Analysis of Induced-radioactivity using DCHAIN-SP for Iron, Copper and Niobium at a Mercury Target Irradiated by 2.83 and 24 GeV Protons

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Reliability estimation was carried out on a radioactivity calculation code system consisting of PHITS, MCNP/4C and DCHAIN-SP 2001 by analyzing an activation experiment performed by using AGS (Alternative Gradient Synchrotron) accelerator at Brookhaven National Laboratory. For induced radioactivity in iron, copper and niobium samples, calculations and experiments were compared indicating that both agreed by a factor of 2 on the average over produced nuclides although the calculation had a tendency to underestimate.

1 Introduction

Many kinds of radioactive nuclides are produced in materials in high-energy intense proton accelerator facilities such as J-PARC [1] conducted by Japan Atomic Energy Agency (JAEA) and the High Energy Accelerator Research Organization (KEK). For calculations of such radioactivity, a high-energy particle induced radioactivity calculation code DCHAIN-SP was developed [2, 3] to combine with such particle transport codes as NMTC/JAM [4, 5] and MCNP[6]. NMTC/JAM estimates nuclide yields produced in target materials mainly by neutrons with energies higher than 20 MeV and protons. MCNP simulates neutron transport phenomena below 20 MeV to provide neutron flux spectra. DCHAIN-SP combines the nuclide yields calculated by NMTC/JAM and those below 20 MeV by folding the activation cross section data with the flux spectra calculated by MCNP, and estimates time-evolution of decaying nuclides by Beteman equation and a decay data library. Physical quantities of radioactivity, decay heat and decay γ-ray spectrum are obtained.

Reliability estimation for this code system was already carried out focusing on either the activation cross section data library below 20 MeV or the nuclide yields calculated by NMTC/JAM. The former was performed using experimental activation data in a 14-MeV neutron field [7, 8], the latter using experimental cross section data in literatures [8]. Although, the reliability estimation was performed using the experimental data in ideal radiation fields, it is also important to estimate the reliability in rather complicated fields including incident protons and spallation neutrons. We utilized an induced-radioactivity experiment[9] performed in a framework of ASTE (AGS Spallation Target Experiment) collaboration [10]. In the experiment, fourteen kinds of materials were activated around the mercury target irradiated by 2.83 and 24 GeV protons. The radioactivity was measured at cooling times from 2 hours to 200 days. In this paper, the radioactivity in iron, copper and niobium is analyzed in order to estimate the reliability of the code system consisting of DCHAIN-SP 2001, PHITS[11] (an upgrade version of NMTC/JAM) and MCNP/4C.

2 Experiment

Figure 1 shows schematic views of the target and the activation samples. Mercury was contained in a cylindrical target container (ϕ=200 mm, L=1300 mm) having a hemisphere beam incident surface

made of stainless steel with 2.5 mm in thickness. The samples of boron-10, boron-11, carbon, aluminum, iron, copper, niobium, mercury-oxide, lead, bismuth, acrylic resin, SS-316, Inconel-625 and Inconel-718 were activated by proton (2.83 and 24 GeV) injection on the target. Sample stacks were set at the on-beam and off-beam position as shown in Fig. 1. The on-beam position samples were irradiated with the incident protons and the spallation neutrons, and off-beam samples were irradiated mainly with the spallation neutrons. An integrating current transformer (ICT) was utilized to measure the total number of protons injected onto the mercury target. The imaging plate (IP) technique was employed for monitoring the incident proton beam profile. A thin aluminum foil was exposed to the proton beam. After the irradiation and cooling, foil was attached to an IP to obtain the image of the distribution of radioactivity mainly induced by the Al(p,x)^{24}Na reaction. To obtain the numbers of protons bombarded the on-beam samples precisely, we measured ^{24}Na activity in copper foil in the stacks induced by the ^{nat}Cu(p,x) reaction. After the irradiation, γ-rays of activated samples were measured with HPGe detectors at cooling times between 0.1 and 200 days. Details are shown in reference [9].

