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RADIATION-INDUCED EFFECTS ON LHC COLLIMATOR MATERIALS UNDER EXTREME BEAM CONDITIONS*

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Abstract

Over the last years, several samples of present and novel LHC collimator materials were irradiated under various beam conditions (using protons, fast neutrons, light and heavy ions at different energies and fluences) in different facilities around the world. This was achieved through an international collaboration including many companies and laboratories over the world. The main goal of the beam tests and the post-irradiation campaign is the definition of a threshold for radiation damage above which LHC collimators need to be replaced. In this paper, highlights of the measurements performed will be presented. First conclusions from the available data are also discussed.

INTRODUCTION

The quest for understanding the behaviour of beam intercepting devices, such as collimators, motivates a rich material R&D in the accelerator beam community. Collimators are the components most exposed to beam losses and high radiation doses. Radiation-induced damage may change the physical-mechanical properties of collimators and therefore their performance in operation. Accelerators with ever increasing beam intensity, brightness and stored energies as for the CERN projects LIU [1] and HL-LHC [2], and other projects such as FAIR [3] or future neutrino facilities, pushes material requirements for collimators into more challenging grounds and unknown territories.

In the framework of the upgrades foreseen for the LHC, very good dimensional stability, resilience to structural degradation and stability of electrical properties are required. The materials presently used for collimators, as carbon fiber-reinforced carbon (CFC) [4], already operated at their physical limits and operation of the LHC has to be adapted to the maximum beam load the presently installed materials allow. A new generation of collimators based on advanced composite materials is needed to fulfil new challenging requirements [5]. Molybdenum-Graphite (MoGr) is a promising candidate [6, 7] and joint efforts between CERN and other international laboratories, such as BNL and GSI, made possible the evaluation of the radiation response against beam impact, for both protons and ions. Composition and properties of MoGr grades involved in irradiation experiments discussed in the paper are compared in Table 1. Note that the materials are orthotropic: the values listed are referred to the most favourable orientation.

The paper collects the most relevant outcomes from irradiation campaigns performed on MoGr over the last few years, with a focus on the results that provided feedback on the material production optimization.

HIGH-ENERGY PROTON IRRADIATION AT BNL

The first irradiation exposure of MoGr (grade MG-1110E) was performed in the 2013-2014 run using the 160 MeV proton beams of the BNL Linac and the BLIP target beamline. The long-term irradiation covered about 8 weeks with a final accumulated fluence of $\sim 10^{21}$ n/cm^2: most of the samples experienced serious degradation and structural collapse (Fig. 1) and therefore it was not possible to perform a complete post-irradiation characterization of thermo-mechanical and electrical properties. The level of 0.3 DPA reached during the irradiation is two orders of magnitude above the damage estimates for LHC primary collimators operating with protons at 7 TeV [8]. Therefore, a new test has been recently launched to evaluate the threshold to damage for MoGr with respect to graphitic composites [9].

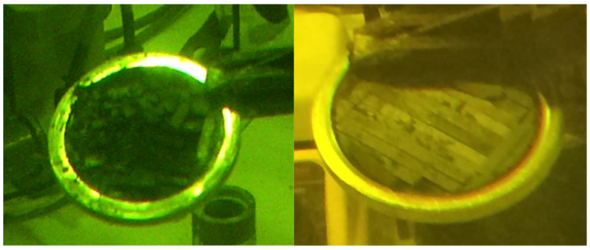

Figure 1: Observed structural degradation in MG-1110E samples after irradiation with 1.1×10^{21} p/cm^2 by 160 MeV protons (left) and exposure to 2.8×10^{18} p/cm^2 at 160 MeV followed by fast neutron fluence of 3.2×10^{18} n/cm^2 (right).

However, results of irradiation with spallation neutrons at BLIP generated by 118 MeV protons are available. In this case, the MoGr samples experienced the effects of short-term proton irradiation (2.8×10^{18} p/cm^2) and the effects of the neutron spectrum (3.2×10^{18} n/cm^2).

Fig. 2 shows load-deflection results of MG-1110E. After irradiation, the slope in the linear trend shows a stiffer behaviour, which is related to the increase of the effective Young's modulus by 20%. The absence of a "continuous" net of carbon fibers in the compound leads to the cross section

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Table 1: Composition and Thermo-Physical-Mechanical Properties (at RT= 20 °C) of MoGr Grades. Coefficient of Thermal Expansion (CTE) is averaged between RT and 200 °C. Abbreviations: Mo = Molybdenum powder, C = graphite flakes, cy = 300μm carbon fibers (from Cytec©), gra = 3 mm carbon fibers (from Granoc©), Ti = Titanium.

