UNDERSTANDING ION INDUCED RADIATION DAMAGE IN TARGET MATERIALS*

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Abstract

Successful operation of next generations of radioactive beam facilities depends on the target survival in conditions of intense radiation field and thermo-mechanical solicitations induced by the driving ion beam. Material property degradation due to ion- beam induced damage will limit target lifetime, either by affecting target performance or, by reducing the material resilience. Similar problems are faced by beam protection elements at LHC. Understanding the mechanism of radiation damage induced by ion beam in these materials provides valuable knowledge for lifetime prediction and for the efforts to mitigate performance degradation. On their way through the target material, energetic heavy ions induce a trail of ionizations and excitations, resulting in formation of ion tracks consisting of complex defect structures. This work reports on the ion-induced structural and thermo-mechanical property degradation studies in high power target materials.

ION- INDUCED RADIATION DAMAGE IN TARGET MATERIALS

The development of high-power heavy ions accelerators poses new challenges to materials that have traditionally served in the nuclear field. Targets, key accelerators components have to perform in severe radiation conditions, experiencing dimensional and structural changes, stresses and severe degradation of properties that control thermal-shock and fatigue resistance. Carbon remains the best choice for high temperature and high dose applications due to the low energy deposited by the primary heavy ion beam, scaling with the atomic number of the target material.

Fine-grained isotropic graphite and carbon-carbon composites have been selected for manufacturing the production target and beam protection elements at the planned Super-FRS fragment separator [1] at FAIR, at FRIB, at LHC and at neutrino facilities. Failure criteria due to irreversible ion-beam induced damage are related to dimensional changes, embrittlement and to degradation of thermal conductivity, leading to increased thermal stresses. To get an estimate for the critical doses, both simulations and experiments are needed.

Samples of high-density, fine-grained, isotropic graphite were exposed to ^{197}Au and ^{238}U ions, at the UNILAC linear accelerator at GSI, Darmstadt, at energies close to Bragg peak, for maximum efficiency of cylindrical damage trails formation. [2]. Irradiated carbon materials present evidence of dimensional changes, induced stress and hardening.

Dimensional changes have been investigated using a Dektak 8, Veeco profilometer. Previous studies have shown that this technique allows a relatively simple, non-destructive test of the sensitivity of a given material to ion-induced damage. The samples were polished to optical quality and partially covered during the irradiation using a thick Al mask. The mean height of the step between pristine and irradiated area has been determined by averaging several individual scans.

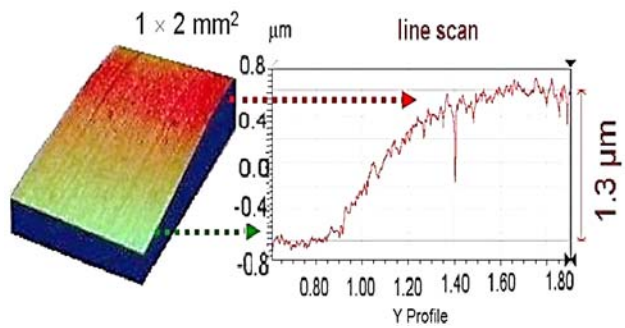

Figure 1: Profilometer mapping at the transition from non-irradiated (front) to irradiated (back) area of a masked polycrystalline graphite sample exposed to 10^{13} ^{238}U ions/cm^2 (11.1 MeV/u). Selected scan displays a step height of 1.3 µm.

Isotropic graphite shows a remarkable resistance to defect production as indicated by the low values of dimensional changes at fluences of up to 10^{12} U ions/cm^2. In the range of 10^{12} to 10^{13} U ions/cm^2 swelling increases steeply. At fluences of 10^{13} U ions/cm^2, ion tracks having a diameter of about 3 nm start to overlap. At this fluence the measured out-of-plane swelling is 1.3 ± 0.1 µm, 1% of the range (120 µm) of the ions (Figure 1). Profilometer mapping of the surface of samples irradiated with 10^{13} U ions/cm^2 reveals crack formation. We also observe a significant bending of this sample due to a strong in-plane stress which develops at the interface between the swollen, irradiated layer and the non-irradiated substrate.

Irradiation induced deformation due to stresses at the interface between irradiated and non-irradiated material has been investigated as a function of fluence. Thin cantilever samples of high density graphite were exposed to GeV heavy ions. The ions are stopped at a depth representing one tenth of the thickness of the sample, inducing strong stresses at this interface and determining the bending of the sample. The radius of curvature was

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measured using a profilometer and the corresponding stress was calculated [3]. Figure 2 presents the evolution of the radius of curvature and of the calculated stress within the irradiated layer, for graphite cantilevers exposed to 4.8 MeV/u, ^{197}Au ions, up to a fluence of 5×10^{13} ions/cm^2.

