Radioactivity of Nb₃Sn/Cu Wires After Proton Irradiation

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Abstract

Superconducting magnets based on Nb₃Sn technology are being developed for the LHC high luminosity upgrade. In this context irradiation induced changes of the superconducting properties of state-of-the-art Ti and Ta doped Nb₃Sn wires are presently studied. During irradiation tests with protons of 65 MeV at the cyclotron of Université Catholique de Louvain (UCL), 1.4 GeV at the CERN ISOLDE facility, and 24 GeV at the CERN IRRAD1 facility, the superconductor samples become radioactive and their handling must follow the legal specifications for radioactive materials. We have estimated the activation and the resulting ambient dose equivalent rate up to a fluence of 1017 p/cm² with the Monte-Carlo Code FLUKA. The estimates were verified with experimental activation spectra. Samples for magnetization measurements with a mass of approximately 20 mg have to be considered as radioactive, but they do not have the potential to harm the operators handling them at radiation levels below 1 µSv/h. Larger samples (longer wires and sample holder materials like Cu and TiAl₆V₄) show correspondingly higher ambient dose equivalent rates and activation levels and they must be handled by radiation workers in designated areas, taking into account the standard precautions for work with radioactive materials. The use of radiation shielding during handling is recommended.

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Abstract—Superconducting magnets based on Nb₃Sn technology are being developed for the LHC high luminosity upgrade. In this context irradiation induced changes of the superconducting properties of state-of-the-art Ti and Ta doped Nb₃Sn wires are presently studied. During irradiation tests with protons of 65 MeV at the cyclotron of Université Catholique de Louvain (UCL), 1.4 GeV at the CERN ISOLDE facility, and 24 GeV at the CERN IRRAD1 facility, the superconductor samples become radioactive and their handling must follow the legal specifications for radioactive materials. We have estimated the activation and the resulting ambient dose equivalent rate up to a fluence of 10^{17} p/cm² with the Monte-Carlo Code FLUKA. The estimates were verified with experimental activation spectra. Samples for magnetization measurements with a mass of approximately 20 mg have to be considered as radioactive, but they do not have the potential to harm the operators handling them at radiation levels below 1 μ Sv/h. Larger samples (longer wires and sample holder materials like Cu and TiAl₆V₄) show correspondingly higher ambient dose equivalent rates and activation levels and they must be handled by radiation workers in designated areas, taking into account the standard precautions for work with radioactive materials. The use of radiation shielding during handling is recommended.

Index Terms—superconducting wires and filaments, Nb₃Sn

I. INTRODUCTION

FOR the LHC high luminosity upgrade [1] it is envisaged to use superconducting quadrupole magnets based on Nb₃Sn superconductors. The magnets will be exposed to various high energy irradiation sources, e.g. neutrons, protons and pions, which have the potential to alter the superconducting properties of the Nb₃Sn conductors [2],[3].

A neutron irradiation study of Nb₃Sn superconductors [4] has started in collaboration with the Atominstutut of the Vienna University of Technology. The neutron study is complemented by proton irradiations of up to 1×10^{17} p/cm² at ambient temperature. The conditions of proton irradiation and safe handling of the samples are described in this paper.

For both RRP and PIT Nb₃Sn wires, a scaling can be established between magnetization and transport critical current measurements [5]. The possibility to characterize small wire samples (typical length: 5 mm) by means of

magnetization measurements, rather than by studying much longer wires for transport critical current (I_c) measurements has strongly simplified the proton irradiation studies for the following reasons.

- 1) Higher proton fluences are possible because the proton beam can be focused to a smaller cross section;
- 2) in smaller samples a more homogeneous proton intensity distribution across the sample can be achieved;
- 3) magnetisation measurements are not sensitive to possible crack formation during the handling of the brittle Nb₃Sn;
- 4) after identical proton fluences the dose rates of the magnetization samples with a mass of typically 20 mg are orders of magnitude lower than the dose rates of I_c transport samples, which simplifies radiation protection for handling and storage of the samples.

Here we report on the proton irradiation procedures and the irradiation induced sample activation. Simulations have been performed with the Monte-Carlo radiation transport code FLUKA in order to estimate the dose rate as a function of decay time after proton irradiation. The estimates were verified with dose rate measurements and experimental activation spectra.