MoGR grade	Composition [%vol]	Density [g/cm^3]	Elec. cond. [MS/m]	Th. cond. [W/m K]	CTE [$10^{-6}K^{-1}$]	Flex strength [MPa]
MG-1110E	20 Mo, 40 C, 20 cy, 20 gra	3.76	1	320	6.8	74.4
MG-3110P	20 Mo, 40 C, 20 cy, 20 gra	2.59	1.13	770	1.60	85.0
MG-5220S	7.2 Mo, 46.4 C, 23.2 cy, 23.2 gra	2.63	0.85	677	2.78	72.7
MG-6400U	4.5 Mo, 95.5 C	2.48	0.86	549	1.62	62.9
MG-6530Aa	4.5 Mo, 90.5 C, 5 gra	2.51	0.90	450	1.52	70.9
MG-6541Aa	4.3 Mo, 90.9 C, 4.75 cy, 0.05 Ti	2.49	1.1	507	2.31	79.5

of the bar type specimen (4 mm × 4 mm) to exhibit brittle fracture. This result was confirmed by ultrasonic tests: low dose irradiation with 160 MeV protons (~ 10^{18} p/cm^2) revealed an increase in the ultrasonic velocity (~30% along the "weak" direction and by ~20% along the "strong" direction). It is clearly related with the change of the elastic constant induced by radiation as shown in Fig. 2.

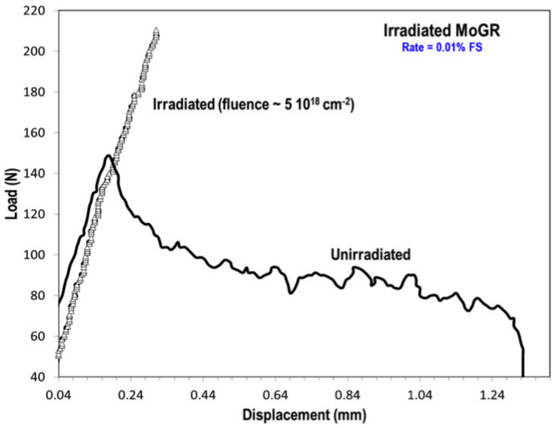

Figure 2: Four-point-bending test comparison of irradiated and unirradiated MG-1110E.

Monochromatic 70 keV X-ray beam and the "white" 200 keV beam at the X17A and X17B1 beamline (respectively) of NSLS were used to assess the microstructural evolution of the MoGr with irradiation. The comparison of EDXRD spectra with "white" beam in Fig. 3 confirms, already in the pristine materials, the high level of graphitization of MoGr with respect to commercial graphite and carbon-carbon composites: the graphite and the fibers in MoGR are well crystallized (narrower 002 diffraction peak) and better than the counterparts shown. However, as shown in the snapshot of the X-ray spectrum for MoGr only in Fig. 4, the shift towards lower diffraction angles of the peak related to the (004) plane of graphite, is correlated to a slight increase of interplanar distance induced by radiation.

At this stage, the sintering cycle was not fully optimized and MG-1110E samples suffered of large non-homogeneity, which may also have affected the characterization. As a follow up of the irradiation campaign at BNL, the production of the MoGr was improved and a higher sintering temperature was reached.

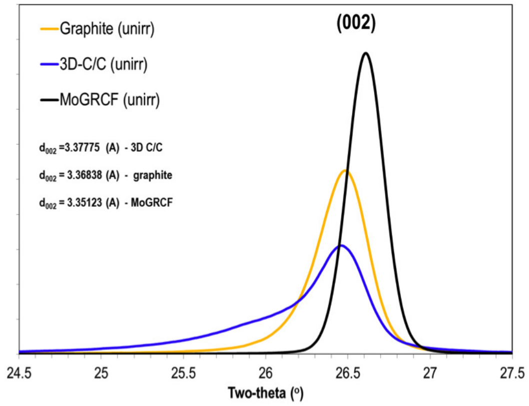

Figure 3: EDXRD on MG-1110E: (002) reflection comparison with isotropic graphite (IG-430) and 3D C/C composite.

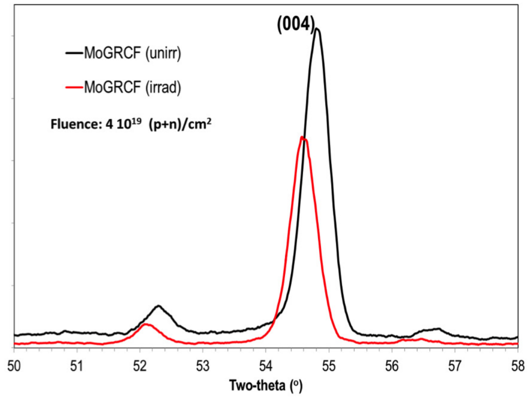

Figure 4: Irradiation-induced changes in the (004) diffraction plane of MG-1110E.

EXPERIENCE OF ION IRRADIATION AT GSI

When swift heavy ions penetrate the matter, a large amount of energy is transferred to the matter through electronic excitation and electron-phonon coupling leading to local damage along the ion trajectory. Irradiation tests were performed at the UNILAC facility at GSI where MoGr samples were exposes to ion beams, with fluence ranging from 1×10^{11} up to 5×10^{13} i/cm^2. 4.8 MeV/u Bi-irradiated MG-3110P samples showed deformation at fluence greater than 6×10^{12} i/cm^2. According to the orientation of carbon fibers embedded in the MoGr specimens exposed to radiation, it was noted that at higher accumulated doses the samples cut along the transversal direction (where fibers and graphitic planes are perpendicular to the surface of the sample) are deforming more with respect to the "in-plane". Raman spectroscopy and thermal conductivity measurements confirm the disorder of graphitic structure as a result of irradiation [10]. In Fig.5, dynamic indentation studies reveal material hardening with fluence increase.

