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ARTICLE

Depth profiling of the residual activity induced in carbon-based materials by heavy ions

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We present new results of the experimental study of the residual activity induced by high-energy heavy ions in carbon-based materials: graphite and carbon composite. The graphite target was irradiated by 500 MeV/u tantalum ions and the carbon composite target was irradiated by 500 MeV/u uranium ions. The targets were assembled from a stack of thin plates and after irradiation were investigated using gamma-ray spectroscopy. Main tasks of the experimental study were: 1) to identify induced radioactive isotopes in the gamma spectra of the measured samples, 2) to estimate residual activity of the identified isotopes and 3) to determine depth profiles of the residual activity of individual isotopes. Depth profiling of the residual activity of all identified isotopes was performed by measurements of individual target plates. According to the depth profiles, the identified isotopes can be classified into two main groups: target-nuclei fragments and projectile fragments. In the measured gamma spectra of the carbon-based materials irradiated by heavy ions only one target-nuclei fragment, ^{7}Be, was identified. All the rest of the isotopes detected using gamma-ray spectroscopy, are the projectile fragments of various masses. The experimental data were compared with Monte Carlo simulations performed by FLUKA code in order to verify validity of physical models and data libraries implemented in the code. A satisfactory agreement between the experiment and the simulations was observed.

Keywords: residual activity; heavy-ion fragmentation; gamma-ray spectroscopy; FLUKA code

1. Introduction

In the frame of the FAIR project (Facility for Antiproton and Ion Research) [1], residual activity induced in various materials by heavy-ion beams are being studied at GSI research centre. The studies include irradiation experiments [2-4] performed on SIS 18 synchrotron and computer simulations using Monte Carlo particle transport codes [5-6]. The experiments are focused on common accelerator construction materials such as copper, stainless steel [2-3] and aluminium [4]. The goals of the experimental studies were to identify the nuclides induced in the irradiated materials, to measure their residual activities and to determine the depth profiles of the residual activities. The experimental data were then used for validation [5-6] of Monte Carlo simulation codes FLUKA [7] and SHIELD [8]. The final goal was to specify tolerable beam losses in high-power heavy ion accelerators [9].

New experimental study of the residual activity induced in carbon-based materials, graphite and carbon-composite AC150, was done within the EU research framework EuCARD, WP 8 (ColMat) [10]. The EU project comprises investigation of materials for beam collimation and machine protection. Graphite and carbon-composite are used as construction materials of collimators and beam dumps in high-power accelerators. The carbon-based materials are chosen due to high stability of the thermo-mechanical properties after irradiation by high energy and high intensity beams [11].

2. Experiment and methods

2.1. Targets and irradiation conditions

The graphite target was irradiated by 500 MeV/u ^{181}Ta beam. The carbon-composite target was irradiated by 500 MeV/u ^{238}U beam. The densities of the graphite and carbon-composite were 1.84 g/cm^3 (measured) and 1.65 g/cm^3 [11], respectively. The graphite target was a cylinder assembled from 30 individual plates. The thickness of each graphite plate was 2.1 mm and the diameter was 12.7 mm (see **Figure 1**). The carbon-composite AC150 target (manufactured in Japan) was a cuboid assembled from 18 individual plates. The cuboid dimensions are presented in **Figure 2**. The thickness of each carbon-composite plate was 3.1 mm.

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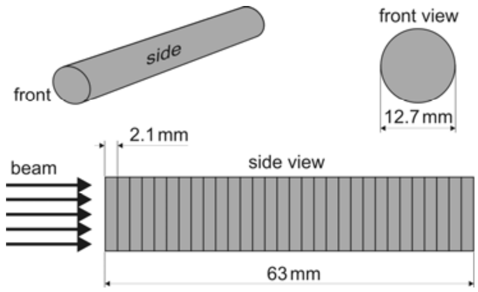

Figure 1. Graphite target irradiated by 500 MeV/u ^{181}Ta ions.

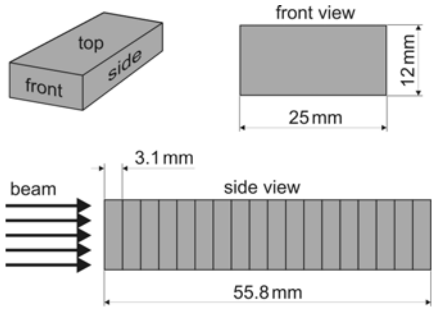

Figure 2. Carbon-composite target irradiated by 500 MeV/u ^{238}U ions.

