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Measurement of induced radioactivity in a spallation neutron field of a mercury target for GeV-proton bombardment

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Abstract
An integral experiment on radioactivity induced in spallation neutron fields was carried out under the ASTE collaboration using AGS at BNL. The spallation neutrons were produced by bombarding a mercury target with protons of 1.6, 12 and 24 GeV. The number of protons was 3~4×10^{13} for each irradiation. The irradiated materials were titanium, nickel, cobalt, yttrium, and bismuth, and placed on the cylindrical surface of the mercury target at the distance of 15~16 cm from the beam-incident-surface of the target. Disintegration rates of induced radioactivities were measured at several cooling-time ranging from hours to months. The principal nuclides contributing to the radioactivity were pointed out for each material. The experimental results for bismuth were compared with the calculations with DCAHIN-SP code.

1. Introduction

The Joint project for high-intensity proton accelerators is being proposed by Japan Atomic Energy Research Institute (JAERI) and the High Energy Accelerator Research Organization (KEK) [1]. Under the project, an intense spallation neutron sources driven by 3 GeV protons with power of MW are planning to be constructed for expanding fundamental research in life science, material science, and so on. In order to design the facility, accurate estimation of radioactivity induced in radiation fields associated with an enormous amount of neutrons as well as GeV protons are required in the viewpoint of radiation safety. For estimating the activities of radioactive products, the DCHAIN-SP code has been developed in JAERI [2]. The DCHAIN-SP code can calculates the nuclide inventory, radioactivity, decay-heat and gamma-ray spectra coupled with the nucleon-meson transport code NMTC/JAM [3, 4]. However the code system have not been validated well due to lack of available experimental data.

An international collaboration on a spallation neutron target experiment at the Alternating Gradient Synchrotron of Brookhaven National Laboratory, namely ASTE collaboration, was organized to study neutronics characteristic of the mercury target and mechanical properties of the target and its container. As for the experiment on neutron characteristic, the reaction rate distributions for some kinds of threshold reactions were measured by means of the activation method [5, 6]. Using the reaction rate data, the neutron yield distribution around the mercury target was obtained, and the neutron spectrum was deduced by means of the unfolding method [7]. These experimental data were used for validating the Monte Carlo code system for simulating nuclear interaction and particle transportation, the NMTC/JAM-MCNP code, which will be used to optimize a spallation target system.

In this work, we focus on the activites induced by the spallation neutrons and their decay properties. In the ASTE experiment, the activities of the irradiated samples were measured at different cooling-time ranging from hours to months after the spallation neutron irradiation. For the validation of DCHAIN-SP code system, we provide the experimental data on the radioactivities for some materials.

The experimental procedure is described in Section 2. In Section 3, some of the experimental data are presented, and principal radioactive nuclei are pointed out for each material. The experimental results for bismuth were compared with the calculations with DCAHIN-SP code as the first step of the code validation.

2. Experiment

The mercury target was set in the U-line tunnel between AGS and Relativistic Heavy Ion Collider. Mercury was contained in a cylindrical target container made of stainless steel with 2.5 mm in thickness. The dimension of the target container was 200 mm in inner diameter and 1300 mm in inner length. The upper and side view of the mercury target are shown in **Fig. 1**. For the neutronics experiment, the activation foils were put on the 4 acrylic bars ("main" and "sub-1~3", see Fig. 1) and then installed on the four radial positions along the beam axis as shown in the **Fig. 2**. More detailed description of the target container is described in Ref. 6.

The shapes of titanium, cobalt, nickel and yttrium-foils are square with 20 mm × 20 mm and 1 mm in thickness. The bismuth-foils have round shape with 15 mm in diameter and 1 mm in thickness.

The incident proton energies were 1.6, 12 and 24 GeV. The number of protons were determined with the activation method using the $Cu(p,x)^{24}Na$ reaction [8]. The number of protons was $3\sim4\times10^{13}$ for each irradiation. The parameters of the proton bombardment are summarized in Table 1.