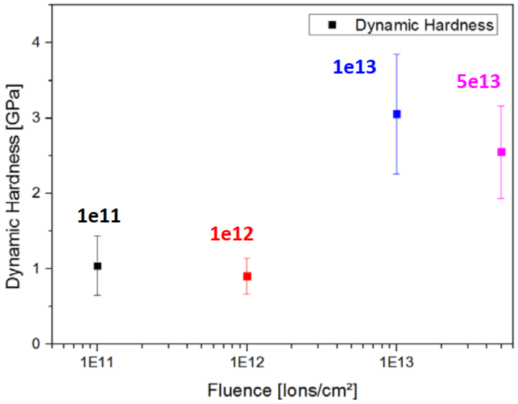

Figure 5: Nano-indentation measurements performed on MG-3110P after Bi ions exposure.

In order to release residual stresses accumulated in the sintering process, which may increase the dislocation density and the grade of distortion of the grains, post-production annealing at 1150 °C or 1300 °C was adopted for MG-5220S. Results after ^{197}Au ion irradiation shown that annealed sample deformation starts at higher fluence (1×10^{13} i/cm^2). However, the accumulation of large doses can heavily affect the property and the microstructure of material.

To get better performance against radiation, additional grades (MG-6400U, MG-6530Aa and MG-6541Aa) followed for which the composition was optimized (see Table 1). Profilometry measurements (Fig. 6) performed after irradiation with 4.8 MeV/u Au ions proved that damage is continuously increasing with fluence for all the grades, with a maximum stress due to swelling between 1×10^{12} and 1×10^{13} i/cm^2. At 1×10^{13} i/cm^2 stress is released by crack formation mostly in the graphite matrix of MG-6530Aa, MG-6541Aa grade experiences less bending at all fluencies.

Experiments with lighter ions were also performed on these grades to simulate the effect of secondary particles and recoil atoms impact on the collimator jaw. The electrical resistivity was monitored on the samples while exposed to Ca ion beam. The change in resistivity is shown in Fig. 7, averaged over the sample thickness in the range of ion penetration (~ 50 μm). High increase in resistivity was measured for MG-6400U after 4×10^{13} i/cm^2, while grades with embedded carbon fibers appear less affected by resistivity increase. Fibers usually provide high thermal and electrical properties to the material along their axis and within the planes within which they are aligned. However, for MoGr a compromise between tolerable deformation and stress by swelling and beam-induced resistivity degradation has to be found.

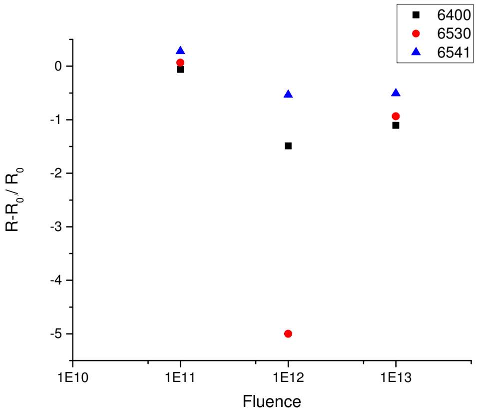

Figure 6: Evolution of beam-induced radius of curvature of various MoGr grades as function of fluence. The samples were irradiated with 4.8 MeV/u Au ion beam.

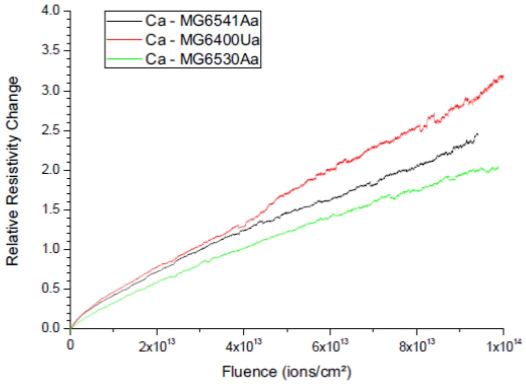

Figure 7: Comparison of relative resistivity changes of various MoGr samples as function of fluence [i/cm^2]. The sample were irradiated with 4.8 MeV/u Ca ions.

CONCLUSIONS AND OUTLOOK

MoGr composite is one of the candidate materials for the new collimators for the HL-LHC upgrade. In order to estimate the lifetime of MoGr collimators in operation the resilience to radiation has been tested with protons, fast neutrons and various ion species. First results provided so far important feedback to the production cycle. Several post-irradiation activities and beam tests are still ongoing in order to get a more complete understanding of how the radiation environment may affect thermo-physical and mechanical properties of graphitic materials. Beyond the relevance for high-energy physics, it is of interest for all applications where equipment may be exposed to high intensity radiation, high-density energy deposition and large thermal loads such as thermal management for electronics, high temperature space applications, fusion and fission reactors.

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