Application of Neutron Flux from PUSPATI TRIGA Mark II Research Reactor in Enhancing Superconducting Properties of BSCCO Superconductor

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Abstract. The PUSPATI TRIGA Mark II Research Reactor is currently been used in various applications such as Neutron Activation Analysis (NAA), Delayed Neutron Activation Analysis (DNAA), radioisotope production for medical, industrial and agricultural purposes, Neutron Radiography and Small Angle Neutron Scattering (SANS). However, there are not much research activities on utilizing neutron flux from the reactor to study properties of superconducting materials. For superconductors to be fabricated into components of electrical and magnetic devices, the sustainability of the transition temperature, TC and the transport critical current density, JC of the materials is vital to ensure the durability and effectiveness of the devices. A strong superconducting flux pinning capability of superconductors is necessary to maintain high TC and larger JC. Meanwhile, exposure of superconductors to neutron flux is able to create the self-organization of defects that are responsible in producing higher TC and JC in superconductors where the defects act as the flux pinning centres. In this paper, we report the utilization of neutron flux from the PUSPATI TRIGA Mark II research reactor in enhancing the TC and JC of bismuth-strontium-calcium-copper oxide (BSCCO) superconductor. Appropriate sample holder and shielding was fabricated as protection from radiation effect. In this work, 2212-phase BSCCO superconductor (Bi-2212 phase) samples were synthesized using the conventional solid-state reaction method. The samples underwent fast neutron irradiation with neutron flux of $2.0 \times 10^{11}$ neutrons/(cm$^2$.s) for 6 hours. Results showed that the TC of the samples improved slightly, and the JC was found to increase by more than 3 folds.

1. Introduction
The PUSPATI TRIGA Mark II research reactor known as RTP is the only nuclear research reactor in Malaysia. The reactor starts its operation in 1982 and reached its first criticality on 28 June 1982. Its compact core allows for a low critical mass and higher neutron fluxes while ensuring a high degree of safety. RTP is a pool type reactor, where the reactor core sits at the bottom of a 7-metre high Aluminium tank surrounded by a biological shield made of high-density concrete [1]. The reactor uses solid fuel elements in which the zirconium-hydride moderator is homogeneously mixed with enriched uranium with 1.0 MW thermal power [2], [3]. Demineralized water acts both as coolant and neutron moderator, while graphite acts as a reflector. The reactor was designed to effectively implement
various fields of basic nuclear science studies and widely used for training and educational purposes. It incorporates facilities for advanced neutron and gamma radiation studies as well as for applications.

On the other hand, high-temperature superconductors (HTS) are widely used in various science and engineering fields such as power transmission lines, motors and generators, transformers, and Computerized Tomography (CT scan) devices. The bismuth-strontium-calcium-copper oxide (BSCCO) superconductor such as the Bi-2212 phase is a good candidate for consideration in many applications because its phase formation is highly stable with relatively high $T_C$ and $J_C$ [4]. In order for the superconducting materials such as the BSCCO superconductors to maintain high $T_C$ and $J_C$, they must have effective flux pinning centers. Innovative technique has to be introduced to enhance the flux pinning capability of BSCCO superconductor to sustain high $T_C$ and $J_C$. One of the methods to improve the flux pinning capability of superconducting materials is through neutron irradiation [5]. The effect of radiation on superconducting materials is depending on the nature and amount of pre-irradiative defects [6]. Neutron irradiation is able to produce point defects and cascades, resulted in enhancement of $T_C$ and $J_C$ in superconductors [7].

Zehetmayer et al. in their study on Hg-based high temperature superconductor discovered that the effect of neutron irradiation is more obvious at elevated temperature due to large defects created by the irradiation [8]. Aoki et al. in their work on the effect of neutron irradiation on 2223-phase BSCCO (Bi-2223 phase) and yttrium-barium-copper oxide (YBCO) superconducting tapes found that the superconducting characteristics of the tapes did not deteriorate under irradiation flux of $6.92 \times 10^{10}$ neutrons/(cm$^2$.s) [9]. Shitamichi et al. stated that neutron radiation not only improve the flux pinning, but also causes the activation of the material that can be prevented by controlling the amount of impurities, using low activation elements and reduced thermal neutron fluence [10]. In addition, elimination of impurities happened when neutron irradiation introduces homogenous distribution of defects in the superconductor microstructure, and this contributed to higher Bi-2212 phase volume fraction [11]. In the YBCO superconductor that has similar characteristics and properties as the BSCCO superconductor, there were evidences of self-organization of defects due to neutron irradiation that creates a sharp distribution of defects concentration within the sink areas [6]. This directly caused defect less superconducting region with size much larger than the coherence length of the materials and promotes high $T_C$ and $J_C$.