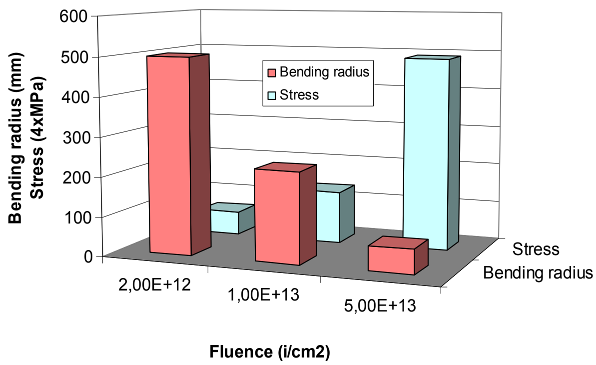

Figure 2: Evolution of radius of curvature and irradiation induced stress with fluence, for narrow graphite cantilever beams exposed to ^{197}Au ions (3.6 Mev/u).

Irradiation induced hardening of graphite samples was investigated using a NanoTest^{TM} Vantage nanoindenter. Hardness values were calculated by averaging experimental data from 10 individual tests. As shown in Figure 3, the irradiated samples exhibit a large increase of the hardness at fluences above 3×10^{13} Au ions/cm^2, indicating this value as a critical dose at which radical irradiation induced structural transformation and stress development take place in graphite.

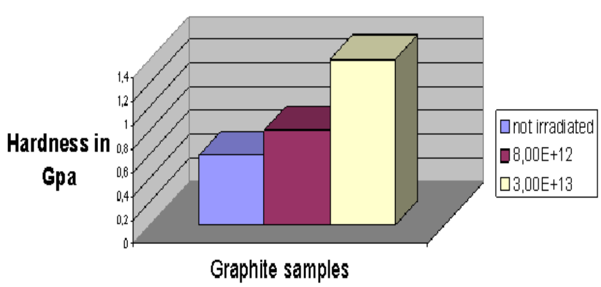

Figure 3: Hardness evolution as a function of fluence for isotropic, fine-grained graphite exposed up to 3×10^{13} ^{197}Au ions/cm^2 (11.1 MeV/u).

Recent experiments, using photo-thermal radiometry techniques showed a pronounced degradation of the thermal conductivity of graphite target materials exposed to heavy ions. High temperature irradiation or post-irradiation annealing brings only partial recovery of the thermal conductivity. [4]

As a result of the radiation induced swelling and stress, the material within the irradiated area of the target will be strongly deformed. An additional contribution to the deformation is probably brought in by creep phenomena, which relax part of the stress by additional deformation. First experiments show that creep might be stronger for high intensity beams, but additional tests are necessary to understand the contribution of creep to target deformation and to distinguish between high temperature and radiation –induced creep. The deformation within the beam spot combined with the stress concentrators at the interface between irradiated and non-irradiated material, hardening and thermal conductivity degradation of the irradiated target area lead to material failure at the edge of the irradiated area. This is a typical failure scenario for targets operating in quasi-continuous irradiation conditions.

At high energies, with low corresponding dE/dx and at high operation temperatures of the target, for which significant defect annealing takes place, we expect that damage is highly reduced. A scaling factor for efficiency of track formation in isotropic graphite, at high energies and target temperatures, based on energy deposition by the beam, can be estimated taking into account the efficiency of track formation determined experimentally from scanning tunneling microscopy studies of Liu at al. on highly oriented pyrolytic graphite (HOPG) [2]. This track yield parameter depends on the energy loss, becoming equal to unity only above an energy loss threshold of 18 KeV/nm. Ion-induced damage is further reduced at temperatures above 800 K, due to track annealing [5]. High temperature irradiation experiments of isotropic graphite have been performed for estimation of the defect recovery at temperatures corresponding to different operating conditions of Super-FRS and FRIB production targets. These experiments show an improved defect recovery at high irradiation temperature [6], helping in delaying the reach of the critical density of tracks and in extending target lifetime.