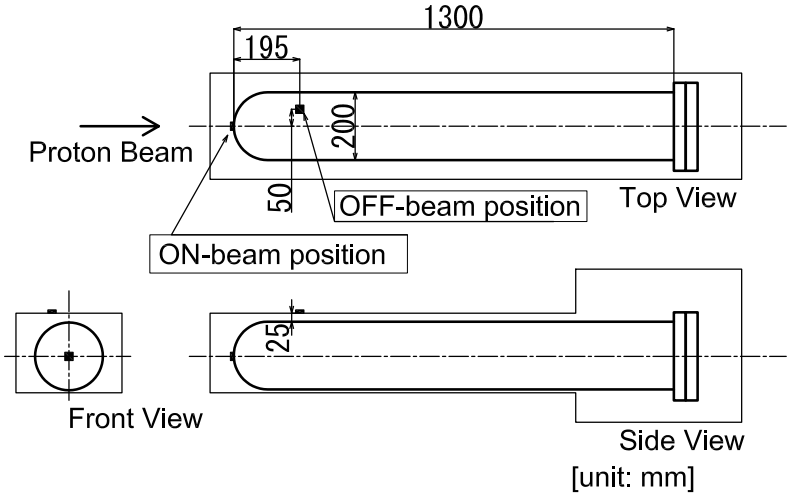

Fig. 1: Side, top and front views of the mercury target. 'On-beam' and 'Off-beam' positions of activation sample stacks are indicated in the top view.

3 Analysis

Figure 2 shows a flow diagram of the radioactivity calculation. At first, full geometry calculations with PHITS were carried out. The target container, the all samples, concrete walls of a irradiation room were included in the calculation model. The proton beam profiles were assumed to exhibit Gaussian distributions judging from the IP measurements. The resultant full widths at half maxima (FWHMs) were (W_x, W_y, E_p)=(40 mm, 19 mm, 2.83 GeV) and (27 mm, 27 mm, 24 GeV), where W_x and W_y were FWHMs in horizontal and vertical directions, E_p the incident proton beam energy. The first PHITS calculations produced proton (>1 MeV) and neutron (>20 MeV) energy spectra at the samples. Using the proton and the neutron energy spectra, nuclear production yields were calculated again by PHITS. Although the nuclide yields are produced in the first PHITS calculation, this two-step calculation method was indispensable to achieve adequate uncertainty associated with the Monte Carlo calculation. Since there are many calculation options to select reaction models in PHITS, default parameters are determined to obtain reasonable results through such benchmark calculations as analyses of neutron spectra produced by the spallation reactions. We utilized the default parameters except for INMED option in the calculation of nuclide yields by PHITS. By default, INMED indicates to use the nucleon-nucleon elastic scattering cross sections in medium [12], while that in free-space was utilized for the present nuclide yields calculations. In DCHAIN-SP, we also adopted non-default parameter for ISOMER, which indicates how to treat isomers in the nuclide yields

file. DCHAIN-SP reads the isomer data as is shown in the files by default, however, the current version of PHITS outputs only nuclides in ground state. To avoid serious underestimation of some important nuclides, we adopted a parameter which assumed that nuclides in the grand state and all isomers were produced with equal probabilities. Assuming the number of isomer is two, underestimation of nuclide production is limited by a factor of 3 although it may cause large overestimation for a nuclide having small production yield in practical.

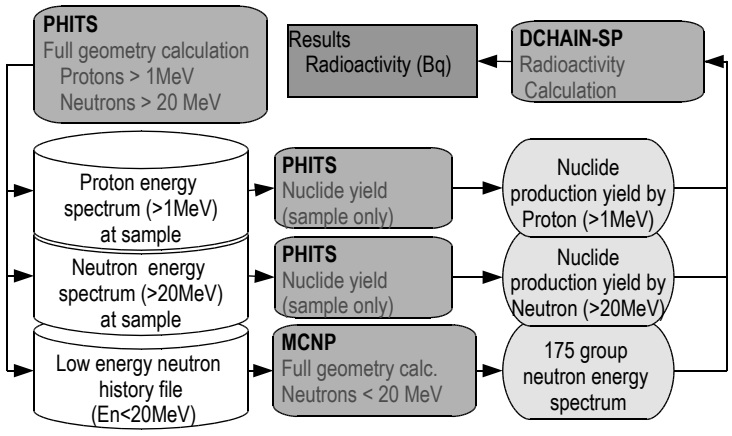

Fig. 2: Flow diagram of radioactivity calculation.

