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Effect of 14.1 MeV fusion neutron irradiation on YBCO thin films and commercial REBCO tapes

V. Pinto, G. Celentano, M. De Angelis, F. Laviano, A. Masi, A. Pietropaolo, M. Tomellini, and D. Torsello

Abstract— The design of new tokamak machines relying on the use of high temperature superconductors (HTS) is promoting the study of HTS properties at the operating conditions required by fusion applications. In particular, the interest in the damage induced by neutron irradiation on $REBa_2Cu_3O_{7-\delta}$ (REBCO, RE = Y or lanthanide series), one of the most used family of HTS, has recently risen and several studies have been devoted to radiation hardness tests performed with ion irradiation or fission neutrons.

In this work, the effect of neutron irradiation on YBCO films and commercial REBCO tapes was investigated using, for the first time, neutrons produced by the D-T fusion reaction. The experiment was carried out at ENEA-Frascati Neutron Generator (FNG) where a deuteron beam is accelerated up to 300 keV and directed on a tritiated target to produce a nearly isotropic 14.1 MeV neutron field via the $T(d,n)\alpha$ fusion reaction. Different YBCO films deposited through metal-organic decomposition (MOD) route on single crystals ($SrTiO_3$ and $LaAlO_3$) and REBCO commercial tapes, grown by pulsed laser deposition, were irradiated. Samples exposed to three fluences were compared with a maximum neutron fluence of $1.2 \cdot 10^{14} \text{ cm}^{-2}$. The properties of HTS materials were assessed before and after irradiation by means of different techniques. From these measurements, no significant effect on the considered properties

was recognized indicating the robustness of films up to the explored irradiation fluences.

Index Terms—coated conductors, fusion neutron, HTS, MOD, neutron irradiation, radiation hardness, $REBa_2Cu_3O_{7-\delta}$ REBCO, $YBa_2Cu_3O_{7-\delta}$, YBCO.

I. INTRODUCTION

THE development of compact fusion reactor designs promises to deliver working fusion plants in just a few years [1] thanks to the possibility of carrying out smaller and cheaper experiments and commercial endeavors [2]. The main enabling technology is that of high temperature superconducting (HTS) materials and cables [3], [4], that can generate and withstand higher magnetic fields, and therefore allow efficient plasma confinement in smaller volumes. However, this new opportunity comes with new difficulties, related to the properties and operation of HTS magnets in a fusion plant. A crucial issue is represented by neutron irradiation, since the plasma will generate a high flux of 14.1 MeV neutrons that will interact with all the reactor materials, including the HTS that will be relatively close to the plasma chamber and protected by limited shielding due to the compactness of the reactor design [5]. It has been shown that the neutron spectrum impinging on the HTS magnets comprises a non negligible component of 14.1 MeV neutrons, in addition to a broad distribution spanning several orders of magnitude in energy [6], [7]. In particular, the fluence of the 14.1 MeV component of the neutron spectrum computed for 10 years of ARC operation was estimated to be between $3.0 \times 10^{18} \text{ cm}^{-2}$ [7] and $6 \times 10^{19} \text{ cm}^{-2}$ [8].

Up to now, experimental investigation of neutron damage in HTS materials mostly employed fission spectra [9]–[17], since experimental fission reactors are available and allow achieving high fluences. However, such spectra do not include the 14.1 MeV component expected in compact fusion reactors employing the D-T reaction, that can produce larger defect cascades than fission neutrons [7]. For this reason, here we focus on the irradiation at room temperature of HTS materials (YBCO thin films on $LaAlO_3$ (LAO) and $SrTiO_3$ (STO) substrates and commercial REBCO tapes) with a

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monochromatic 14.1 MeV neutron beam obtained from the D-T reaction accelerating a high intensity D beam on a tritiated target at the Frascati Neutron Generator (FNG) facility in the Frascati Research Center of ENEA. Structural, by means of X-Ray diffraction (XRD), d.c. electric and electromagnetic (magnetic measurements and visualization) measurements were carried out on pristine and irradiated samples. The results indicate that at the explored neutron fluences, negligible effects are observed on YBCO films and REBCO tapes.

In the next future, improvements of the FNG facility are planned in order to allow irradiation experiments at higher neutron fluences and at cryogenic temperatures.

II. EXPERIMENTAL

A. Frascati Neutron Generator Facility

The FNG facility [18], [19] is an accelerator-driven fusion neutron source operating at the ENEA Frascati Research Center.

FNG relies on an ion source which produces deuterons. These are accelerated up to about 30 keV by means of an electric field and focalized by an Enizel lens towards a bending magnet which acts as a mass selector, allowing only D^+ ion to enter into the linear electrostatic accelerator to be further accelerated up to about 270 keV.