II. THE SAMPLES

Nb₃Sn/Cu superconducting strands with an approximate length of 5 mm and a mass of roughly 20 mg are used for magnetisation measurements. Two wires of the Restacked Rod Process (RRP) type produced by Oxford Superconducting Technology [6] (Ta alloyed, billet #7419, and Ti alloyed, billet #11976) and one of the Powder-in-Tube (PIT) type produced by Bruker-EAS [7] (Ta alloyed, billet #0904) have been used for proton irradiation studies. The cross sections of the Ti-alloyed RRP wire and the PIT wire are shown in Fig.1.

Fig. 1. Cross sections of the PIT # 0904 (a) and RRP #11976 (b) wires used for proton irradiation studies.

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The diameters before and after reaction heat treatment (HT) and the densities after HT of the three samples are reported in Table I.

TABLE I. CHARACTERIZATION OF THE IRRADIATED WIRES

	Ø before HT (mm ²)	Ø after HT (mm ²)	ρ after HT (g/cm ³)
RRP #7419	0.507	0.532	8.46
RRP #11976	0.506	0.524	8.33
PIT #0904	0.792	0.825	8.36

III. IRRADIATIONS AND DOSIMETRY

A. 65 MeV proton irradiation at the Cyclotron of Université catholique de Louvain (UCL)

Proton irradiations in the targeted fluence range from 2×10^{16} p/cm² to 10×10^{16} p/cm² with a proton energy of 65 MeV have been performed at the Cyclotron of UCL. A photograph of the proton irradiation beam line at the cyclotron is shown in Fig. 2.

Fig. 2. Proton irradiation beam line at the cyclotron of UCL.

Twenty wire samples were placed in an Al sample holder, in such a way that the samples were in tight contact with the Al plate. The Al sample holder was positioned in a water-cooled Cu holder (see Fig. 3).

Fig. 3. Aluminum sample holder with 20 magnetisation samples with collimator and beam stop.

The beam-dump placed behind the samples and the sample holder was used as Faraday cup for monitoring the proton current. The surface of the beam is delimited by a collimator

placed in front of the sample holder. The proton beam intensity homogeneity inside this collimator with 3.4 cm diameter has been measured with alanine dosimeters [8], which were placed on the water cooled Cu plate that is used as a beam stop (see Fig. 4).

Fig. 4. Alanine dosimeters placed on the Copper plate used as beam stop.

After irradiation the proton dose received by the dosimeters was read with Bruker e-scan Alanine Dosimeter Reader, the estimated error being 2%. The proton intensity inside the collimator area in the horizontal and vertical directions varies less than 20% from the peak intensity.

The proton fluence is calculated from the proton current measured at the beam dump. During sample irradiation the proton current was around 3 μA, which corresponds to a proton flux density of 7.6×10^{15} p/cm² per hour. The integrated proton current to achieve one irradiation step of 2×10^{16} p/cm² is 8.05 μA·h. Thus, the irradiation duration for one step of 2×10^{16} p/cm² was about 2.8 hours, and the total irradiation duration to achieve a fluence of 10^{17} p/cm² was about 14 hours.

The energy loss of the 65 MeV protons in the sample material was calculated with the SRIM program [9] as approximately 6 MeVcm²/g. With a linear density of 0.84 g/cm of the 1 mm diameter samples a maximum energy loss of about 5 MeV of the initial 65 MeV protons is calculated. The estimated radiation induced temperature increase of the samples in tight contact of the Al sample holder is less than 10 °C.

B. 24 GeV proton irradiation at the CERN IRRAD1 facility

Proton irradiations in the targeted fluence range from 1 to 10×10^{16} p/cm² with a proton energy of 24 GeV have been performed at the CERN IRRAD1 facility [10]. The proton flux density at IRRAD1 reaches up to 3×10^{13} p/cm² per hour, i.e. more than two orders of magnitude lower than the fluence that can be reached with the UCL set-up. Fluences of up to 10×10^{16} p/cm² have been accumulated during about 2 years of IRRAD1 operation.

Fig. 5. (a) Nb₃Sn samples packaged in paperboard sample holder. (b) Sample holders on the shuttle in IRRAD 1.

