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EFFECT OF FAST NEUTRON IRRADIATION ON CURRENT TRANSPORT PROPERTIES OF HTS MATERIALS

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The effect of fast neutron irradiation with energy up to 35 MeV and integrated fluence of up to 5 x 10^{15} cm^{-2} on the current transport properties of HTS materials Bi-2212 and Bi-2223 has been studied, both at liquid nitrogen and at room temperatures. The samples irradiated were selected after verification of the stability of their superconducting properties after temperature cycling in the range of 77 K - 293 K. It has been found that the irradiation by fast neutrons up to the above dose does not produce a significant degradation of critical current. The effect of room temperature annealing on the recovery of transport properties of the irradiated samples is also reported, as is a preliminary microstructure investigation of the effect of irradiation on the soldered contacts.

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Abstract. The effect of fast neutron irradiation with energy up to 35 MeV and integrated fluence of up to 5 x 10^{15} cm^{-2} on the current transport properties of HTS materials Bi-2212 and Bi-2223 has been studied, both at liquid nitrogen and at room temperatures. The samples irradiated were selected after verification of the stability of their superconducting properties after temperature cycling in the range of 77 K - 293 K. It has been found that the irradiation by fast neutrons up to the above dose does not produce a significant degradation of critical current. The effect of room temperature annealing on the recovery of transport properties of the irradiated samples is also reported, as is a preliminary microstructure investigation of the effect of irradiation on the soldered contacts.

1. Introduction

The current leads for superconducting magnetic systems such as will be used in the Large Hadron Collider (LHC) at CERN are an evident application of High Temperature Superconductors (HTS) in large-scale projects. It is the possibility of transporting large currents without Joule losses, coupled with the low thermal conductivity of the ceramics, which makes these materials attractive for current leads. A further advantage favouring the use of HTS for these devices is that they are only required to work in relatively low magnetic field at high temperature. Thanks to the commercialization of these materials, high-current HTS current leads can now be considered as a realistic alternative to traditional resistive current leads. Potential candidates for the current-carrying elements of an HTS current lead are multifilament composite tapes using Bi-2223, prepared via the powder in tube (PIT) method [1], and rods of bulk material, Y-123 and Bi-2212 [2-4]. In particular HTS tape is manufactured in large volume, has a sufficient critical current density, and current leads based on tape can be easily prepared for a specific current value by choosing the number of tapes to be used in parallel.

In installations working in strong radiation fields current transport properties may degrade due to radiation damage in HTS material. However, the global effect of fast particle irradiation on the physical properties of the HTS material is not well understood. On the one hand, irradiation creates atomic cascades and structural defects (point defects, clusters or columnar tracks) that can in some cases give rise to an increase of critical current due to the vortex pinning on these defects, but on the other hand radiation disordering of microstructure reduces the critical temperature of HTS material, and influences negatively the inter-granular critical current [5]. There have been a number of studies on the effect of irradiation using various fast particles (ions, protons, electrons) on the physical properties of HTS materials [6-11], but the effect of fast neutron irradiation of HTS material is less well documented [12]. Moreover, tests to date have been made at reactors, where the neutrons have average energy of about 0.2 MeV, whereas the average energy of neutrons emanating from the LHC may be up to 100 MeV in the vicinity of some current leads. It is therefore of interest to know whether fast particle irradiation can give rise to a significant degradation of transport properties of HTS material. For this purpose a number of irradiation cycles with sufficient energy and fluence were carried out on samples at both cryogenic and room temperatures.

2. Preparation of samples and experimental procedure

Samples of HTS material were prepared at CERN. These consisted of two tape samples, A1 and A2, made of multifilamentary Bi-2223 in an Ag-Au matrix, and a rod sample, D2, made of 2 mm diameter bulk Bi-2212. The tape samples were tested in a sample holder consisting of two copper terminal blocks to which the edges of the tape were soldered with indium. The copper terminals were bolted to a fibreglass plate of cross-section 25 mm x 10mm. The HTS tape had no intermediate points of fastening to the fibreglass plate. The HTS rod D2 was similarly installed but was also glued with epoxy to the fibre-glass plate. Each sample was supplied with four potential contacts, two for critical current measurement and two for measuring contact resistance.