The accelerated ions are focalized by means of a set of quadrupoles magnets onto a titanium-coated copper substrate where deuterium or tritium is implanted (Fig. 1).

This way, FNG can be operated in two different modes: 1) DD mode; 2) DT mode.

In DD mode, almost monochromatic 2.45 MeV neutrons are produced with a nominal emission rate of 10^9 s^{-1} .

In DT mode, almost monochromatic 14.1 MeV neutrons are produced with a nominal emission rate of 10^{11} s^{-1} . Fig. 2 shows the iso-flux contours plot calculated per primary neutron emitted at the target. The neutron emission features an energy span over the interval $0-90^\circ$ from $\sim 14.7 \text{ MeV}$ to 14.1 MeV , respectively, because of the kinematics due to the incident deuteron momentum.

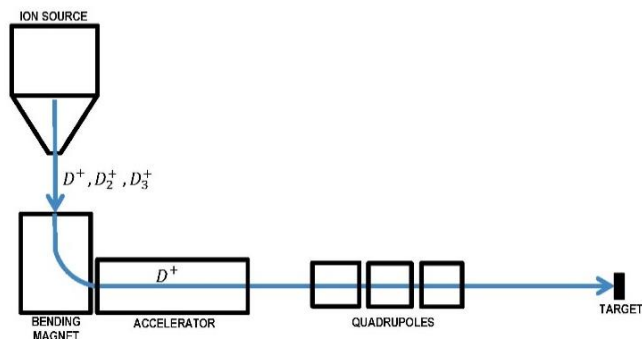


Fig. 1. Schematic representation of Frascati Neutron Generator.

B. Sample Details and Experimental Setup

YBCO samples were prepared by the low-fluorine metal-organic decomposition (MOD) approach according to the procedure described in [20]. In order to check whether the substrate nature has an effect on the modification of properties induced by neutron irradiation, the films were deposited both on STO and LAO single crystals supplied by MaTeck (Material Technologie & Kristalle GmbH). A final thickness of 80 nm was obtained with a single deposition. Bilayer films with a final thickness of 150 nm were produced performing two consecutive pyrolysis steps.

The irradiation test was carried out also on REBCO commercial tapes grown by pulsed laser deposition, namely i) YBCO + Y_2O_3 supplied by SuperOx (SO), batch 1175R, with Ag and Cu stabilizer thickness of 2 and 5 μm respectively, characterized by $I_c @ 77\text{K}, \text{S.F.} \sim 180\text{A}$; and ii) EuBCO+BaHfO₃ supplied by Shanghai Superconducting Technology Co., Ltd. (SST), with Cu stabilizer thickness of 10 μm characterized by $I_c @ 77\text{K}, \text{S.F.} \sim 200\text{A}$.

For this experiment, samples were attached through a kapton biadhesive tape to a polystyrene sheet. A multilayered structure was obtained superimposing three polystyrene sheets that were consecutively removed at different time intervals corresponding to three irradiation doses. The multilayered structure was protected by teflon tape and located at the maximum of the neutron radiation flux by fixing it in direct contact with the FNG target back side (Fig. 3).

Commercial tapes and 80 nm thick YBCO-MOD films were irradiated with three different fluences, namely 0.4, 0.8 and $1.2 \cdot 10^{14} \text{ cm}^{-2}$, by varying the exposure time. The 150 nm thick YBCO-MOD films were irradiated with the $1.2 \cdot 10^{14} \text{ cm}^{-2}$ only. The irradiation test was carried out at room temperature and with a flux of $1.9 \cdot 10^9 \text{ cm}^{-2} \cdot \text{s}^{-1}$.

C. Characterizations

The YBCO-MOD structural properties were assessed before and after irradiation through X-Ray diffraction analysis performed by a Rigaku Geigerflex Diffractometer with Cu K_α radiation in the Bragg-Brentano configuration.

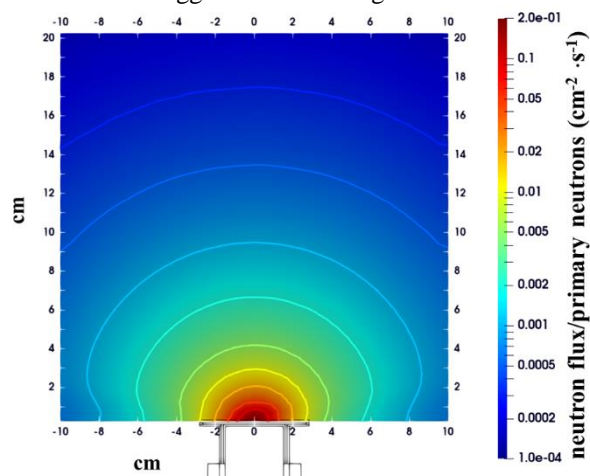


Fig. 2. The iso-flux contour plot relative to D-T reaction. The intensity is given in neutrons per primary neutron emitted at the target (see text for details).

