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SIMULATION OF RESIDUAL ACTIVITY IN STEEL AND COPPER TARGETS INDUCED BY 950 MeV/NUCLEON URANIUM IONS

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Radioactive nuclides will be produced in structures and surrounding materials of the relativistic heavy ion collider NICA. Predicting radioactivity in the materials, air, cooling water, and ground is an important part of the technical project. Some Monte Carlo transport codes (FLUKA, GEANT4, SHIELD) can simulate the production of radionuclides induced by heavy ions. The preliminary verification of MC codes with experimental data is necessary before using them for radiation protection purposes. In the present work, comparisons of the experiment and GEANT4 and SHIELD simulations of induced activity in thick stainless steel and copper targets irradiated with 950 MeV/nucleon uranium ions are presented.

Пучки тяжелых релятивистских ядер ускорительного комплекса NICA будут формировать наведенную активность в элементах и окружающих материалах ускорителя. Прогнозирование уровней радиоактивности в материалах конструкций, воздухе, охлаждающей воде и почве является важной частью технического проекта. В настоящее время ряд программ транспорта излучений в веществе на основе метода Монте-Карло (FLUKA, GEANT4, SHIELD) позволяет оценить образование радионуклидов под воздействием тяжелых ионов. Тем не менее для использования данных программ в целях радиационной безопасности необходимо предварительно выполнить сличение результатов расчетов с экспериментальными данными. В данной работе приводятся результаты сравнения с экспериментом расчетов по программам GEANT4 и SHIELD наведенной активности в мишенях из нержавеющей стали и меди, облученных ядрами урана с энергией 950 МэВ/нуклон.

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Radioactivity induced by primary ions and secondary hadrons in the beam line equipment of the NICA accelerator complex at JINR will make the main contribution to the personnel dose during the maintenance work. The sources of the strongest radioactivity will be the beam catchers irradiated with 4.5 GeV/nucleon gold ions within the NICA collider. The prediction of the short- and long-time radioactivity of the beam line elements and shielding structures is an important part of the NICA project. Due to the complexity of the activation mechanism stipulated by the intra- and internuclear cascades in the matter, it is hardly possible to obtain

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reliable radionuclide production data from radiation transport codes using different models of nucleus–nucleus collision and data libraries. Thus, experimental data have to be used as an indicator for the evaluation of various MC transport codes estimating residual radioactivity.

As for the NICA project, the most suitable experimental data on residual radioactivity were obtained in an experiment in which thick stainless steel and copper targets were bombarded with 950 MeV/nucleon ^{238}U ions [1]. Recently, the first benchmark study of activation prediction using the FLUKA code [2, 3] was done based on experimental data [4]. The aim of the present work is to verify experimentally other MC codes: GEANT4 [5, 6] and SHIELD [7, 8].

The target material densities were 8.93 and 7.9 g/cm^3 for copper and stainless steel, respectively [1]. The targets had a cylindrical shape with a diameter of 5 cm and thicknesses of 133.09 cm for copper and 138.38 cm for stainless steel. The targets were assembled from disks with thicknesses varying from 0.1 to 20 mm. After irradiation, the gamma spectra from all disks were measured by an HP(Ge) spectrometer at two cooling times. For the code verification, only few isotopes with the decay time of more than two days, large gamma yield, and easy identification were chosen. The measured isotope activities were then extrapolated in time to the end of target irradiations.

The GEANT4 (version 9.3) Binary Cascade was used to model the intranuclear cascade within a nucleus and isotope production. The real spatial distribution of the narrow ion beam was not taken into account for computer simulations. The composition of stainless steel was chosen according to [4]. For secondary neutrons with energies below 100 MeV, MENDL [9] data libraries are used to evaluate isotope production in the internuclear cascade within the target. For all other reactions, the isotopes are derived from the final state of the transport simulation engines. The G4RadioactiveDecay process is applied to simulate the decays of radioactive isotopes by α, β^+, and β^- emission and by electron capture. The G4RadioactiveDecay process is based on the decay chains and time structure of real experiments. If the daughter of a nuclear decay is an excited isomer, its prompt nuclear deexcitation is treated using the G4PhotoEvaporation process. A rough estimation (equal sharing among states) of isomer production is used in this case. The excited state is assumed to survive if its lifetime is longer than decay time of the isotope in ground state.

The SHIELD code does an exclusive simulation of nuclear reactions in a target (the MSDM generator). For low-energy neutron transport, the LOENT code is based on the 28-group neutron data library. Since SHIELD has no special process to take into consideration the isotope decay chains and isomer transitions, the DCHAIN-SP [10] code is used for the further processing of the calculated isotope production.

