PRODUCTION YIELDS OF THE RADIONUCLIDES
INDUCED IN VARIOUS TARGETS
FIXED IN CONCRETE SHIELD
AT THE 500-MEV NEUTRON IRRADIATION FACILITY OF KENS

H. Matsumura¹, N. Nakao¹, K. Masumoto¹, K. Oishi², M. Kawai¹, T. Aze³, A. Toyoda¹,
M. Numajiri¹, K. Takahashi¹, M. Fujimura⁴, Q. Wang⁵, K. Bessho¹, T. Sanami¹
¹ High Energy Accelerator Research Organization (KEK), Tsukuba, Japan
² Institute of Technology, Shimizu Corporation, Tokyo, Japan
³ Graduate School of Science, The University of Tokyo, Tokyo, Japan
⁴ Graduate School of Integrated Basic Sciences, Nihon University, Tokyo, Japan
⁵ The Institute of High Energy Physics (IHEP), Beijing, China

Abstract

We performed a shielding experiment with high-energy neutrons produced from a tungsten target bombarded with 500-MeV protons and penetrated through a concrete shield in the 0 degree direction. Using an activation method, we observed many radioactive products induced by neutrons with energies ranging from thermal to 500 MeV and obtained their production yields from various target elements at depths of 0 to 4 m from the surface of the concrete shield.
Introduction

In high-energy proton accelerator facilities, high-energy neutrons are secondarily produced upon bombardment with a proton beam. Such neutrons have high penetrability, and induce activation of the surrounding materials. The spectra of the neutrons and activation of the materials are generally estimated using Monte-Carlo simulation codes. It is very important to confirm experimentally their confidence from the benchmark.

However, only few experiments for the benchmark have been performed so far. In particular, there are no experimental data pertaining to the conditions of “high energy neutron”, “0 degree direction”, and “thick concrete shield.” We therefore constructed a high-energy neutron-beam course at KEK Neutron Science Laboratory (KENS) at the High Energy Accelerator Research Organization (KEK) [1, 2]. The course has seven irradiation slots inside a 4-m-thick concrete shield. High-energy neutrons were guided to the shield through a collimator of 2.19-m length from a tungsten-target assembly which completely stopped 500-MeV primary protons.

Using this course, we have been accumulating experimental data for following: (1) the spectrum and attenuation characteristics of the neutrons in the concrete shield and (2) the activation products induced by high-energy neutron irradiation in the well-known system at KENS. With regard to the first point, an activation method is most suitable for detecting neutrons in the limited spaces in the concrete. Therefore, we have measured the yields of the radioactive products from various targets and observed the attenuation profiles of the neutrons. The yields obtained from the various nuclear reactions with different $Q$-values can indirectly provide information on the energy spectra of the neutrons and their intensities. With regard to the second point, we measured many yields of the spallation products from various targets induced by high-energy neutrons. Following this, charge distributions and mass yield curves were elucidated from the yields. In this paper, we introduce a part of our experimental data [1-7], including the preliminary data, obtained using the high-energy neutron-beam course at KENS.

Experimental procedure

The 500-MeV neutron irradiation facility of KENS

The high-energy neutron-beam course of KENS was designed for this shielding experiment. Figure 1 shows the cross-sectional views of the KENS high-energy neutron-beam course and the arrangement of the shielding experiment. Secondary neutrons were produced by bombarding with 500-MeV protons on four tungsten blocks placed in the tungsten-target assembly. Since the total thickness of the tungsten blocks was 11.68 cm, the primary protons were completely stopped in the tungsten blocks. The produced secondary neutrons were passed through a 10-cm stainless-steel block placed downstream in the target assembly, and collimated with an iron beam guide to lead in zero-degree direction. The collimated beam finally reached to a beam exit of 20 cm width × 15 cm height located in a distance of 250 cm from the target assembly and irradiated on an ordinary concrete of 4 m thickness which borders on the beam exit. The concrete has eight irradiation positions referred
to as “slot 1” to “slot 8”. They are positioned along the neutron beam direction in the concrete shield. The slots 1, 2, 3, 4, 5, 6, 7, and 8 were located on 0, 40, 80, 130, 185, 250, 320, and 400 cm depth from the surface of the concrete shield, respectively. The more details of this course were described in [1, 2].

**Irradiation**

The targets of Na$_2$CO$_3$, MgO, Al, Si, KCl, K$_2$CO$_3$, CaCO$_3$, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, In, Au, Au covered with Cd, and Bi were irradiated at slots 1, 2, 3, 4, 5, 6, 7, and/or 8. The targets were individually sealed in polyethylene bags. Plastic capsules containing the targets were fixed at the bottom of the shield plugs and inserted into the 8 slots, and set on the beam axis. Nine irradiations were carried out. The irradiation duration was typically 1 week. The average current of the primary protons was typically 6-7 $\mu$A. The fluctuation of the primary proton current was monitored during the irradiation. The beam fluctuation was corrected for radioactive decays if necessary.