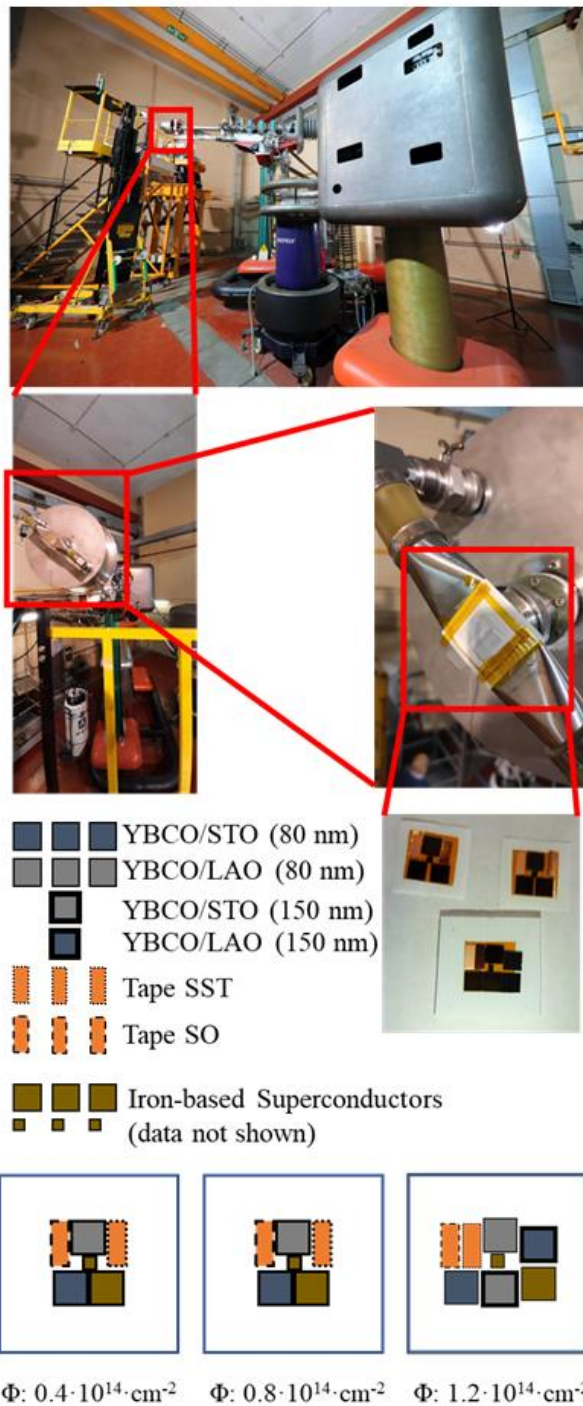


Fig. 3. Pictures of FNG facility and schematic representation of samples setup.

The zero-resistance critical temperature (T_c) value of pristine and irradiated samples was determined by means of $R(T)$ measurements carried out through d.c. electric analysis using the four-probe configuration. The residual resistance ratio (RRR) was calculated as the ratio among the resistances measured at 290 K and 100 K ($R_{290\text{ K}}/R_{100\text{ K}}$). Characterizations on irradiated samples were carried out within a few weeks after the exposure to the neutron radiation.

Magneto-optical imaging (MOI) with an indicator film [21]

was used for visualizing the magnetic field distribution in the critical state, in order to evaluate macroscopic inhomogeneities/damages and differences of the critical current density between samples.

III. RESULTS

No relevant effect on the structural properties of YBCO-MOD films was evidenced by XRD analyses. In fact, each sample was analysed pre and post irradiation. Comparable spectra were recorded independently from the neutron fluence without any relevant change as regards (00 ℓ) YBCO peaks position, shape and intensity distribution (Fig. 4). The calculated c lattice parameters are summarized in Table I. Moreover, no changes are detected on substrate related peaks as well. Likewise, similar T_c values were measured in pristine and irradiated YBCO-MOD and REBCO commercial tapes. Negligible differences were observed also in measurements of the RRR parameter of MOD deposited films (Table I), as well as in MOI characterization where no damages or appreciable differences in the critical current density were found.

Therefore, regardless of substrate, deposition technique and film thickness, it would appear that, up to the explored doses, neutron irradiation does not damage the HTS materials considered in our study.