Detailed current transport measurements were carried out at RRC "Kurchatov Institute" (KI). Three voltage-current characteristics (VCC) were measured for each sample, one for the superconducting (central) part of a sample, and two for the contact bonds of the sample, using an automated system. All VCC-measurements were made in a bath of saturated liquid nitrogen. The critical current (J_c) of the sample was defined by the customary 1 μV/cm criterion. The contact resistance was defined by the slope of the linear part of the contact VCC.

Irradiation of samples by fast neutrons was made using a tailored source at KI.

3. Neutron source facility for irradiation of HTS material

The cyclotron at KI can accelerate fast protons in the energy range 10 - 35 MeV with a maximum proton current of up to 30 μA. Fast neutrons were produced at the cyclotron by bombarding a beryllium target with 35 MeV protons. The Be target, which was 6.5 mm thick and 30 mm in diameter, was mounted at the end of a long metallic tube from the cyclotron, and was insulated. During irradiation the target was air-cooled. The beam current on the target was measured by a current integrator, which in addition to instantaneous current also measured the charge of protons arriving at the target. The beam was positioned on the target by a 20 mm diameter orifice in a thick carbon block. A second current integrator also controlled the current on the carbon block. To minimize the impact of secondary electrons on the accuracy of the measured neutron beam current, the target system took the form of a deep Faraday cup.

Measurements of neutron yield and spectral distributions of fast neutrons were made in the experimental hall by a neutron time-of-flight (TOF) spectrometer at four angles between 0 and 40 degrees. The neutron events were separated from the gamma-ray events by pulse shape discrimination. The measured TOF spectra were corrected for dead time and detector efficiency, and were converted to differential neutron yields. The detector efficiency was obtained from a Monte-Carlo calculation. Neutron detection threshold was ~ 0.6 MeV.

The total neutron intensity and mean neutron energy for the full spectrum were estimated from the measurements. For small samples the total intensity is about 5 x 10^{11} neutrons $cm^{-2}s^{-1}$ and mean neutron energy is ~ 12 MeV when a beam of 32 MeV protons is used. For 100 cm^2 samples the total intensity is about 5 x 10^9 neutrons $cm^{-2}s^{-1}$, with the average energy of fast neutrons 17 MeV.

The absolute accuracy of total neutron flux is estimated to be 30%, but the relative accuracy is better at ~ 5%. The typical measured energy distribution is shown in Fig. 1.

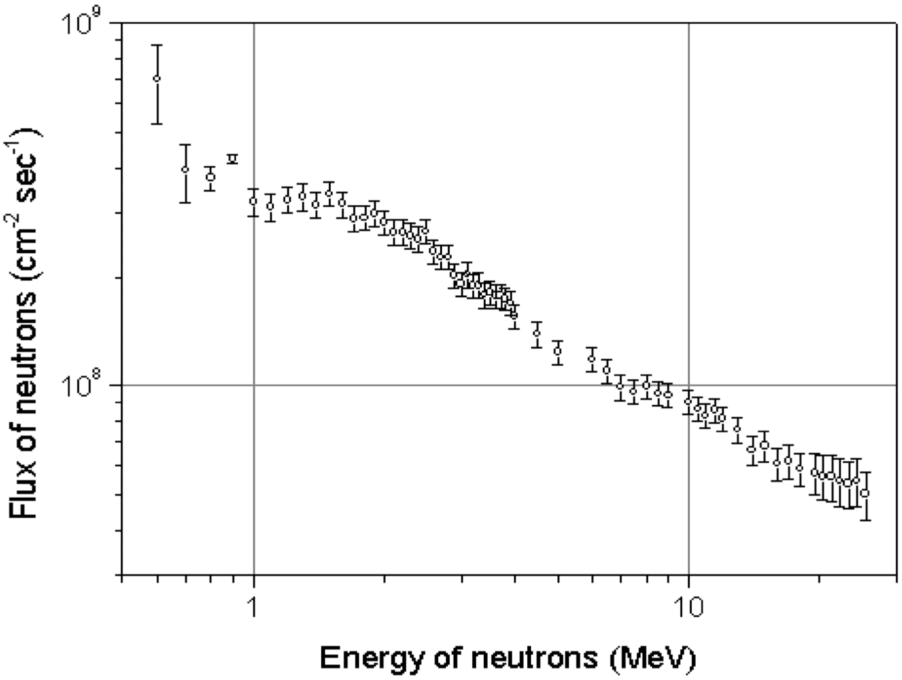

Figure 1. Energy spectrum of fast neutrons used to irradiate samples.