The number of protons in the off-beam samples and the number of neutrons (both below and above 20 MeV) were normalized to the incident protons measured by ICT. For the number of protons injected into the on-beam samples, we considered it was more reliable to utilize the results by the copper activation method using the foil with same size to the samples since not all protons were injected in the on-beam samples.

4 Results

4.1 Energy Spectra

Figure 3 shows calculated neutron and proton energy spectra in the iron samples at the on-beam and off-beam positions for the E_p=2.83 GeV case. At the on-beam position, the incident proton and the secondary neutrons produced in the Hg target are dominant. Spallation reactions, which produce many nuclides having quite different mass from the sample material, are mainly caused by incident protons. The dominant neutrons around 1 MeV initiate reactions producing nuclides having mass numbers close to the sample nuclides. At the off-beam position, the contribution of proton is almost negligible.

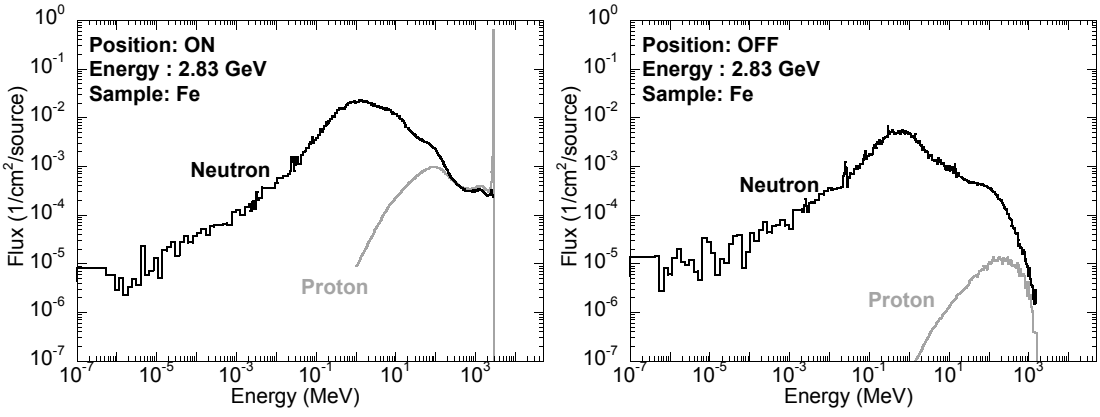

Fig. 3: Proton and neutron energy spectra at the on- and off-beam positions in the iron samples for the $E_p = 2.83$ GeV case.

4.2 Radioactivity

Figure 4 shows time evolution of radioactivity in the iron sample with E_p=2.83 GeV. Although most of the calculation results are lower than the experimental ones, the time evolution is almost consistent between them. Therefore, it is appropriate to take average over the cooling-time in discussions about ratios of the calculation to the experiment (C/E). Figure 5 shows C/E values for various radioactivity taking the average considering inverse squares of the experimental errors as weights. Minimum relative error among the original data was adopted as the error of the averaged C/E. Upper four figures in Fig. 5 show C/E of radioactivity in the iron samples at the on-beam and off-beam positions for the E_p=2.83 and 24 GeV cases. The measured nuclides are analogous among these results. Except for the E_p=24 GeV case at the off-beam position, the calculated radioactivity agrees with the experimental results by a factor of 2∼3 although the calculation trended to underestimate. The radioactivity of ^{24}Na is underestimated by one order of magnitude only in the E_p=24 GeV case at the off-beam position. For other nuclides, the C/E values are between 0.3 and 1.0. The reason why only the estimation of ^{24}Na in this case exhibited such large discrepancy is not understood. Excluding this data, the averages of C/E are 0.67, 0.84, 0.66 and 0.49 for the cases of E_p=2.83 GeV at the on-beam, E_p=24 GeV at the on-beam, E_p=2.83 GeV at the off-beam and E_p=24 GeV at the off-beam positions, respectively.