Beams from the SIS-18 synchrotron at GSI were used to irradiate the targets. The beam profile was approximately Gaussian according to the profile-meter. The beam intensity was monitored by a current transformer. In front of both targets a thin aluminium foil was placed in order to monitor the number of ions impinging on the target. It is done by activity measurement of radioactive isotopes (e.g. ^{7}Be) induced in the part of the foil which covers the target. The thickness of the foil was 0.1 mm and 0.5 mm for graphite and carbon-composite target, respectively. The number of tantalum ions delivered to the graphite target was 2.0×10^{12}. The number of uranium ions delivered to the carbon-composite target was 1.0×10^{13}.

2.2. Gamma spectra measurement

After irradiation of the targets gamma spectra of individual plates were measured repeatedly. The first measurement of the graphite plates started 6 days and lasted until 232 days after the end of irradiation. The first measurement of the carbon-composite plates started 15 days and lasted until 292 days after the end of irradiation. The spectra of the samples were measured using a HPGe detector. The target-to-detector distance was 7 cm for both targets. The spectra were analysed by the GammaVision-32 software package.

2.3. Range of the primary ions

Range of the primary ions was calculated using the FLUKA code [7]. Energy losses of the primary beam in the 100 μm thick vacuum window made of stainless steel, air drift space between the end of the beam pipe and the target (90 cm and 60 cm for the graphite and carbon-composite, respectively) and the aluminium foil, were taken into account. Range of the 500 MeV/u tantalum ions in the graphite target is 23.11 mm. The range of the 500 MeV/u uranium ions in the carbon-composite target is 21.14 mm. The range of the tantalum ions in the graphite target is on the border of the 11th and 12th plate. The range of the uranium ions in the carbon-composite target is at the end of the 7th plate.

3. Experimental results

3.1. Identified isotopes and residual activities

The isotope identification was based on their half-life and on the energy and emission probability of the gamma lines. Activities of the isotopes were obtained from the peak-net-areas calculated using GammaVision-32. The activities were then extrapolated backwards in time to the end of the irradiation (cooling time = 0) and normalized per one incident ion. The combined standard uncertainty of the data shall then comprise two components: (1) uncertainty of the isotope activity and (2) uncertainty of the beam intensity.

As expected from our previous experience [2-6], two types of the identified isotopes could clearly be distinguished: (1) target-nuclei fragments and (2) projectile fragments. The depth profiles of the target-nuclei fragments cover the whole length of the target, upstream as well as downstream of the range of the primary ions. In contrast to that, the depth profiles of projectile fragments start at the range of the primary ions and extend to various depths downstream of the range.

3.2. Depth profiles of the residual activities

In the measured gamma-spectra of the samples only one target-nuclei fragment, ^{7}Be, was identified. Its profile in the graphite and in the carbon-composite target is presented in **Figures 3** and **4**, respectively. The tail beyond the range is caused by the isotopes produced by secondary particles such as neutrons, protons, projectile fragments and decreases modestly with depth. The vertical dashed line in the graphs represents the range of the primary ions calculated using FLUKA code.

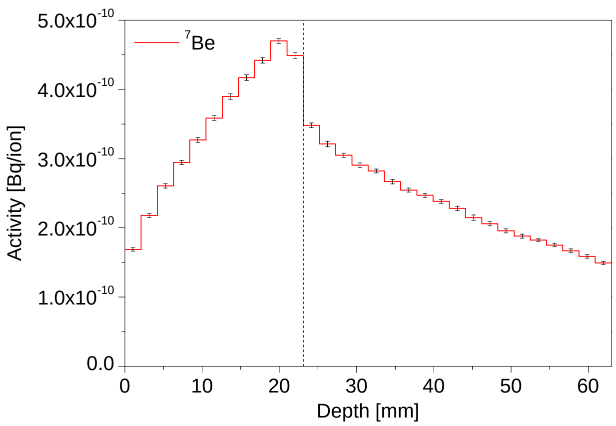

Figure 3. Depth profile of ^{7}Be in the graphite target irradiated by tantalum ions. The activity is calculated at cooling time = 0.