4. Results

For irradiation of the samples at liquid nitrogen temperature a cryostat was manufactured using foam thermo-insulation. The cryostat was equipped with a liquid nitrogen level control system to ensure a constant temperature of the sample during the cycle of the experiment. Voltage-current characteristics were recorded directly before and after irradiation, so that samples were at all times in liquid nitrogen. Superconducting transport properties were re-measured for the irradiated samples after heat-cycling to room temperature for several days to measure the effect of such annealing on eventual radiation-induced defects.

4.1. Preliminary test of stability

Before performing the experiment on the samples by irradiation with fast neutrons it was necessary to check that eventual changes of the superconducting properties are caused by irradiation rather than by degradation due to temperature cycling. Examination of temperature cycling effects is also of some practical interest for HTS current-carrying elements. For this purpose the VCC of all samples were measured several times with intermediate sample warming and drying at room temperature. The samples were cooled by insertion into liquid nitrogen, the process taking ~ 30 seconds. After measuring in liquid nitrogen samples were withdrawn from the dewar and allowed to warm up to room temperature during a few hours. Such cooling and warming was certainly hard on the samples, exposing them to temperature shock and mechanical loads, but the goal was to select sufficiently robust samples. Tape samples showed a slight inelastic slackening; those samples for which critical current did not change were deemed suitable candidates for the irradiation tests. Due to a fastening problem with sample A1 the tape was slightly torn near to a copper terminal. The sample was repaired with a soldered shunt and performed well. The bulk rod samples were unchanged after heat cycling.

4.2. Irradiation of HTS materials by fast neutrons

For the first cycle of neutron irradiation at liquid nitrogen (LN_2) temperature the bulk rod sample D2 was selected. The sample was equipped with three nickel foils for radiation dose measurement, one at the centre of the rod, and two near the contacts. The designed dose for the central part of a sample was 10^{15} cm^{-2}, for contacts 4 x 10^{14} cm^{-2}. The analysis of nickel data confirmed the radiation dose received by the sample.

Directly before the irradiation process (in the cryostat) the VCC measurement for the superconducting part of the sample and the contact bonds were carried out. It appeared that the critical current had decreased by 2 A (to 42.5 A) compared with the measurement 10 months earlier. In addition the contact resistance had increased by 10 %. As temperature cycling does not give rise to a change in critical current, this degradation must be attributed to aging. Just after irradiation, without warming of the sample, VCC measurements were carried out, and a further decrease of the J_c of 1.5 A (to 41.0 A) was observed. The contact resistance remained constant. The sample was slowly warmed up to room temperature and left to deactivate. After two weeks when the activation had decreased to a safe level, the VCC measurements were repeated. The critical current was found to have increased by 2.7 A (up to 43.7 A), i.e. to more than the value of J_c before irradiation. In Fig. 2 are shown the VCC of sample D2 at all stages of the experiment.

The second cycle of an irradiation, up to the same dose as the first run, was carried out on tape sample A1. A third cycle was made to see the effect of irradiation of the samples at room temperature (RT). Samples A1, A2 and D2 were assembled as a single package and installed on an insert for VCC measurements and irradiated. As the cryostat was not required, samples were placed closer to the source so the radiation dose was higher than for the low temperature irradiation, being ~2 x 10^{15} cm^{-2}. Dose was measured using nickel foils.

Observations concerning results after irradiation cycles are summarized as follows:

1) Sample D2 (bulk) was irradiated twice: at LN_2 and at room temperature. Low-temperature irradiation and subsequent annealing gave a slight increase of Jc. Aging for another six months at RT led to a diminution of J_c of 1 A (from 43.5 A to 42.5 A). The irradiation at room temperature of up to 2 x 10^{15} n/cm^2 did not affect J_c (see Fig. 2). It was noted that the contact resistances did not change significantly on irradiation and subsequent annealing at room temperature.

2) Sample A1 (tape) was also irradiated at both LN_2 and room temperatures. Fourteen months of aging at room temperature before the first irradiation gave rise to a negligible degradation of critical current (0.3 A or 1%). Low temperature irradiation with a dose of 10^{15} cm^{-2} did not produce a change, and subsequent annealing at room temperature gave a small increase in J_c (0.5 A or 2%). The subsequent dose (2 x 10^{15} cm^{-2}) at RT produced a further increase in J_c of 0.5A (2 %). The contact resistance remained unchanged. The VCC of sample A1, which was subjected to total dose of 5x10^{15}n/cm^2, are shown in Fig. 3.