The aim of this work is to apply the electron flux from the PUSPATI TRIGA Mark II research reactor at the Malaysian Nuclear Agency in conducting research on superconducting materials. Bi-2212 phase superconductor was used as the samples with small addition of nanosized magnesium oxide (MgO) particles. The nanosized MgO particles were added to improve the texture and reducing the formation of secondary phases in the Bi-2212 phase superconductor [12]. In this work, neutron flux of $2.0 \times 10^{11}$ neutrons/(cm$^2$.s) was utilized and the samples were irradiated for 6 hours. The transition temperature, $T_C$ and the transport critical current density, $J_C$ were determined using the cryogenic four-point probe equipment.

2.2. Experimental procedure

2.1. Neutron Irradiation
Neutron irradiation of samples was conducted at the Nuclear Malaysia Agency using the PUSPATI TRIGA MARK II research reactor. Water acted as coolant of the reactor since it has good thermal and nuclear properties. Furthermore, it does not capture much neutrons throughout the irradiation process. Before irradiating the samples, there were initial preparations that need to be done such as fabrication of the aluminum sample holder, and the shielding using aluminum sheet and boron carbide powder. Estimation in details of elements’ activation after termination of irradiation were also estimated. In the irradiation, neutron flux of $2.0 \times 10^{11}$ neutrons/(cm$^2$.s) was applied and the samples were irradiated for 6 hours. Thus, total fluence of $4.32 \times 10^{15}$ neutrons/cm$^2$ were exposed to the samples.
2.2. Sample preparation and characterization

The conventional solid-state method was used to synthesize the samples. Chemical powders were acquired from Sigma-Aldrich, USA. Molar ratio of bismuth (II) oxide (Bi$_2$O$_3$), strontium (II) carbonate (Sr$_2$CO$_3$), calcium carbonate (CaCO$_3$) and copper oxide (CuO) were mixed according to its ratio into composition of Bi: Sr: Ca: Cu = 2:2:1:2. The mixture was ground and heated at temperature of 800°C for 24 hours to remove impurities, and eventually resulted in the formation of Bi-2212 phase superconductor. The resultant powder was added with 5% weight percentage of nanosized MgO before reground and pressed with pressure of 7.0 tons. Each sample with mass of 2.0 g was palletized into a pellet with 13 mm in diameter and 3.0 mm in thickness. The samples were sintered at 840°C for 48 hours, then furnace-cooled to room temperature. Microstructure investigation was carried out using the Hitachi S3400N Scanning Electron Microscope (SEM). Table 1 shows the composition of each element in preparing each sample.

| Element | Mass (g) |
|---------|----------|
| Bi      | 0.9388   |
| Sr      | 0.3936   |
| Ca      | 0.0900   |
| Cu      | 0.2855   |
| O       | 0.2875   |
| MgO     | 0.0045   |

2.3. Estimation of elements’ neutron activation

Stability of nucleus in the elements may change their nucleus composition with or without any exterior influence and this feature is known as radioactivity. During nuclear decay, emission of radiation may occurs. Rate of radioactive decay of each type of sample was quantified using characterization of radionuclides half-life, $T_{1/2}$. Since radioactive decay represents the transformation of an unstable radioactive nuclide into a more stable nuclide, which may also be radioactive, it is an irreversible event for each nuclide. Mass of elements in the samples is very crucial in estimating their $T_{1/2}$. For estimation of elements’ neutron activation, the online WISE Uranium Project Calculator (https://www.wise.org) was used. Examples of input data and output parameters from the calculator are as shown in Figure 1.

2.4. Sample holder and shielding

A cylindrical aluminium sample holder of 0.05 m in diameter and 0.15 m in height was fabricated for the irradiation. Boron carbide (B$_4$C) powder was used as the filler in the sample holder. The filler is able to control the neutron irradiation from the reactor since it contains high boron’s density, non-reactive towards chemicals and excellent refractory material. During irradiation, B$_4$C absorbed neutron and decayed from boron ($^{10}$B) to helium ($^4$He) and lithium ($^7$Li). Maximum weight of 15 grams B$_4$C is needed to occupy the empty spaces inside the sample holder. Radioactivity for the 30 g of Aluminium sample holder and 15 g of B$_4$C powder was 8.359 mCi ($^{27}$Al $\rightarrow$ $^{28}$Al) and 1.103 pCi ($^{13}$C $\rightarrow$ $^{14}$C), respectively.
2.5 Measurement of $T_C$ and $J_C$

Determination of superconducting properties for superconductors are extremely crucial for industrial applications. Transition temperature ($T_C$) and transport critical current density ($J_C$) for each sample were determined using the cryogenic four-point probe equipment. Silver conductive paint was applied at the joints in between the probes and surface of the sample to ensure the connections are firm. The onset transition temperature, $T_{C,onset}$ and the zero transition temperature, $T_{C,zero}$ were determined from the plotted resistivity against temperature graph. The transport critical current, $I_C$ was determined from the $I-V$ curve using the $1\mu V/cm$ criterion. The transport critical current density, $J_C$ was calculated by dividing $I_C$ with the sample cross-sectional area, $A$.