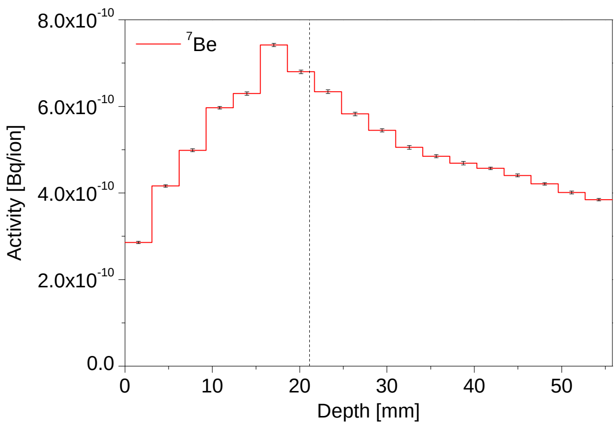

Figure 4. Depth profile of ^{7}Be in the carbon-composite target irradiated by uranium ions. Cooling time = 0.

All the rest of the identified isotopes are the projectile fragments which profiles start at the range area. As an example, the profiles of four fragments of the tantalum primary ion ^{46}Sc, ^{83}Rb, ^{127}Xe and ^{170}Lu, are presented in **Figure 5**. The profiles of four fragments of the uranium primary ion ^{95}Zr, ^{124}Sb, ^{149}Gd and ^{237}U, are presented in **Figure 6**. The dashed line again represents the range of the primary ions calculated using FLUKA.

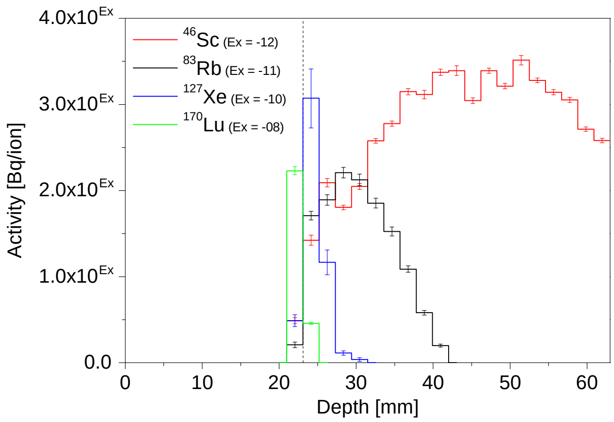

Figure 5. Depth profiles of the residual activity of ^{46}Sc, ^{83}Rb, ^{127}Xe and ^{170}Lu in the graphite target irradiated by tantalum ions. Cooling time = 0.

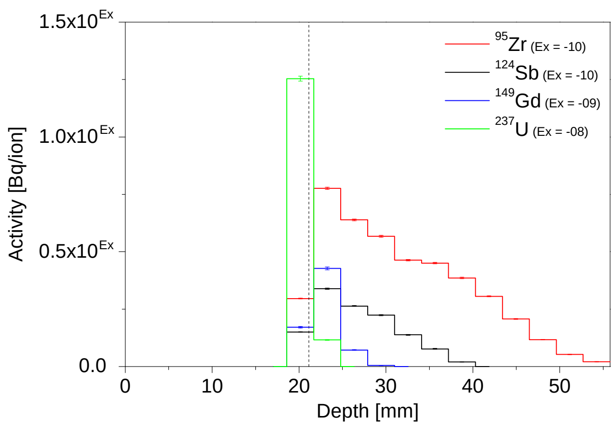

Figure 6. Depth profiles of the residual activity of ^{95}Zr, ^{124}Sb, ^{149}Gd and ^{237}U in the carbon-composite target irradiated by uranium ions. Cooling time = 0.

It can be seen that the light fragments penetrate to the considerably longer depth than the heavy fragments. This is due to the fact that the range of ions at the same initial energy per nucleon is roughly proportional to A/Z^2 (A is the mass number and Z is the proton number of the ion). Heavy fragments with A/Z^2 very close to the original projectile (e.g. ^{237}U) can be used for the range verification of the primary ions [12].

The uncertainty bars in the depth-profile graphs do not include uncertainty of the beam intensity and include only uncertainty of the residual activity. Addition of the beam-intensity uncertainty to the depth profiles could cause distortion of the presented data. It should be included only in the uncertainty of the isotope's total activity induced in the whole target.