The FLUKA code (version 2006.3b) was used [4] for the simulation of experimental data. The residual nuclide activities were building up in several decay chains. FLUKA involves isotope production only in the ground state; therefore, the post-processing of the results was done to take into account some isomers. The results calculated in [4] are presented as experimental-to-simulated data ratios. The FLUKA results were recovered on the basis of the experimental data from [1] and the corresponding ratios [4].

The statistical uncertainties of the FLUKA, GEANT4, and SHIELD simulations were within a few percent.

The comparisons of the FLUKA, GEANT4, and SHIELD simulations are presented in Tables 1 and 2. The rows set in bold correspond predominantly to the residual radionuclides from the target nuclei; the light-faced rows correspond to the radioactive fragments of the

Table 1. **A comparison of the partial and total activities of the residual radionuclides (with $T_{1/2} > 2$ days) in a thick stainless steel target [1] simulated by the FLUKA, GEANT4, and SHIELD codes**

Isotope	Experiment, Bq·ion^{-1}	FLUKA, Bq·ion^{-1}	GEANT4, Bq·ion^{-1}	SHIELD, Bq·ion^{-1}
Be-7	**3.06E–08**	**1.81E–08**	**1.38E–08**	**1.73E–08**
Na-22	**5.40E–10**	**3.75E–10**	**1.02E–10**	**4.57E–10**
Sc-44m	**1.09E–6**	**1.18E–06**	**2.46E–06**	**4.94E–06**
Sc-46	**4.99E–08**	**6.16E–08**	**4.29E–09**	**1.75E–08**
Sc-47	**5.56E–07**	**3.31E–07**	**1.25E–07**	**1.44E–07**
V-48	**5.46E–07**	**4.63E–07**	**3.29E–07**	**7.29E–07**
Cr-51	**1.02E–06**	**8.09E–07**	**5.68E–07**	**1.13E–06**
Mn-52	**8.80E–07**	**1.57E–06**	**9.17E–07**	**3.10E–06**
Mn-54	**7.66E–08**	**6.53E–08**	**1.96E–08**	**5.10E–08**
Co-56	**4.09E–08**	**3.09E–08**	**2.55E–08**	**6.30E–08**
Co-57	**1.62E–08**	**2.03E–08**	**1.75E–08**	**1.53E–08**
Ni-57	**5.24E–07**	**1.05E–06**	**1.35E–06**	**1.38E–08**
Co-58	**5.11E–08**	**7.48E–08**	**3.63E–08**	**2.36E–06**
Zr-95	5.65E–10	2.15E–10	5.81E–10	2.60E–10
Mo-99	1.67E–08	5.02E–09	2.47E–08	1.54E–10
Ru-103	1.24E–09	9.84E–10	1.13E–09	6.11E–09
Te-121	4.41E–10	8.82E–10	3.95E–10	5.64E–10
I-126	4.92E–09	2.90E–09	5.83E–11	2.87E–10
Sb-126	5.78E–10	2.66E–10	2.56E–09	4.46E–10
Xe-127	6.97E–10	4.93E–10	1.12E–10	1.07E–10
I-131	2.81E–09	7.15E–10	2.86E–09	9.52E–10
Ba-131	2.83E–09	1.57E–09	2.21E–10	1.45E–10
Ce-139	7.16E–11	1.21E–10	7.40E–12	4.19E–10
Ce-141	2.06E–10	2.75E–10	5.25E–10	2.49E–11
Gd-146	4.25E–10	1.66E–10	8.44E–11	2.33E–10
Gd-149	9.85E–10	6.12E–10	3.02E–10	8.98E–11
Yb-169	7.11E–10	5.09E–10	1.19E–10	3.53E–10
Bi-206	1.71E–09	3.35E–09	4.73E–09	2.58E–11
U-237	9.03E–08	1.46E–08	2.59E–08	2.57E–10
Total activity	5.01E–6	5.71E–6	5.93E–6	1.26E–5

uranium projectiles. The experimental data are the gamma spectra measured during the period of the 3rd – 23rd day (for a stainless steel target) and the 14th – 38th day (for a copper target) after the end of irradiation. The experimental standard deviations including detector-efficiency calibration errors and net-peak-area errors are dependent on the radionuclide and vary to tens of percent.