The activities of each foil were measured with Germanium detectors at the cooling-time from 2 hours to 2 months. Experimental data were derived as the radioactivity intensity per unit weight (Bq/g). The radioactivity was identified by gamma-ray energies and their intensity relationship. The activity of each radioactive product was deduced by considering the radioactive decay during a measurement. The decay property data were taken from Table of Isotopes [9]. When there were multiple gamma-ray emission associated with decay, the most intense gamma-ray line was used for the data processing. The gamma-ray detection efficiency was corrected for geometric effects of foil sizes, gamma-ray self-absorption in foils and coincidence-summing effect.

The sources of experimental errors were mainly attributed to the following items: 1) statistical error of gamma-ray peak count, 2) error of the gamma-ray detection efficiency, and 3) error of the half-life and the gamma-ray emission rate. In this work, the data with the statistical error more than 50% were not adopted. The error of detector efficiency was estimated to be less than 3~5% by considering the error of the standard source activities for the calibration. The errors of the decay data were negligibly small in comparison with the other sources in almost all cases.

3. Results and Discussion

According to the measured reaction rate of $^{115}In(n,n')^{115m}In$, the peak position of the spallation neutron yield distribution is ranging the distance of 10~20 cm from the beam-incident-surface of the target as shown in **Fig. 3**. In this paper, we present the results for the titanium, cobalt, nickel, and bismuth foils irradiated on the "main"-bar at the distance of 15~16 cm from the beam-incident-surface of the target.

The experimental results for 1.6 GeV proton bombardment are shown as a function of a cooling time in **Figs. 4~8**, respectively. In the figures, decay curves were drawn by fitting the experimental data. For titanium (**Fig. 4**), the principal nuclei contributing to the radioactivities were ^{44g}Sc, ^{47}Sc and ^{46g}Sc. For cobalt (**Fig. 5**), the radioactive nuclide of ^{56}Mn with half-life of 2.58 h is the main contributor to the total activities before 14 h cooling. After the cooling time more than 1 day, the radioactivity of ^{58g}Co was the strongest radioactive source in the sample. The activity of ^{58g}Co had increased before 2 days cooling due to the isomeric transition from ^{58m}Co with the half-life of 9.2 h. The decay curve was drawn by considering the contribution of the isomer in **Fig. 5**. The radioactivity of ^{60}Co with half-life of 5.2 y can be expected to be the pricncipal radioactivity after 100 days cooling. The principal nuclei for the nickel sample are ^{57}Ni at $T_c < 2$ d, where T_c is the cooling time, and ^{58}Co at $T_c > 2$ d, as sown in **Fig. 6**. For yttrium (**Fig. 7**), the principal radioactive products were ^{87m}Y ($T_c < 2$ d), ^{87g}Y ($T_c < 2$~15 d) and ^{88}Y ($T_c > 15$ d). For bismuth (**Fig. 8**), ^{204}Bi ($T_c > 1$ d), ^{203}Pb ($T_c \sim 2$ d), ^{206}Bi (3 d $< T_c <$ 20 d) and ^{205}Bi ($T_c > 20$ d) were the principal radioactive products. The activity of ^{203}Pb had been increasing before 2 days cooling due to the decay of ^{203}Bi to ^{203}Pb. It is expected that ^{207}Bi with the half-life of 33.4 y is the principal activity at $T_c > 100$ d.

The experimental data for bismuth were compared with the calculation with DCHAIN-SP code system. Flow of the calculation using DCHAIN-SP is illustrated in **Fig. 9**. In the code system, the nuclide production induced by the neutrons with the energy below 20 MeV were calculated using the neutron cross section data taken from FENDL/A-2 data library and the neutron energy spectrum calculated with the NMTC/JAM-MCNP code system. For the nuclide production induced by the neutrons with energies more than

20 MeV, the production rates calculated with the nucleon-meson transport code NMTC/JAM were used as input data. It is noted that, in the present version of DCHAIN-SP code, the nuclide production rate calculated with the NMTC/JAM code were considered as the "ground-state" nuclide production rate. This means that isomer production for the neutron with >20 MeV was not taken into account.