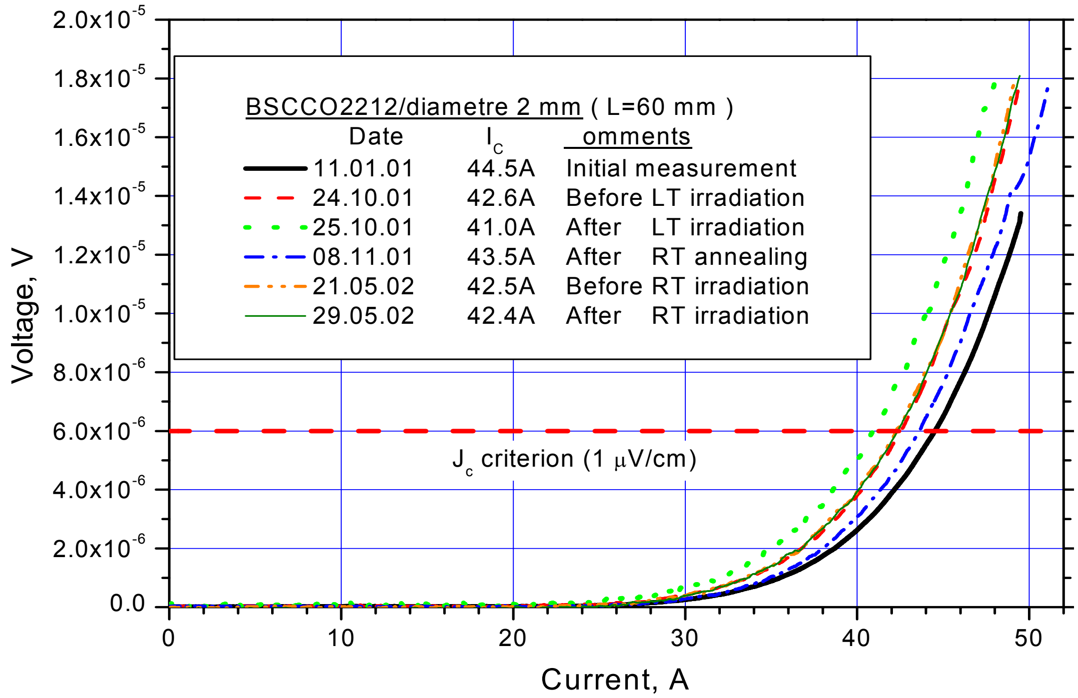

Figure 2. Voltage–current characteristics of sample D2 (bulk material).