In middle of Fig. 5, C/E values are shown for the radioactivity in the copper samples. The results are only for the on-beam position since no copper sample was put at the off-beam position in the experiment. The calculated radioactivity agrees with the experimental results almost by a factor of 2∼3 although rather large discrepancies are found in ^{52}Mn. The averages of C/E are 0.58 and 0.80 for the E_p=2.83 and 24 GeV cases, respectively. The calculated cross sections by PHITS were compared with existing experimental ones for further discussions. The $^{\rm nat}$Cu(p,x)^{24}Na reaction was utilized as a reference to obtain the number of protons in the experiment. In E_p=2.83 GeV case, the cross section of 3.5±0.5 mb was adopted [9] while those of 2.4 mb was calculated by PHITS. The C/E value of 0.52 for ^{24}Na is reasonable considering the difference of the cross section, the error of the cross section (15%) and the uncertainty of ^{24}Na activity (5%).

In bottom four figures of Fig. 5, C/E values are shown for the radioactivity in the niobium samples. In all cases, good agreement is shown for $^{92\rm m}$Nb indicating that the calculated neutron fluences were reasonable between 11 and 20 MeV, in which the ^{93}Nb(n,2n)$^{92\rm m}$Nb reaction is sensitive. However, exceptional underestimation is recognized for ^{67}Cu, ^{76}As and ^{82}Br at the on-beam position, and for ^{46}Sc ^{57}Co and ^{60}Co at the off-beam position by one to three order of magnitude. On the other hand, ^{44}Sc and ^{76}Br in the E_p=24 GeV case at the on-beam

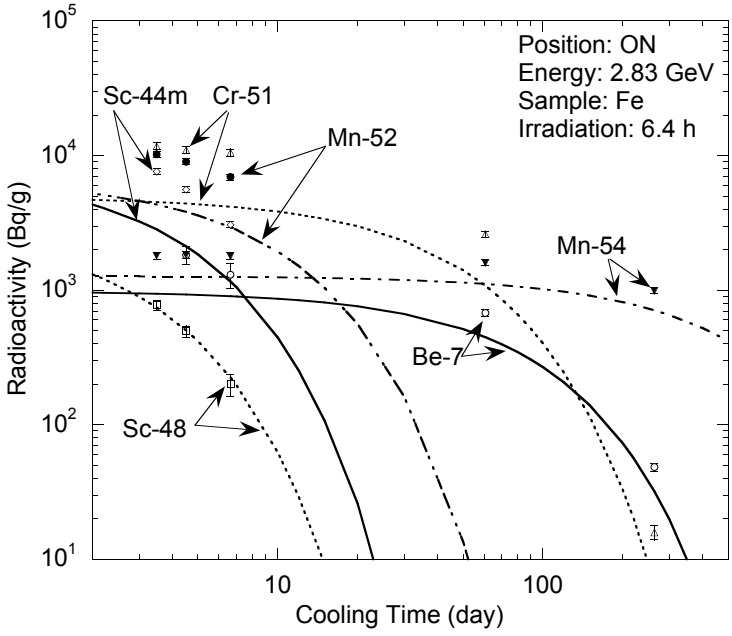

Fig. 4: Time evolution of radioactivity in the iron sample at the on-beam position in case of E_p=2.83 GeV. Symbols are the measured ones, lines are the calculated ones.