The calculated decay curve for bismuth (E_p=1.6 GeV) are shown in **Fig. 10**. For all proton energies, the calculated values show underestimation. In particular, for E_p= 12 and 24 GeV, the calculated values are lower than the experimental ones by a factor more than 2 for all nuclide except ^{206}Bi. These discrepancies are due to the underestimation of the nuclide production yields calculated with NMTC/JAM because all the nuclei listed in Table 4 are produced via the reactions induced by neutrons with energy more than 20 MeV.

5. Conclusion

The activity of the metal foils irradiated by the spallation neutrons generated by bombarding 1.6, 12 and 24 GeV protons with the mercury target were measured using AGS accelerator. The titanium, cobalt, nickel, yttrium and bismuth-foils were placed on the cylindrical surface of the mercury target at the distance of 15~17 cm from the beam-incident-surface of the target. After the irradiation, induced activities were measured at several cooling-time ranging from hours to months. The principal radioactive nuclei are pointed out for each material. The experimental results for bismuth were compared with the calculations using the DCAHIN-SP code system as the first step of the code validation. The calculation showed the underestimation. It is due to the to the underestimation of the nuclide production yields calculated with NMTC/JAM. This work provided the first experimental data to validate the DCAHIN-SP code system.

Acknowledgement

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Table 1 Parameters of the proton bombardment

E_p (GeV) [a]	N_p ($\times10^{13}$) [b]	Time(s)
1.6	3.7±0.3	3150
12	4.8±0.4	1078
24	4.2±0.4	240

[a] Proton Energy
[b] The number of protons
[c] Time between the first and last proton pulse bombardment

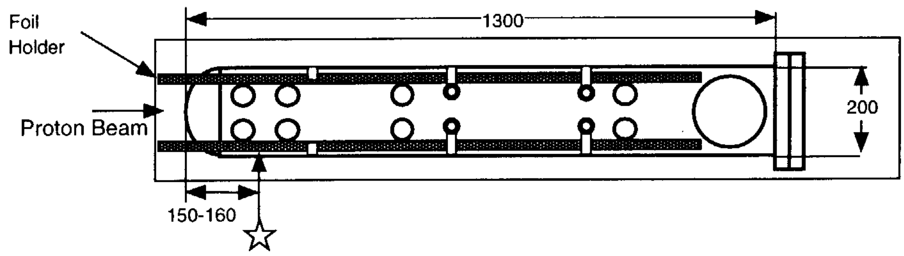

Fig. 1 Upper view of the mercury target. The activation foils on which we focused in this study were set on the position at the distance of 15~16 cm from the beam-incident-surface of the target. The foil-position is marked in "☆" in the figure. The units of values in the figure are in mm.

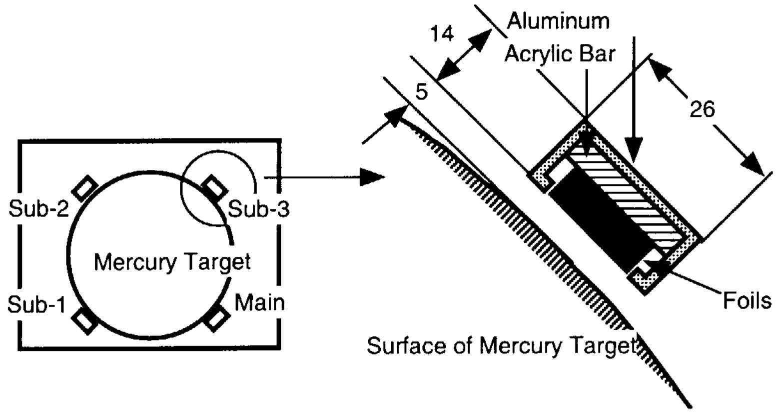

Fig. 2 Arrangement of activation-foil holders. The incident protons penetrate from the front to back surface. The samples in the present work were put on the "Main"-bar. The units of the values in the figures are in mm.