4. Monte Carlo simulations using the FLUKA code

Monte Carlo code FLUKA was used to simulate the experiment in order to validate implemented physical models and data libraries. The simulated and measured activities were compared in the same time-points at the beginning of the measurement: 6 days and 15 days after irradiation for the graphite and carbon-composite, respectively. Choosing the comparison time-points following some cooling-time after the end of irradiation takes into account a possible contribution from short-lived isotopes [6]. This is due to the fact that some isotopes of interest may be produced additionally after the end of irradiation being daughter products of the decaying short-lived isotopes. While this contribution can be treated correctly in FLUKA simulations, it is not possible to be taken into account in experimental data.

Comparisons of the measured and simulated depth profiles of the residual activity of ^{7}Be, ^{75}Se and ^{131}Ba induced in graphite and of ^{95}Zr, ^{139}Ce and ^{233}Pa induced in carbon-composite are presented in **Figures 7** and **8**, respectively. Ratio Experiment/FLUKA of the total activities induced in the whole graphite target is presented in **Figure 9**. In case of the total activities the uncertainty of the beam intensity is included in the presented uncertainty bars.

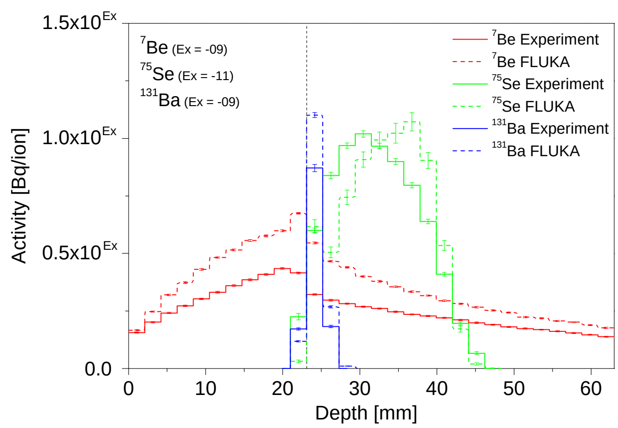

Figure 7. Measured and simulated depth profiles of ^{7}Be, ^{75}Se and ^{131}Ba in the graphite target irradiated by tantalum ions. Cooling time = 6 days.

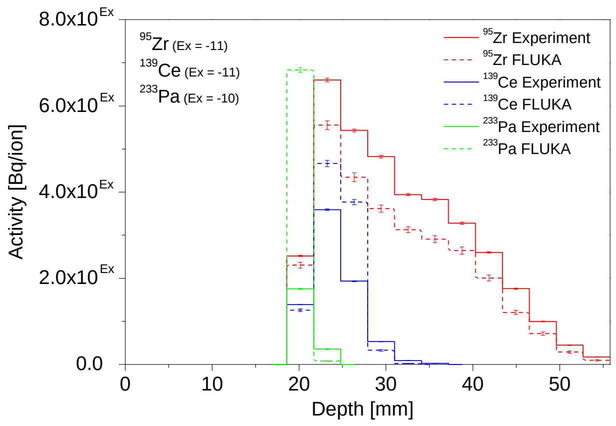

Figure 8. Measured and simulated depth profiles of ^{95}Zr, ^{139}Ce and ^{233}Pa in carbon-composite target irradiated by uranium ions. Cooling time = 15 days.

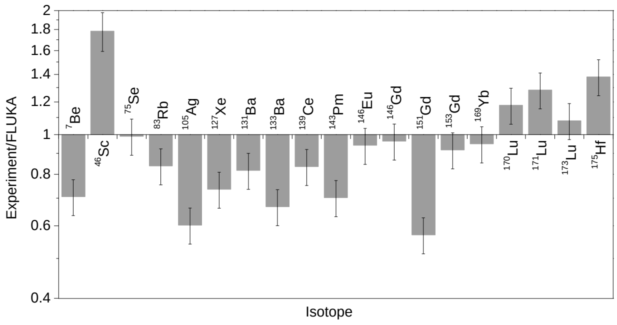

Figure 9. Ratio Experiment/FLUKA for the isotopes identified in the spectra of the whole graphite target irradiated by tantalum ions calculated for the cooling time = 15 days.

It was found out that FLUKA mostly overestimates the residual activities of the induced isotopes. A significant overestimation was observed especially in case of some heavy fragments of the uranium primary ions. This can be seen for example in the comparison of the measured and simulated depth profile of the ^{233}Pa fragment (see Figure 8). Nevertheless, the comparison of the experimental data with the FLUKA simulations can be interpreted as a reasonably good agreement. It allows us to extend the simulated data over the range that can be practically covered by experiments.