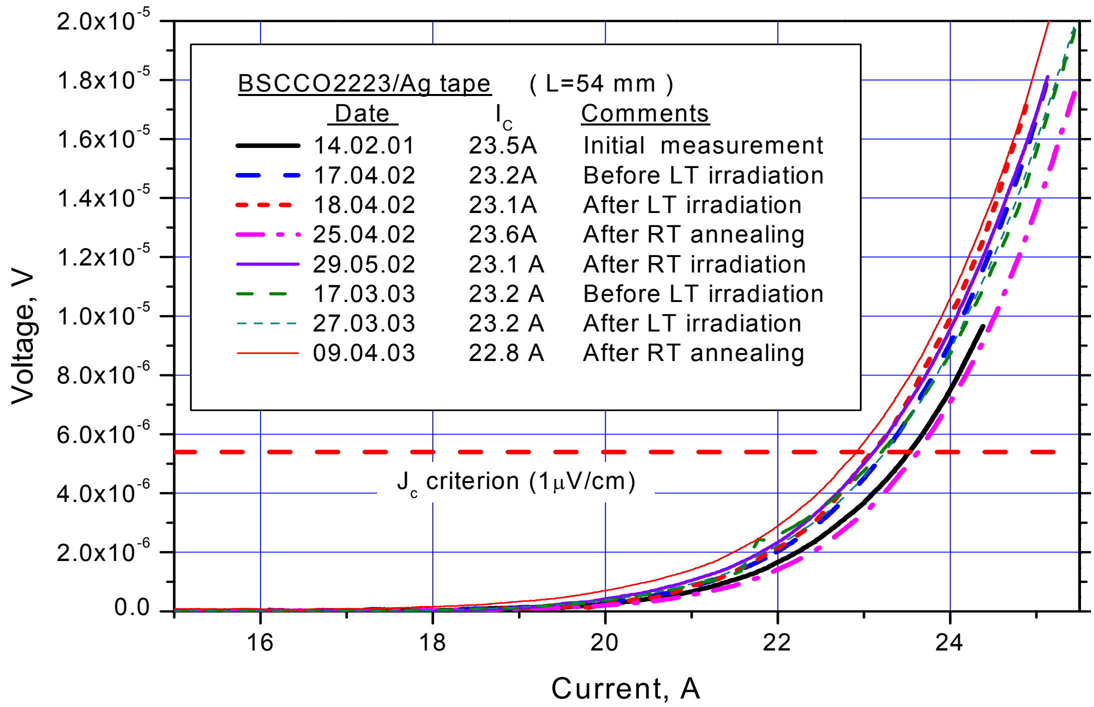

Figure 3. Voltage-current characteristics of sample A1 (tape material).

3) Tape sample A2 was irradiated only at room temperature, with a dose of 2 x 10^{15} cm^{-2}. Fifteen months of aging at room temperature gave a very small degradation of critical current, to 26.4 A (by 0.5 A or 1.7 %). The irradiation did not produce a change of critical current or contact resistance.

5. Discussion

Experiments on the irradiation by fast neutrons have unequivocally shown that a fluence of up to 5 x 10^{15} cm^{-2}does not give in an appreciable degradation of critical current of the HTS samples prepared both from Bi-2223 tape and Bi-2212 bulk material. Any changes are at the level of some percent only. A careful literature search on the effect of irradiation by fast particles on HTS properties was also made. It is well known that the neutron irradiation gives rise to the formation of spherical clusters with the average size of 1-5 nm and with linearly increasing density of clusters up to ~10^{16} cm^{-3} when the dose of fast neutrons is increased to 10^{18} cm^{-2} [13]. This implies that the cluster density is about 5 x 10^{13} cm^{-3} in our experiment, and the mean distance between clusters is about 300 nm. While not being sufficient to seriously affect the fundamental superconducting properties of a material (e.g. critical temperature), the size of defect clusters is enough that they can serve as additional pinning centres for vortices and can affect current transport properties. In papers [14-17] irradiation by fast neutrons was reported to produce an increase of critical current of materials in which initially the critical current density was low (in our case this is the bulk ceramic rod). In these materials the initial density of pinning centres is insufficient, and irradiation leads to the formation of additional pinning centres, thus raising critical current density. For the tape, however, where initially there are a large number of defects, a reduction of critical current density in a zero external field has been observed at a radiation dose of 4 x 10^{17} cm^{-2} [12]. The authors explain such a reduction by fracture of loose couplings on inter-granular boundaries. It should be noted that this radiation dose is two orders of magnitude higher than in our experiments. The somewhat unexpected result observed with the low-temperature irradiation of our bulk HTS rod sample and its subsequent annealing is explained as follows. The small reduction of critical current following the low-temperature irradiation is probably caused by point defects and dislocations of an oxygen sub-lattice in the crystal. The subsequent annealing at room temperature led to the recovery of an affected oxygen subsystem and consequent restitution of critical temperature, but has not changed the defects of the cationic subsystem providing pinning centres. This led to a perceptible growth of the critical current.

While it was not the primary subject of the study, it is relevant to discuss a problem of stability of superconducting properties of some samples at temperature cycling. The method of support should preclude exposure to strain exceeding 0.15% in all operation modes. Special attention should also be paid to the most critical section of a sample immediately near to the soldered contact, where the peak mechanical stress can be expected. The LHC lead design satisfies these requirements [18].

With regard to the degradation of critical current of the bulk rod D2 due to aging, our explanation is a possible chemical interaction between the HTS material and components of the epoxy compound used for fixing the sample to the fibreglass plate. The properties of another sample which was self-supporting remained stable during temperature cycling and after aging for more than a year.