The samples are packaged in paperboard sample holders and fixed with adhesive Kapton tape (see Fig. 5(a)). The beam homogeneity within the 5 mm² active area of the sample holder is 97%. The samples in the paperboard sample holders are placed on the IRRAD1 shuttle (Fig. 5(b)) and transported remotely into the irradiation zone.

The accumulated proton dose is determined by measuring the activation of aluminum witness samples that are irradiated simultaneously with the superconductor samples. The accuracy of these fluence measurements is $\pm 7\%$ [11]. It is assumed that due to the comparatively low proton fluence proton irradiation induced sample heating can be neglected.

C. 1.4 GeV proton irradiation at the CERN ISOLDE facility

In order to achieve proton fluences above 10^{17} p/cm² within a reasonable duration, proton irradiations have been performed with the CERN booster beam on an ISOLDE liquid metal target. Irradiation has been performed in parasitic mode, by placing eight #7419 Nb₃Sn samples inside a polyethylene sample holder (see Fig. 6), which then has been mounted onto an ISOLDE target.

Fig. 6. Polyethylene sample holder with eight samples for irradiation on an ISOLDE target.

The 1.4 GeV proton beam spot has an area of approximately 1 cm². The samples will be exposed to different fluences. In the beam center relatively high proton fluences in the order of 10^{18} p/cm² can be achieved within about one week. The accumulated proton fluence will be determined for each sample by measuring the activation of high purity Al wire witness samples that have been placed behind each of the Nb₃Sn wires [11].

Because of the comparatively high proton fluences and the lack of good thermal contact between the samples and the sample holder, in this case a significant sample heating is expected. This will be estimated for each sample when the exact fluences will have been determined from the Al witness samples activation.

IV. FLUKA SIMULATIONS

The activation and the ambient dose equivalent rate measured in a distance of 10 cm ($H^*(10)$) of the Nb₃Sn/Cu RRP strand samples #7419 (Ta-alloyed) and #11967 (Ti-alloyed) with a mass of 21 mg have been estimated with the radiation transport program FLUKA 2008. The result of the estimate for 3 proton energies (35 MeV, 1.4 GeV and 24 GeV) as a function of decay time after an accumulated fluence of 10^{15} p/cm² are shown in Fig. 7 for the Ta alloyed RRP wire. An approximate elemental wire composition of 0.59 vol.% Cu, 0.28 vol.% Nb, 0.13 vol.% Sn and either 0.09 vol.% Ta or 0.02 vol.% Ti has been assumed.

After identical irradiation conditions the activation and $H^*(10)$ of the Ti and Ta alloyed wire differ only slightly. For the estimates it is assumed that the Nb₃Sn/Cu samples are

irradiated free-in-air with 10^{15} p/cm² during 1 week (similar to the IRRAD1 conditions).

The irradiation conditions in the simulation calculation deviate insofar slightly from those in the UCL and ISOLDE set-up because secondary particles backscattered from the target will add to the primary protons. Since this effect is not assumed to give rise to significant increases of activity or dose rate it has been neglected here.

Fig. 7. Calculated decay time dependence of the ambient dose equivalent rate at 10 cm distance ($H^*(10)$) of Nb₃Sn/Cu RRP type wire #7419 with a mass=21 mg, after irradiation to a maximum fluence of 10^{15} p/cm² accumulated during 5 days.

One hundred days after proton irradiation to 10^{17} p/cm² the simulated $H^*(10)$ values are in the order of 1 μSv/h. 35 MeV proton irradiation leads to significantly lower Nb₃Sn wire activation as compared to 1.4 GeV and 24 GeV irradiation. No significant difference is observed in the activation of Ti and Ta alloyed wires (unlike after neutron irradiation, where Ta alloying causes a much stronger sample activation).

In later studies it is planned to compare magnetization and transport I_c measurements results of Nb₃Sn samples that will be irradiated with 35 MeV protons. In order to estimate the activation and the ambient dose equivalent rate $H^*(10)$ of such sample holders, $H^*(10)$ of bulk Nb₃Sn, pure Cu and the commonly used sample holder material TiAl6V4 have been estimated as well. The FLUKA simulation results are shown in Figs. 8-10. The activation depends on the thickness of the material, and has been calculated for thicknesses ranging from 0.5 mm to 4 mm.

Fig. 8. Calculated ambient dose equivalent rate at 10 cm distance per g of bulk Nb₃Sn irradiated with 35 MeV protons to a maximum fluence of 10^{15} p/cm² accumulated during 5 days, as a function of decay time for 3 different sample thicknesses.