5. Conclusion

Residual activities and their depth profiles in graphite and carbon-composite targets irradiated by tantalum and uranium ions were measured using a gamma-ray spectroscopy. The characteristic shape of the depth profiles for target-nuclei fragments and projectile fragments was described. The depth profiles of the target-nuclei fragments cover the whole length of the targets whereas the profiles of the projectile fragments start at the range of the primary ions. The projectile fragments penetrate to various depths which depend on their A/Z^2 ratio. The production of the target-nuclei fragments downstream of the range of the primary ions is caused by secondary particles.

Experimental results were compared with the FLUKA simulations. The comparison showed a reasonably good agreement of the experimental data with the simulated ones.

Acknowledgements

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References

[1] H.H. Gutbrod (Editor in Chief), *FAIR - Baseline Technical Report*, GSI Darmstadt, (2006).

[2] I. Strasik, E. Mustafin, A. Fertman, R. Hinca, M. Pavlovic, D. Schardt, N. Sobolevsky, A. Golubev, B. Sharkov, G. Fehrenbacher, I. Hofmann, H. Iwase, E. Kozlova and G. Mustafina, Experimental study of the residual activity induced by 950 MeV/u uranium ions in stainless steel and copper, *Nucl. Instrum. Methods Phys. Res.* Sect. B 266 (2008), pp. 3443-3452.

[3] I. Strasik, E. Mustafin, T. Seidl and M. Pavlovic, Experimental study and simulation of the residual activity induced by high-energy argon ions in copper, *Nucl. Instrum. Methods Phys. Res.* Sect. B 268 (2010), pp. 573-580.

[4] V. Chetvertkova, I. Strasik, A. Belousov, H. Iwase, N. Mokhov, E. Mustafin, L. Latysheva, M. Pavlovic, U. Ratzinger and N. Sobolevsky, Activation of aluminum by argon: Experimental study and simulations, *Nucl. Instrum. Methods Phys. Res.* Sect. B 269 (2011), pp. 1336-1340.

[5] I. Strasik, E. Kozlova, E. Mustafin, I. Hofmann, A. Smolyakov, N. Sobolevsky, L. Latysheva and M. Pavlovic, Simulation of the residual activity induced by high-energy heavy ions, *Nuclear Technology* 168 (2009), pp. 643-647.

[6] E. Kozlova, I. Strasik, A. Fertman, E. Mustafin, T. Radon, R. Hinca, M. Pavlovic, G. Fehrenbacher, H. Geissel, A. Golubev, H. Iwase and D. Schardt, Benchmark test of the FLUKA Monte Carlo code for residual production with 500 and 950 MeV/u uranium beam on copper and stainless steel targets, *Nuclear Technology* 168 (2009), pp. 747-751.

[7] A. Ferrari, P. R. Sala, A. Fasso and J. Ranft, Reports No. CERN-2005-10, INFN/TC_05/11, and SLAC-R-773.

[8] A.V. Dementyev and N.M. Sobolevsky, SHIELD - universal Monte Carlo hadron transport code: scope and applications, *Radiation Measurements* 30 (1999), pp. 553-557.

[9] I. Strasik, E. Mustafin and M. Pavlovic, Residual activity induced by heavy ions and beam-loss criteria for heavy-ion accelerators, *Phys. Rev. ST Accel. Beams* 13 (2010), 071004.

[10] J. Stadlmann, L. Bozyk, H. Kollmus, P. Spiller, I. Strasik, N.A. Tahir, M. Tomut and C. Trautmann, Collimators and materials for high intensity heavy ion synchrotrons, *Proc. Int. Part. Accel. Conf.*

IPAC'12, New Orleans, Louisiana, USA, May. 20-25, 2012, (2012). pp. 2564-2566.

[11] A. Bertarelli, G. Arnau Izquierdo, F. Carra, A. Dallocchio, M. Gil Costa and N. Mariani, Research and development of novel advanced materials for next-generation collimators, *Proc. Int. Part. Accel. Conf. IPAC'11*, San Sebastian, Spain, Sept. 4-9, 2011, (2011), pp. 2888-2890.

[12] I. Strasik, V. Chetvertkova, E. Mustafin, M. Pavlovic and A. Belousov, Depth profiling of residual activity of ^{237}U fragments as a range verification technique for ^{238}U primary ion beam, *Phys. Rev. ST Accel. Beams* 15 (2012), 071001.