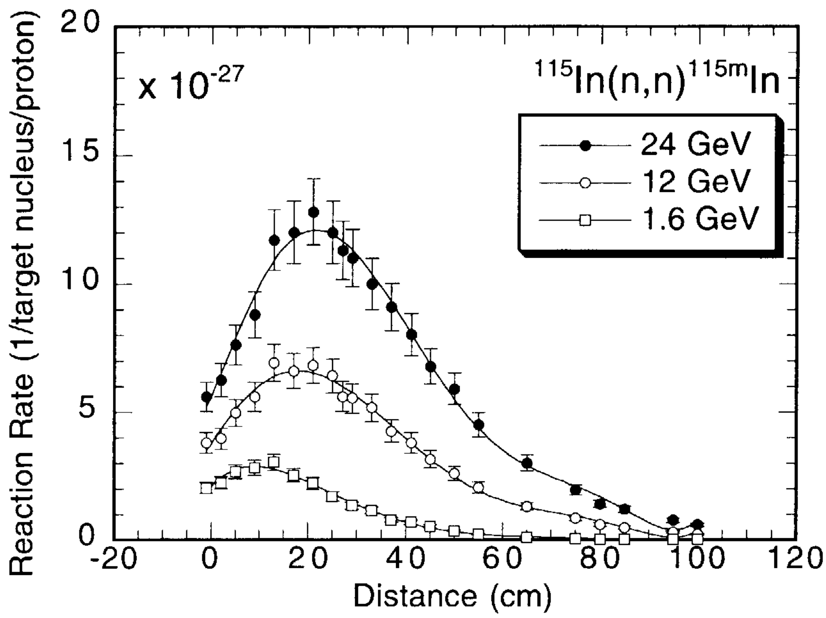

Fig. 3 The reaction rates of the $^{115}In(n,n')^{115m}In$ reactions on the main bars are plotted as a function of the distance from the beam-incident-surface of the target. The peak position of the spallation neutron yield distribution is ranging the distance of 10~20 cm from the beam-incident-surface of the target

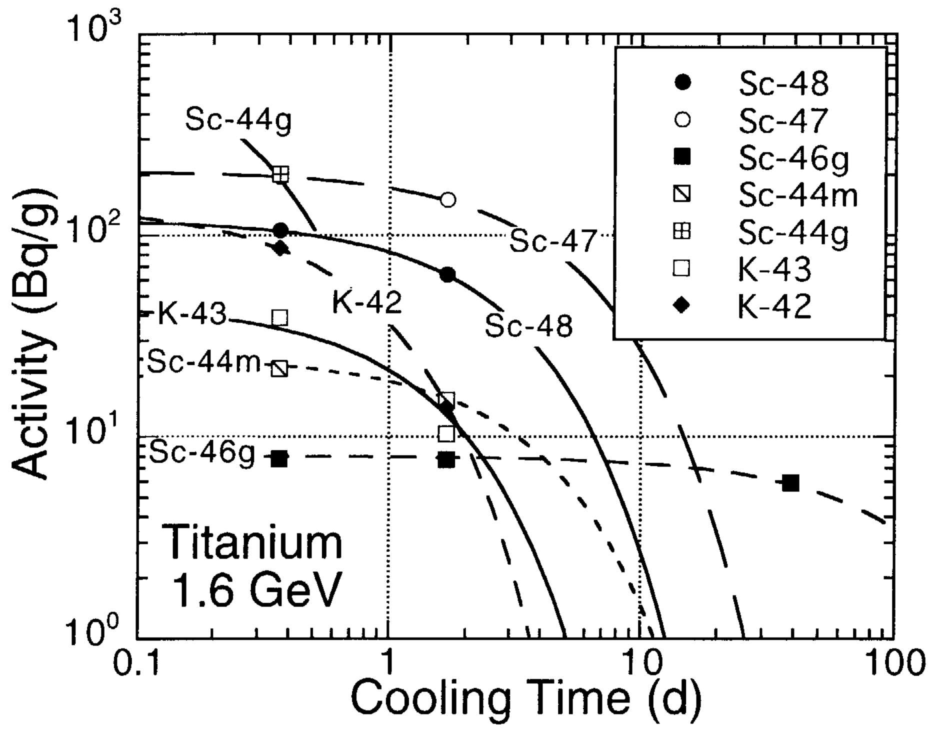

Fig. 4 Measured radioactivities of the titanium sample for 1.6 GeV proton bombardment at different cooling times and the decay curves fitted to the experimental data.