**Figure 1. Cross-sectional views of the KENS high-energy neutron-beam course and the arrangement of the shielding experiment**
Measurements of the radioactive products

Spallation products from $^{197}$Au

After irradiation, $\gamma$-ray spectrometry was performed with HP Ge detector systems. The radioactivities of $^{196}$Au, $^{194}$Au, $^{192}$Au, $^{188}$Pt, $^{189}$Ir, $^{185}$Os, $^{183}$Re, $^{181}$Re, and $^{175}$Hf were determined non-destructively. On the other hand, $^{173}$Lu, $^{171}$Lu, $^{169}$Yb, and $^{167}$Tm could not be detected directly because of their low activity and high activity of $^{198}$Au and neighbor spallation products. Therefore, they were measured after radiochemical group-separation of rare-earth elements. The radioactivity measurements of target set in the backward from slot 5 were more difficult than that set at front side due to their attenuation. For the targets of slots 5 to 7, low background $\gamma$-ray counting was performed with an anti-Compton and/or X-$\gamma$ coincidence system which HP Ge detector coupled with 3 NaI scintillation counters. All of the yields were cumulative for the respective $\beta^+$-decay and/or electron-capture chains. The details of the samples for the targets of slots 1 to 5 and the chemical procedure were described in [5]. Since the details of the samples and data analysis for the targets of slots 5 to 7 are preliminary, they have not published yet.

Chlorine-36

After irradiation, chlorine was chemically separated from each irradiated target and finally precipitated as AgCl. The amount of the $^{36}$Cl was determined by accelerator mass spectrometry (AMS) in the Micro Analysis Laboratory, Tandem Accelerator (MALT) at the Research Center for Nuclear Science and Technology, the University of Tokyo. The details were described in [6].

Other products

After irradiation, $\gamma$-ray spectrometry was nondestructively performed with HP-Ge detectors to determine the radioactivities of the products from the various targets. The yields of $^{22},^{24}$Na from Cl were obtained by subtracting the yields in K$_2$CO$_3$ from those in KCl. In order to detect $^{198}$Au from Au and Au covered with Cd, the irradiated gold foils were also simultaneously exposed to an imaging plate (IP). The exposed IP was scanned with a bio-imaging analyzer. The details of the activity determination with the IP were described in [3]. The yields induced by epithermal neutrons were measured from the Au targets covered with Cd. We could obtain the yields induced by thermal neutrons by subtracting the yields in the Cd-covered Au from those in the non-covered Au.

Results and discussion

Many yields were collected in this work. The unit of the yield is atom$^{-1}$μC$^{-1}$, which indicates the production probability per target atom and per μC of the primary protons. We explain the measured yields in following two separate sections.
In the activation method, the detectable energy of the neutrons depends on the reaction threshold energy. Using the energy dependence, we can indirectly derive information on the spectrum profile, intensity, and attenuation of the neutrons from the product yields in the various reactions. Accordingly, at the slots 1-8, we measured the yields of the following products:

1. 13 spallation products ($^{196}$Au, $^{194}$Au, $^{192}$Au, $^{188}$Pt, $^{189}$Ir, $^{185}$Os, $^{183}$Re, $^{181}$Re, $^{175}$Hf, $^{173}$Lu, $^{171}$Lu, $^{169}$Yb, and $^{167}$Tm) from a $^{197}$Au target,
2. $^{202}$-$^{206}$Bi in $^{209}$Bi(n, xn) reactions,
3. $^{198}$Au from a $^{197}$Au target and a $^{197}$Au target covered with Cd,
4. $^{115m}$In from $^{115}$In,
5. $^{36}$Cl from natCl, natK, and natCa targets, and,
6. $^{24}$Na and $^{22}$Na from $^{23}$Na, natMg, $^{21}$Al, natSi, natCl, natK, and natCa targets.

The effective energies of the yields are estimated at to be 8-500 MeV for (1), 10-100 MeV for (2), thermal and epithermal reactions for (3), and 1-10 MeV for (4), and thermal to several 100 MeV for (5) and (6). In particular, the threshold energy in the Au spallation products covers the widest energy range among the measured targets in this work. The threshold energy of $^{197}$Au(n, 2n)$^{196}$Au is 8.1 MeV, which is the lowest among the (1) products. The effective neutron energy for this reaction is estimated at around 10 MeV. On the other hand, the threshold energy of $^{197}$Au(n, 10p21n)$^{167}$Tm is 202.9 MeV, which is the highest among the measured reactions. The effective neutron energy for this reaction is estimated to be in the range of 350 to 500 MeV [5]. As the results, the effective energies of the 13 Au spallation products are in the range of 10 to 500 MeV. Furthermore, whole neutron energies were completely covered using all the productions of (1) to (6).