Fig. 9. Calculated ambient dose equivalent rate at 10 cm distance per g of pure copper irradiated with 35 MeV protons to a maximum fluence of 10^{15} p/cm² accumulated during 5 days, as a function of decay time for 3 different sample thicknesses.

Fig. 10. Calculated ambient dose equivalent rate at 10 cm distance per g of TiAlV irradiated with 35 MeV protons to a fluence of 10^{15} p/cm² accumulated during 5 days, as a function of decay time for 4 different sample thicknesses.

V. GAMMA-RAY SPECTROMETRY AND DOSE RATE MEASUREMENTS

Gamma-ray spectrometry of the #7419 and #0904 samples after an accumulated 65 MeV proton fluence of 6×10^{16} p/cm² and of 8×10^{16} p/cm² has been performed at CERN with a Canberra XiRa co-axial high purity Ge detector. In Table II the activities measured for the #7419 wire nine months after the end of irradiation to a fluence of 8×10^{16} p/cm² are compared with the simulation result. The mass of the two magnetisation samples that have been measured together is about 35 mg. The strongest gamma emitter in the Nb₃Sn wires is Co-57 ($T_{1/2}=271$ days). Similar results have been found for the #0902 wires.

TABLE II. ACTIVITIES OF MAIN GAMMA EMITTING ISOTOPES IN THE #7419 WIRE WITH $M=35$ MG MEASURED 9 MONTHS AFTER THE END OF 65 MEV PROTON IRRADIATION TO 8×10^{16} p/cm².

Isotopes	Half live $T_{1/2}$ (days)	Measured activity (Bq)	Calculated activity (Bq)	Calculated /Measured
Co-57	271	10500	12500	1.2
Co-58	71	3670	5080	1.4
Y-88	107	3460	3940	1.1
Zr-88	83	2320	1580	0.7
Mn-54	312	1110	1310	1.2
Co-56	77	982	2280	2.3
Zn-65	244	945	1140	1.2
Co-60	1924	725	880	1.2
Hf-175	70	235	880	3.7

The calculated dose rates have been compared with the $H^*(10)$ values that have been measured after 65 MeV and 24 GeV proton irradiation. $H^*(10)$ of about 1 μSv/h has been measured 1 month after the end of 65 MeV irradiation of the #7419 wire with a weight of 20 mg to 10×10^{16} p/cm². About 9 months after the end of 24 GeV proton irradiation to 10×10^{16} p/cm² $H^*(10)$ of about 0.5 μSv/h has been measured for the #7419 and #0902 wires with an approximate mass of 20 mg. Since the main gamma emitters in the Nb₃Sn wire have a half live in the order of 100 days, the measured dose rates can be compared to those shown in Fig.7, by multiplying the simulation results for 10^{15} p/cm² by a factor of 100.

VI. DISCUSSION AND CONCLUSION

The activation of Nb₃Sn wires induced by irradiation with protons of different energy has been simulated, and the simulation results have been validated by comparison with dose rate measurements and experimental activation spectra of Nb₃Sn samples irradiated with up to 10^{17} p/cm².

The magnetization measurements of proton irradiated Nb₃Sn samples are scheduled about one year after the end of irradiation. The calculated and measured ambient dose equivalent rates show that 1 year after irradiation to 10^{17} p/cm² $H^*(10)$ values do not exceed 1 μSv/h.

Unlike in neutron irradiation where the 182-Ta isotope with a half live of 116 d is formed from Ta, the type of the doping elements (Ti or Ta) has no significant influence on the proton induced Nb₃Sn wire activation.

The technique of measuring the magnetization of small samples ($m = 20$ mg), showing negligible activation and dose rate after a suitable waiting time, enables studies of radiation effects on superconductors in nearly standard laboratory environments with easy applicable measures to avoid disseminating the samples in the environment. In particular, the personnel is not exposed to doses of ionizing radiation of any concern.

In contrast, $H^*(10)$ values exceeding 100 μSv/h are expected after irradiation of I_c transport current measurement samples with a mass of several grams to fluences of interest. Transport, storage and handling of such samples would have to be authorized under the local regulations for radioactive materials, and the personnel would likely fall under the legislation for radiation workers.

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