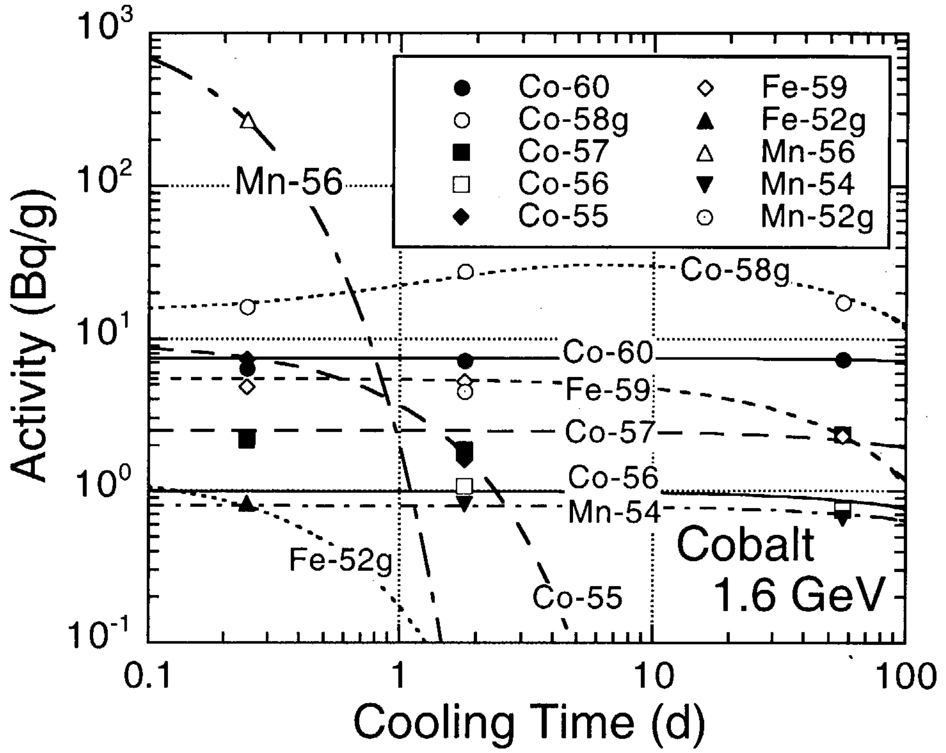

Fig. 5 Measured radioactivities of the cobalt sample for 1.6 GeV proton bombardment at different cooling times and the decay curves fitted to the experimental data.