3. Results and Discussion

Table 2 shows the radioactivity decay of the Bi-2212 phase superconductor sample. According to the estimated calculation via WISE Uranium Project Calculator (https://www.wise.org), the highest radioactivity detected is copper-63 (Cu-63) that decayed to copper 64 (Cu-64) with radioactivity of 12.76 mCi. Particular protection such as shielding is required in handling radionuclide materials and it has to take into consideration on all the safety measures to avoid any health issue and uncontrolled circulation of radioactive materials.
Table 2. Radioactive decay of each element in the Bi-2212 superconductor sample.

| Element | Mass of element | Activity | Half-life, $T_{1/2}$ |
|---------|----------------|----------|---------------------|
| Bi-209  | Bi-210         | 938.8 mg | 16.8 µCi            | 5.012 d  |
| Sr-84   | Sr-85          | 2.111 mg | 135.4 nCi           | 64.84 d  |
| Ca-40   | Ca-41          | 86.99 mg | 9.841 pCi           | $140 \times 10^3$ a |
| Cu-63   | Cu-64          | 195.5 mg | 12.76 mCi           | 12.70 h  |
| O-18    | O-19           | 650.7 µg | 18.03 pCi           | 26.91 s  |

Nuclear radiation such as neutron flux is able to create defects in the microstructure of superconducting materials, and formed flux pinning centers that will improve their superconducting properties [7]. Nevertheless, it may also lead to reduction of $T_c$ due to disorientation of grain boundaries that is strongly related to oxygen vacancy in the copper dioxide ($\text{CuO}_2$) planes and multiple lattice distortion around them [13]. Figure 2 shows the SEM micrographs of Bi-2212 phase superconductor for both the non-irradiated and neutron irradiated samples. The micrograph of sample that was exposed to neutron flux shows more random grain orientation and higher degree of texturing. However, several types of material defects such as dislocation networks, impurities and precipitates may exists within the superconducting matrix of the irradiated sample [7,11].

![SEM micrographs of (a) non-irradiated, and (b) neutron irradiated Bi-2212 superconductor samples.](image)

Table 3 shows the $T_c$ and $J_c$ of non-irradiated and neutron irradiated Bi-2212 phase superconductors. The $T_{C,zero}$ for both samples are almost similar but the $T_{C,conset}$ of neutron irradiated sample increased significantly. Variation of $T_c$ relies on the number of holes provided on the CuO$_2$ layers [13]. In such situation, the number of holes are artificially modified by the interaction between neutron flux and the CuO$_2$ layers of the Bi-2212 phase superconductor, and contributed to the enhancement of $T_c$. 
Table 3. $T_C$ and $J_C$ of non-irradiated and neutron irradiated Bi-2212 superconductor samples.

| Sample               | $T_{C,\text{zero}}$ (K) | $T_{C,\text{onset}}$ (K) | $J_C$ (A/cm$^2$) |
|----------------------|-------------------------|--------------------------|------------------|
| Non-irradiated       | 84                      | 100                      | 2.86             |
| Irradiated           | 85                      | 120                      | 10.44            |

In addition, Table 3 shows the results of $J_C$ for both of the Bi-2212 phase superconductor samples. There is a significant increase of $I_C$ in the neutron irradiated sample. Critical current, $I_C$ and $J_C$ can be enhanced by incorporating a high density of extended defects that act as effective pinning centers and resulted in significant supercurrents flow along the CuO$_2$ planes [14]. The existence of a strongly linked network of percolative paths, combined with good interplanar coupling also contributed to higher $J_C$ [15].

4. Conclusion

The potential to utilize neutron flux from the PUSPATI TRIGA Mark II research reactor to study superconducting properties of superconductor materials such as BSCCO superconductor is very encouraging. In this study, neutron flux has shown its ability to improve superconducting properties of Bi-2212 phase superconductor with neutron fluence of $4.32 \times 10^{15}$ neutrons/cm$^2$. From the SEM analysis and results of $T_C$ and $J_C$, neutron flux is able to create homogenous distribution of defects in the superconductor microstructure. There is also the possibility that the neutron flux initiates production of holes in the CuO$_2$ planes of the Bi-2212 superconductor. This indicates that neutron irradiation has the ability to enhance the superconducting properties of superconductor. As such, there are potentials for superconductors to be applied in various electrical and magnetic devices and equipment.

5. References

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