FAILURE MODES FOR TARGET MATERIALS EXPOSED TO PULSED, INTENSE BEAMS

One of the important challenges for the design of accelerator targets is posed by the regime of short intense pulsed-beam. Super-FRS target at the future FAIR facility and collimators of LHC will work in this regime. Stress waves are generated by the intense fast-extracted primary ion beams which deposit a high amount of energy within a very short time interval. Within such short pulses, the thermal expansion of the heated material is prevented, producing a large compressive stress which propagates through the material with the velocity of sound. These stress waves can reflect as rarefaction waves at the free edges of the target and interfere in a constructive manner. They excite also its natural oscillations, leading to material damage, fatigue and finally to failure. This scenario will be dominant in the fast extraction regime.

Using the quasi-static approach, the pressure rise inside the target scales with the temperature jump induced by

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ion beam energy deposition. For target survival the radial pressure induced by the beam should not be higher than the mechanical strength of the target material. For this regime, the target can mechanically fail due to stress wave solicitations before the temperature rises above melting or sublimation points. For graphite, rarefaction waves are more dangerous than compressive waves because the tensile strength of the graphite material is smaller than the compressive strength. The bending strength of high density graphite is between 45 MPa and 65 MPa. For fast strain rates (shock conditions), the brittle spall strength increases, according to Grady [7], with the strain rate. For high strain rates, the strength of isotropic graphite can double and these values should be used as limits for the lifetime of the target materials.

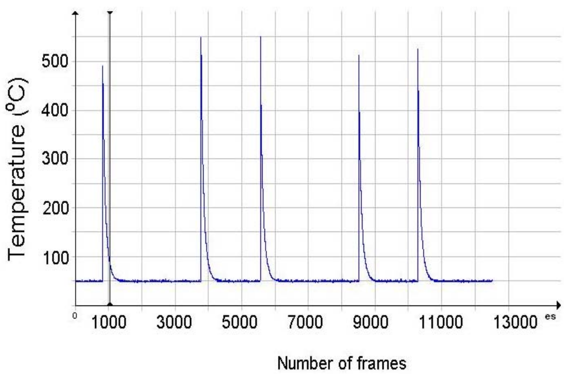

Figure 4: Experimentally determined beam induced cyclic temperature rise in a graphite foil exposed to 1.14 GeV ^{238}U beam with 100 μs pulse length and 0.5 Hz repetition rate.

Pulsed beam operation of target, induces periodic temperature changes in the samples and consequently cyclic thermal stresses in the material. The mechanical integrity of the target will be affected by thermal fatigue effects. Little is known about fatigue behaviour of irradiated graphite, but one expects a decrease of lifetime due to degradation of fatigue resistance in radiation-hardened materials. First test on the fatigue resistance on graphite materials irradiated with pulsed swift heavy ion beams have been done using 1.14 GeV ^{238}U with 100 μs pulse length and 0.5 Hz repetition rate. Cyclic variation of temperature within the beam spot has been measured with very good temporal resolution using a high sensitivity FLIR SC 7500 thermal camera (Figure 4). For this experiments maximum beam- induced temperature jumps within the beam spot reached 600 °C, an amplitude estimated to be reached with enlarged fast extracted beam on the Super-FRS target at FAIR. Fatigue induced target failure after 10^4 pulses are illustrated in Figure 5 for the transmission case, a, and for the situation when the beam is stopped in the target, b. Additional stress at the interface between the irradiated layer and non-irradiated substrate induces a premature failure of the material. Heating the target during irradiation and delaying the degradation of thermal conductivity and the material hardening will improve also the fatigue response of the target.

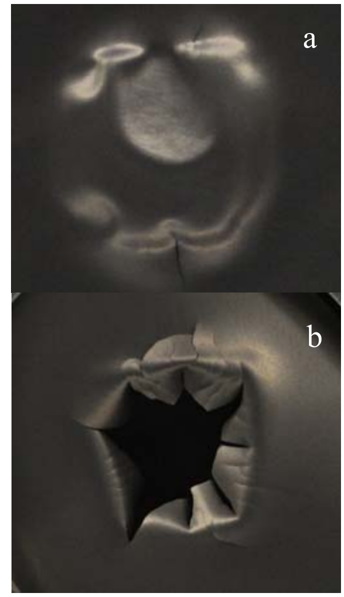

Figure 5: Mechanical failure of thin graphite targets exposed to 1.14 GeV ^{238}U beam with 100 μs pulse length and 0.5 Hz repetition rate:a) beam passes through the foil; b) beam stopped in the foil.

New carbon-based materials, like doped graphite and metal matrix-diamond composite, are currently under investigation at GSI and CERN for improving the response of targets and collimators to fast-extracted proton and ion beams.

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