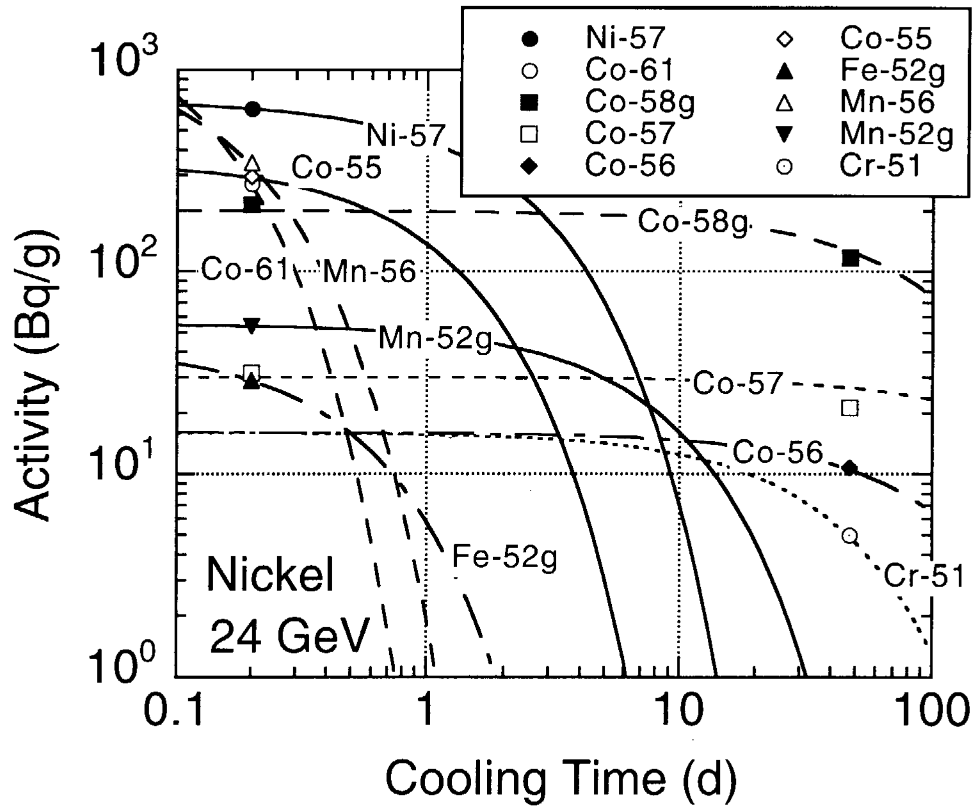

Fig. 6 Measured radioactivities of the nickle sample for 24 GeV proton bombardment at different cooling times and the decay curves fitted to the experimental data.

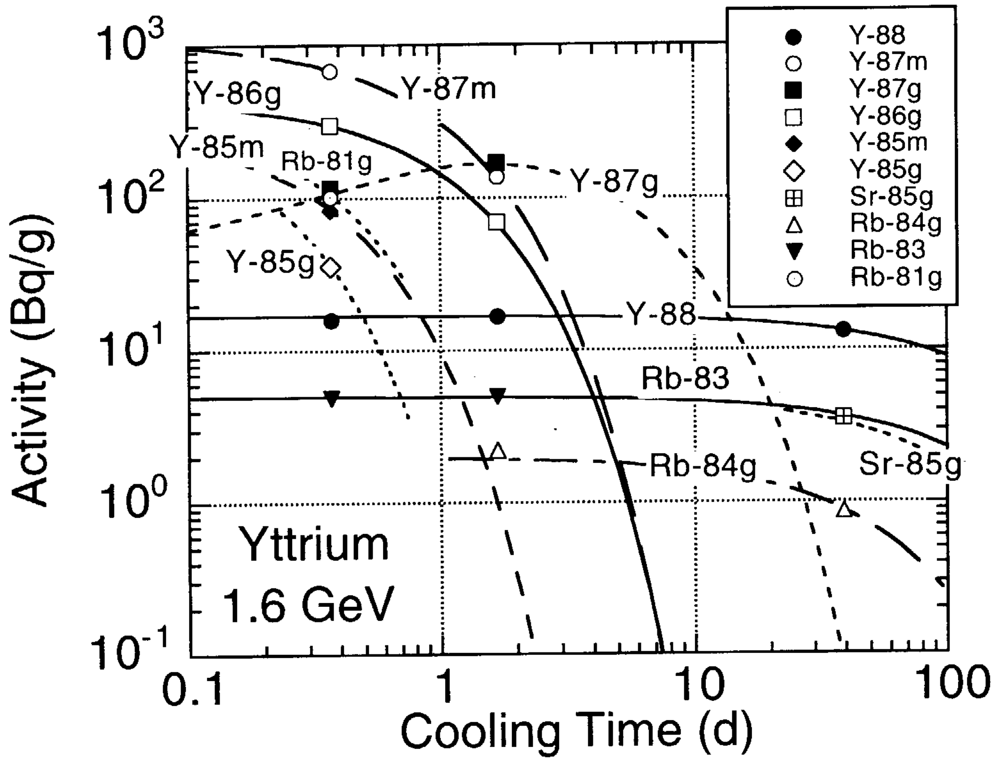

Fig. 7 Measured radioactivities of the yttrium sample for 1.6 GeV proton bombardment at different cooling times and the decay curves fitted to the experimental data.

