV neutron will continue, the increase of B_{C2} will be demonstrated empirically in the near future.

2. The qualitative explanation on the I_C change was attempted based on the irradiation defects which would strengthen the flux pinning force. Also, the tiny degradation of the T_C (0.2 K) was recognized as the indication of the disordering of the crystal caused by the knock-on effect. Higher neutron fluence would increase the I_C but destroy the crystal at the same time. The balance of the strengthening and the disordering would determine the superconducting status under the neutron irradiation.

3. Magnetization of irradiated samples at fission reactors suggested that 1.0 x 10^{21} n/m^2 irradiation would increase I_C but 1.1 x 10^{24} n/m^2 irradiation would degrade I_C. The irradiation of 4.3 x 10^{24} n/m^2 would make Nb_3Sn strand non superconductivity any more. When the Nb_3Sn crystalline becomes isolator, the pure niobium still keeps the superconductivity but it becomes very weak because of the heavy irradiation.

Acknowledgments

The authors would like to express their thanks to M. Sakamoto at The Graduate University for Advanced Studies, Y. Hishinuma, T. Tanaka, T. Muroga at NIFS, Y. Izumi at University of Fukui for their contribution to perform this study, K. Takakura, K. Ochiai, T. Konno at FNS in JAEA for supports of 14 MeV neutron irradiation tests, M. Yamasaki, M. Narui, H. Kurisita, T. Shikama, K. Shirasaki, I. Sato, H. Oguro, S. Awaji at IMR in Tohoku University for supports of the fission neutron irradiation at JRR-3, operation of the radioactive materials and operation of the high field magnets, D.X. Li, Y. Honma at Tohoku University, Y. Haga at JAEA for operation of the MPMS and T. Hemmi, H. Nakajima at JAEA for the fruitful discussion, and also to Furukawa Electric Corporation Ltd. for providing the superconducting wires.

Part of this work was performed at FNS and JRR-3 in JAEA, High Field Laboratory for Superconducting Materials, Laboratory of α-Ray Emitters and International Research Center for Nuclear Materials Science under the Inter-university Cooperative Research Program of the Institute for Materials Research, Tohoku University.

This work was supported in part by Grant-in-Aid for Scientific Research (#16560725 and #22560824), NIFS-JAEA collaboration program in 2008 to 2011, NIFS collaboration research programs (NIFS04KOBF008, NIFS07KOBF014, and NIFS10KOBF018), Research programs at NIFS (NIFS06UCFF013, NIFS08UCFF005, NIFS09UCFF005, and NIFS10UFFF006).

References

[1] ITER Home page: http://www.iter.org/
[2] T-60SA Home page: http://www.jt60sa.org/
[3] T. Kuroda, K. Katagiri, H. Kodaka, M. Yuyama, H. Wada, K. Inoue, and T. Okada, "Effects of Neutron Irradiation on the Critical Temperature and Critical Current of Nb-Tube Processed Nb3Al Multifilamentary Wires," J. of Atomic Energy Society Japan, 37 (1995) pp. 652-659 (in Japanese).
[4] H. Weber, "Irradiation Damage in Superconductors," Advances in Cryogenic Engineering, 32 (1986) pp.853-872.
[5] M. W. Guainan, P. A. Hahn, and T. Okada, "Studies of Superconductors and Stabilizers for Fusion Magnets," Summary Report on RTNS-II Collaboration Research, UCID 21298 (1988).
[6] A. R. Sweedler, D. E. Cox, S. Moehlecke, "Neutron Irradiation of Superconducting Compounds," Journal of Nuclear Materials, 72 (1978) pp. 50-69.
[7] A. Nishimura, T. Takeuchi, S. Nishijima, G. Nishijima, T. Shikama, K. Ochiai, and N. Koizumi, "Neutron Irradiation Effects on Superconducting Wires and Insulating Materials," Fusion Engineering and Design, 84 (2009), pp. 1425–1428.
[8] A. Nishimura, T. Takeuchi, S. Nishijima, K. Ochiai, G. Nishijima, K. Watanabe, and T. Shikama, "14 MeV Neutron Irradiation Effect on Critical Current and Critical Magnetic Field of Nb3Sn and Nb3Al Wires," Advances in Cryogenic Engineering, 56 (2010), pp. 255-262.
[9] A. Nishimura, T. Takeuchi, S. Nishijima, K. Ochiai, G. Nishijima, K. Watanabe, M. Narui, H. Kurishita, T. Shikama, "Study on Neutron Irradiation Effect of Superconductors and Installation of 15.5 T Magnet in Hot Laboratory at IMR in Tohoku University" to be published in Fusion Engineering and Design (2010).