6. Study of contact microstructure in HTS samples after neutron irradiation

The soldered contacts between the shell (matrix) of the HTS material and copper bus in the tape samples were also investigated. This study considered the uniformity of the connection, composition of the solder, and characteristics of electrical joints. A scanning electron microscope DSM-960 equipped with a crystalline spectrometer 'Microspec' was used for the microstructure investigations. The spectrometer is used to determine the composition by measuring the wavelengths of characteristic X-ray radiation.

Before the investigation the irradiated sample was polished in three steps. First, a polishing paper with grain size of 5 μm was used, followed by polishing with a diamond paste having grain size 1 μm. After the mechanical polishing, the surface was polished chemically by immersion in carbon tetrachloride and ultrasonic cleaning.

One can see in photos made by a secondary electron method that the contact between the solder material and copper terminals differs from that between the solder and the shell of HTS tape. There is a transitional layer (of less than 10μm thick) between the solder and the copper. This layer provides a good contact. The contact between the solder and the HTS shell resembles more a mechanical junction between two metals. In Fig. 4 are shown the characteristic forms of the junctions. The boundary region between the solder material and transitional layer contains many precipitates. The boundary region between the HTS shell and solder has many inclusions that can lead to cracks and have a strong impact on the electrical characteristics. The solder is indium; both In and O atoms are observed in its composition. Solder regions enriched by Cu and Ag were also observed.

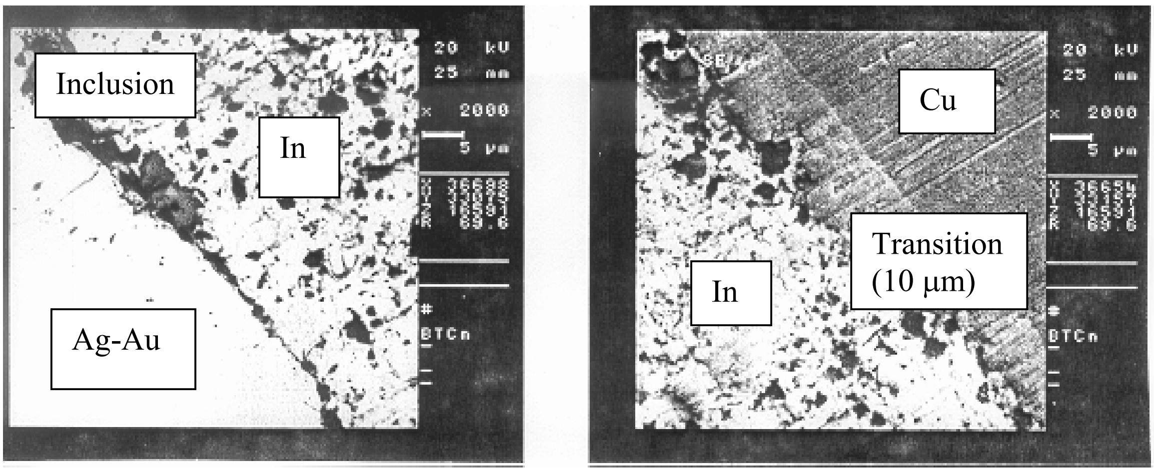

Figure 4. Micrographs of soldered contacts after irradiation.

A secondary conclusion of the present study is thus that one should avoid the use of hand-soldered indium electrical joints for cryogenic use in radiation fields. Future investigations of radiation resistance of current leads used in the LHC should address in more detail the effect of high doses of radiation on the change of electrical and mechanical properties of joints, including the micro-crack formation. In the LHC leads the solders used are Sn-Ag and Pb-Sn eutectics, and soldering is done in vacuum [18].

7. Conclusions

Irradiation of HTS material samples by fast neutrons, simulating actual radiation fields in the LHC better than is possible at a reactor facility, was carried out at room temperature and at liquid nitrogen temperature. It was found that irradiation up to integrated doses of up to 5 x 10^{15} cm^{-2}, equivalent to about 500 kGy, does not give a significant degradation of critical current of the HTS elements. It has also been observed that annealing of bulk samples irradiated at low temperature re-establishes, and may even slightly increase, the critical current. This can be explained by the generation of additional pinning centres due to irradiation by fast neutrons.

Microstructure investigations of contacts in current leads show that the boundary region between the solder material and transitional layer on the terminal side includes many precipitates and the boundary region between the HTS tape matrix and solder can have micro-cracks that could affect the electrical characteristics in the longer term. Future tests on the vulnerability of contacts should feature both joints and support which resemble those chosen for the LHC leads.

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