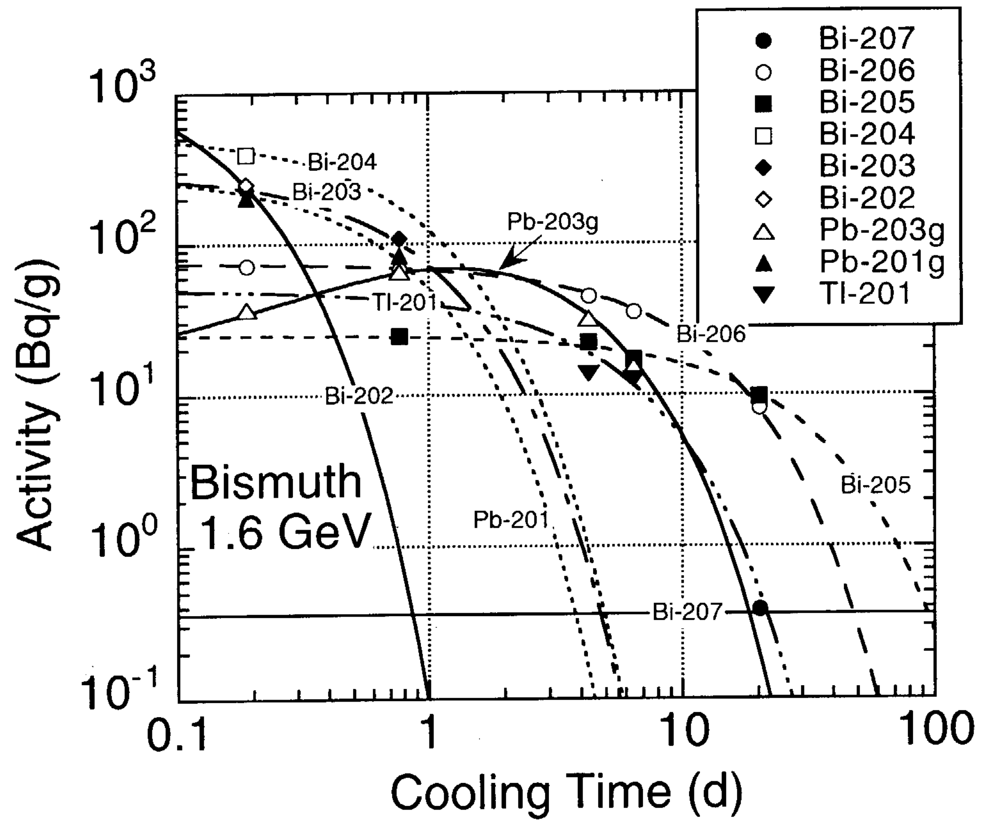

Fig. 8 Measured radioactivities of the bismuth sample for 1.6 GeV proton bombardment at different cooling times and the decay curves fitted to the experimental data.