This outcome is consistent with the results of damage calculations carried out through Monte Carlo simulations. The code PHITS can give a direct estimate of the damage in the target material in terms of displacement per atom (dpa) [22]. In the simulation, pure YBCO was exposed to a flux of 14.1 MeV neutrons. For a fluence of $1.2 \cdot 10^{14} \text{ cm}^{-2}$ the expected damage is $6 \cdot 10^{-7}$ dpa. Typically, HTS materials show properties degradation starting from 0.1-1 mdpa. Our finding is in line with the T_c degradation that can be estimated employing the two T_c degradation rates given by [16], for which using our dpa value the expected T_c variation is about 2-3 mK. Instead, using the degradation rate reported in [17], the expected T_c variation should be around 1 mK. Such a low expected T_c variation is significantly below our instrumental resolution and compatible with our results. In the future, a different experimental setup for $R(T)$ measurement will be tested in order to reduce our error for commercial tapes to tens of mK.

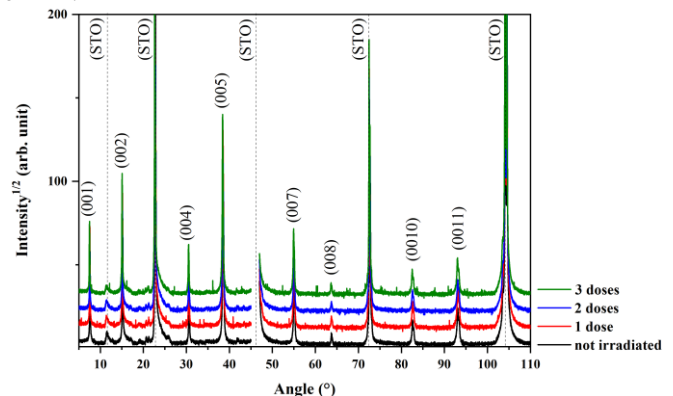


Fig. 4. XRD θ - 2θ patterns of pristine and irradiated YBCO-MOD films deposited on STO.

TABLE I

c LATTICE PARAMETER, OBTAINED FROM XRD ANALYSES, CRITICAL TEMPERATURE (T_c) AND RESIDUAL-RESISTANCE RATIO R_{290K}/R_{100K} (RRR), DERIVED BY ELECTRIC MEASUREMENTS, FOR PRISTINE AND IRRADIATED SAMPLES

| | | Not irradiated | 1 dose | 2 doses | 3 doses |
|---|---------------------------------------|-------------------|----------------|----------------|----------------|
| Lattice parameter c (Å)^a | | | | | |
| YBCO/STO 80 nm | pre ^b post ^b | 11.67 | 11.69 11.68 | 11.69 11.69 | 11.69 11.70 |
| YBCO/LAO 80 nm | pre ^b post ^b | 11.68 | 11.69 11.68 | 11.69 11.69 | 11.69 11.69 |
| YBCO/STO 150 nm | pre ^b post ^b | 11.70 | - | - | 11.69 11.69 |
| YBCO/LAO 150nm | pre ^b post ^b | 11.68 | - | - | 11.67 11.69 |
| T_c (K) | | | | | |
| YBCO/STO 80 nm | | 90.2 ^c | 89.8 | 90.0 | 90.5 |
| YBCO/LAO 80 nm | | 90.2 ^c | 89.6 | 89.8 | 90.6 |
| YBCO/STO 150 nm | | 89.5 | - | - | 90.2 |
| YBCO/LAO 150nm | pre ^b post ^b | 90.5 | - | - | 90.5 90.5 |
| Tape SST | | 93.2 ^d | 93.2 | 93.2 | 93.2 |
| Tape SO | | 87.8 ^d | 87.9 | 87.8 | 87.9 |
| RRR | | | | | |
| YBCO/STO 80 nm | | 3.4 ^e | 3.0 | 3.2 | 3.3 |
| YBCO/LAO 80 nm | | 3.0 ^e | 3.2 | 2.9 | 3.2 |
| YBCO/STO 150 nm | | 2.7 | - | - | 3.3 |
| YBCO/LAO 150nm | | 3.0 | - | - | 3.3 |

^a to be considered ± 0.01 Å due to instrumental error
^b value measured on the same sample pre and post irradiation
^c ± 0.5 K due to sample reproducibility
^d ± 0.1 K due to instrumental error
^e ± 0.1 due to sample reproducibility

V. CONCLUSION

This paper presented the first study on irradiation of REBCO films and tapes with D-T fusion neutrons. The experiment was carried out at the FNG facility on laboratory-scale YBCO samples and commercial REBCO tapes. This work represents a first step toward filling the gap of experimental approaches available to study neutron radiation damage expected in HTS at relevant fusion conditions, by considering also the high energy portion of the fusion spectrum. The characterization of samples irradiated with 14.1 MeV neutrons showed that, despite the large expected size of defects cascades, the small dpa values introduced at the reachable fluences results in negligible structural and electromagnetic effects.

In the future, higher irradiation fluences will be considered as well as the possibility of monitoring the superconducting properties during the irradiation tests performed at cryogenic temperatures.

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