It should be emphasized that the contributions of the decay chains and isomers to the total activities are quite large. As the codes consider it differently, a precise comparison of the simulations is difficult. On the other hand, the experimental results were not quite adequate because of the procedure of time extrapolation to the end of irradiation. It was assumed that the selected isotopes underwent a purely exponential decay; i.e., isomers and decay chains leading to the selected isotopes in the ground states were not taken into account. As a result, *Table* 2. **A comparison of the partial and total activities of the residual radionuclides (with $T_{1/2} > 2$ days) in a thick copper target [1] simulated by the FLUKA, GEANT4, and SHIELD codes**

Isotope	Experiment, Bq·ion^{-1}	FLUKA, Bq·ion^{-1}	GEANT4, Bq·ion^{-1}	SHIELD, Bq·ion^{-1}
Be-7	**2.09E–08**	**1.25E–08**	**7.41E–10**	**1.26E–08**
Na-22	**3.51E–10**	**1.01E–10**	**3.25E–10**	**1.35E–10**
Sc-44m	**4.02E–07**	**3.47E–07**	**9.98E–07**	**1.20E–06**
Sc-46	**1.87E–08**	**2.17E–08**	**1.25E–08**	**4.12E–09**
Sc-47	**2.51E–07**	**2.20E–07**	**2.11E–07**	**3.07E–08**
V-48	**1.64E–07**	**1.11E–07**	**3.56E–07**	**1.92E–07**
Cr-51	**2.54E–07**	**1.72E–07**	**4.68E–07**	**2.47E–07**
Mn-52	**4.09E–07**	**4.09E–07**	**1.83E–06**	**1.11E–06**
Mn-54	**2.62E–08**	**1.97E–08**	**3.48E–08**	**7.48E–09**
Co-56	**4.52E–08**	**4.66E–08**	**2.09E–07**	**1.78E–08**
Co-57	**4.64E–08**	**4.46E–08**	**1.14E–07**	**1.28E–07**
Co-58	**2.41E–07**	**3.83E–07**	**3.38E–07**	**4.26E–08**
Fe-59	**2.41E–08**	**1.60E–08**	**6.38E–09**	**1.25E–07**
Co-60	**4.59E–09**	**6.85E–09**	**4.12E–08**	**2.32E–09**
Zn-65	**1.92E–09**	**1.49E–09**	**6.40E–09**	**3.52E–09**
Zr-95	8.17E–10	1.58E–10	6.20E–10	2.48E–10
Mo-99	1.33E–08	4.96E–09	2.11E–08	1.29E–10
Ru-103	8.45E–10	7.02E–10	1.28E–09	6.13E–09
Te-121	3.71E–10	7.27E–10	6.13E–10	5.40E–10
I-126	8.73E–10	1.04E–10	5.16E–10	2.82E–10
Sb-126	3.40E–10	2.68E–10	1.46E–09	3.96E–10
Xe-127	4.63E–10	5.73E–10	1.94E–10	7.47E–11
I-131	2.94E–09	7.72E–10	2.42E–09	9.47E–10
Ba-131	7.26E–10	1.65E–09	7.67E–10	1.40E–10
Ce-139	7.35E–11	1.75E–10	1.05E–10	3.57E–10
Ba-140	2.15E–09	3.32E–10	1.66E–09	5.64E–10
Ce-141	1.14E–10	3.04E–10	6.60E–10	7.46E–10
Eu-146	1.77E–09	2.16E–09	1.98E–09	2.14E–11
Gd-146	3.73E–10	1.71E–10	1.40E–10	2.14E–10
Gd-149	1.34E–09	8.93E–10	9.18E–10	9.35E–11
Yb-169	1.04E–09	4.44E–10	6.88E–10	8.17E–11
Bi-206	6.14E–09	4.12E–09	1.97E–08	3.21E–10
U-237	1.50E–07	1.42E–08	5.03E–08	3.03E–11
Total activity	2.09E–06	1.84E–06	4.73E–06	3.13E–06

the comparison does not claim to be a perfect conclusion; rather, it can be used only for radiation protection needs.

The overwhelming part of the total activities is caused by the target residual nuclides. It means that these nuclides are a result of the hadrons' internuclear cascade within thick targets. The partial activities in Tables 1 and 2 differe very significantly for some isotopes. Though, the total activities simulated by SHIELD and GEANT4 codes are overestimated up to 2.5 times only. It is quite acceptable for the radiation protection purposes as overestimation is preferable to underestimation in this case. Thus, all the three codes — GEANT4, SHIELD, and FLUKA — can be confidently used for the gamma dose rate estimation at a rather short time after the end of irradiation. As concerns the prediction of the residual activity gamma dose rate for a longer time (for example, several months or years), the level of computer simulation reliability is now insufficient ex facte because it is necessary in this case to simulate exactly the long-term partial activities and take into account the big amount of long-lived isotopes with small production rate and low gamma yields.

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