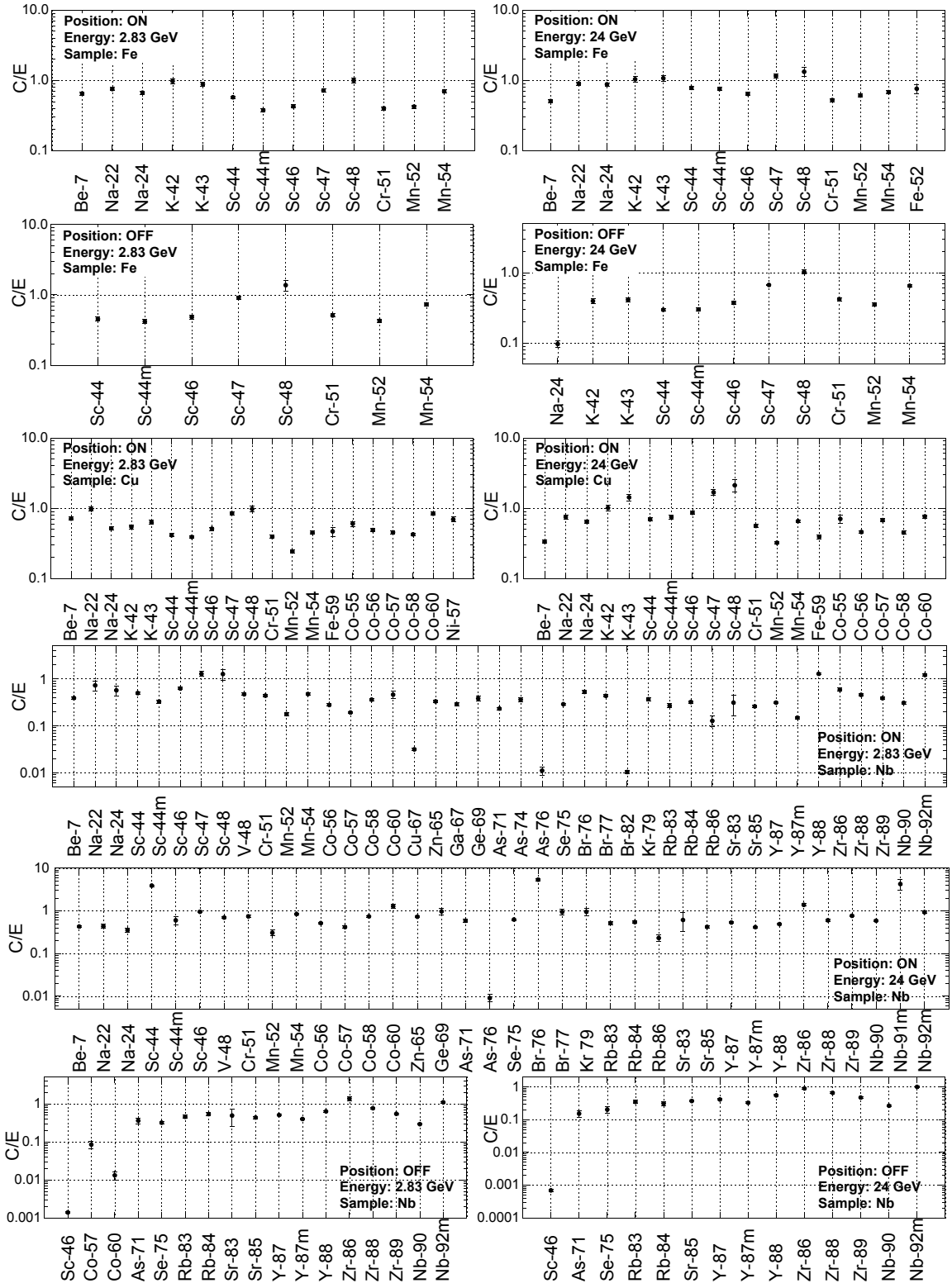

Fig. 5: C/E values for radioactivity in iron, copper and niobium samples. Sample positions, incident proton energies and sample materials are shown in figures.

position are overestimated by a factor of 4 and 5, respectively. The reason for the overestimation was found to be large C/E values at the shortest cooling time. Excluding such data, the C/E values are improved to be 0.56 for ^{44}Sc and 1.1 for ^{76}Br. Although C/E for ^{91m}Nb is also large, there is not such a discrepancy as in the above case. Assuming all nuclides were produced in the grand state by selection of the ISOMER parameter in DCHAIN-SP, C/E for ^{91m}Nb was underestimated by a factor of forty. As mentioned in the section 3, one of the reasons for the overestimation is considered to be that nuclides in the grand state and all isomers were assumed to be produced with equal probabilities. Excluding the exceptional data, the averages of C/E are 0.47, 0.78, 0.59 and 0.46 for the cases of E_p=2.83 GeV at the on-beam, E_p=24 GeV at the on-beam, E_p=2.83 GeV at the off-beam and E_p=24 GeV at the off-beam positions, respectively.

5 Conclusion

The radioactivity experiment was analyzed to estimate the calculation reliability of the code system. The calculations were consistent with the experiments within a factor of 2 on the average over the produced nuclides excluding the data exhibiting the exceptional discrepancies although the calculations were lower than the experiment on the whole. The overestimation for ^{91m}Nb produced from the niobium sample was caused by the assumption that a nuclide in the grand state and all isomers were produced with equal probabilities. Further discussions are important to improve the calculation reliability, however, we concluded that the code system was available to estimate the radioactivity in iron, copper and niobium. We considered this conclusion was also valid for other nuclides having middle mass number.

Acknowledgements

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