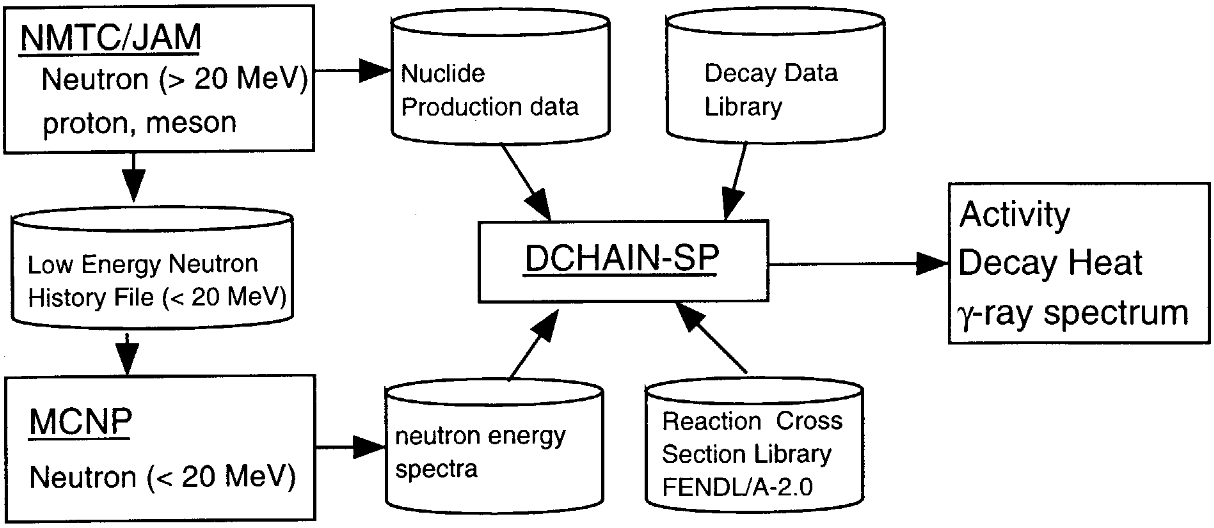

Fig. 9 Flow of the calculation using DCHAIN-SP.

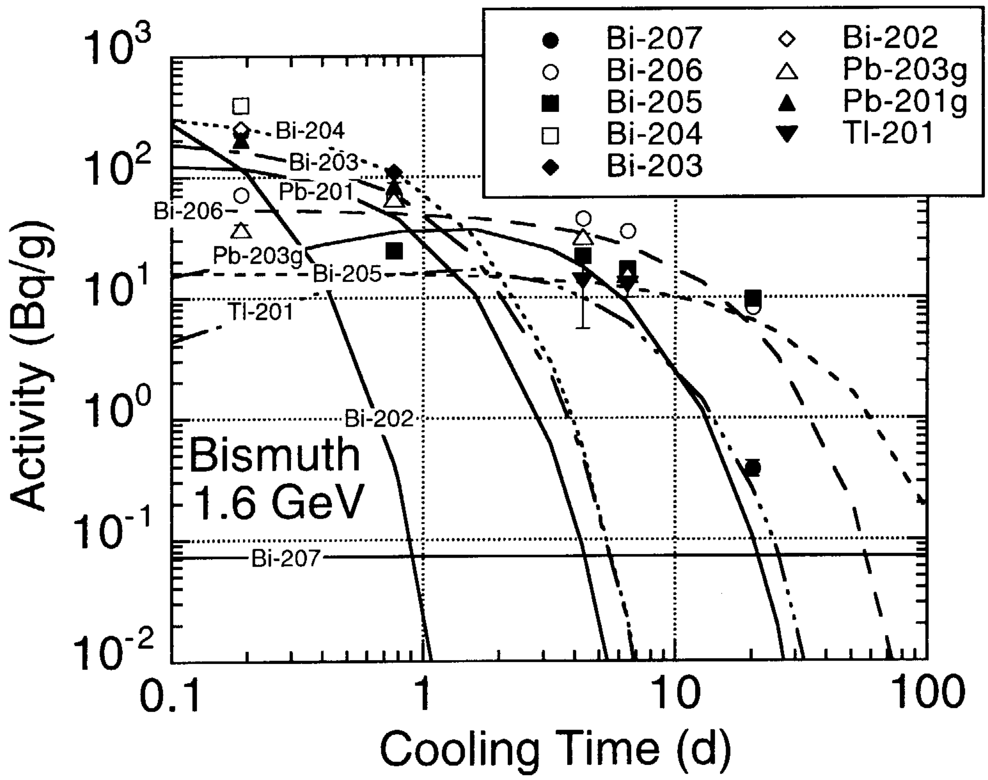

Fig. 10 Measured radioactivities of the bismuth sample for 1.6 GeV proton bombardment at different cooling times and the decay curves calculated with DCHAIN-SP code.