In Figure 2, the measured yields of (a) the spallation products from $^{197}$Au, (b) the products of the $^{209}$Bi(n, xn) reactions, (c) $^{198}$Au from $^{197}$Au, $^{36}$Cl from Cl, K, and Ca, and $^{115m}$In from $^{115}$In, and (d) $^{24}$Na and $^{22}$Na were plotted as a function of the depth from the surface of the concrete shield. The open and the closed symbols joined by solid lines represent the reported ([1, 5, 6]) and preliminary experimental yields, respectively. All the yields are exponentially attenuated in the concrete shield and their slopes appear similar regardless of the reactions.

In order to compare the details of the relative yield variations, the ratios of the yields at each slot to those at slot 2 are plotted in Figure 3. The open symbols denote the experimental values published in [1, 5, 6], while the closed symbols denote the preliminary values. The solid lines indicate the average for the Au spallation products. This figure shows the relative change in the energy spectra of the neutrons passing through the concrete shield. The ratios are almost constant except in the low-energy reactions such as $^{35}$Cl(n, $\gamma$)$^{36}$Cl, $^{197}$Au(n, $\gamma$)$^{198}$Au, $^{197}$Au(n, 2n)$^{196}$Au, and $^{115}$In(n, n'$)^{115}$In where the threshold energy is lower than approximately 10 MeV. The ratios of the yields of the low-energy reactions are slightly higher than the others at slots deeper than slot 3. Therefore, without changing the profile of the neutron spectrum in the range of 10 to 500 MeV, the intensity is exponentially attenuated with an increase in the depth of the concrete, although there was a small build-up of low-energy neutrons at depths below 40 cm.
Figure 2. Measured attenuation profiles of the various yields of
(a) the spallation products from $^{197}$Au, (b) products of $^{209}$Bi(n, xn) reactions,
(c) $^{198}$Au from $^{197}$Au, and $^{36}$Cl from Cl, K, and Ca, and (d) $^{24}$Na and $^{22}$Na from various targets
Production yields of radioisotopes from various targets

The yields obtained from the Au target are also available for the benchmark of the residual products from heavy targets. All the yields are cumulative for the respective $\beta^+$-decay and/or electron-capture chains. Since the measured nuclides are closed to stable isotopes, the yields can be approximately regarded as mass yields according to the spallation characteristics. In Figure 4, the yields from Au target are plotted as a function of the product mass number. The open symbols denote the experimental values published in [5], while the closed symbols denote the preliminary experimental values. The solid lines drawn through the points are provided as a visual guide. On the semi-log graph, it is evident that each mass yield curve decreases steeply in an arc with a decrease in the product mass number.

For the benchmark of the residual products from medium targets, we measured the yields of the spallation products from the $^{45}$Sc, $^{nat}$Ti, $^{nat}$V, $^{nat}$Cr, $^{55}$Mn, $^{nat}$Fe, $^{59}$Co, $^{nat}$Ni, $^{nat}$Cu, and $^{nat}$Zn targets at slot 2. Although the charge distribution characteristics were unclear for the neutron-induced spallation, we attempted to fit the spallation yields to an empirical five-parameter formula derived by Rudstam [9] that was successfully applied to photospallation reactions [8]. Rudstam’s formula (CDMD formula) for charge distribution (CD) and mass yield distribution (MD) is

$$Y(Z, A) = \frac{\sigma P R^{23} \exp[PA-R | Z-SA+TA^2 | 3/2]}{1.79 \{\exp(PA)-1\}}$$

, where $Y(Z, A)$ indicates the independent yield of a nuclide $(Z, A)$ obtained from the target $(Z_t, A_t)$ and $P, \sigma, R, S,$ and $T$ are free parameters. The parameter $P$ defines the slope of the MD; $\sigma$, the total
inelastic cross section; $R$, the width of the CD; and $S$ and $T$, the location of the CD through the most probable charge $Z_p = S - TA^2$. Figure 5 shows the CD of the spallation products from the $^{45}$Sc, $^{nat}$Ti, $^{nat}$V, $^{nat}$Cr, $^{55}$Mn, $^{nat}$Fe, $^{59}$Co, $^{nat}$Ni, $^{nat}$Cu, and $^{nat}$Zn targets. The open squares indicate independent yields (I); reverse triangles, cumulative ones for $\beta^+$ and/or electron-capture decay (C+); and triangles, cumulative ones for $\beta^-$ decay (C-). The solid lines indicate the CD obtained by the fitting, and the dashed and dotted lines represent the cumulative yields for C+ and C- estimated using the CDMD formula, respectively. As shown in the figure, the results of these fittings are very good for all the products and targets. Therefore, it was confirmed that the CDMD formula was useful for the yields of the neutron spallation residues obtained at KENS.

Mass yield curves were calculated using the CDMD parameters. Figure 6 shows the MD for the $^{45}$Sc, $^{nat}$Ti, $^{nat}$V, $^{nat}$Cr, $^{55}$Mn, $^{nat}$Fe, $^{59}$Co, $^{nat}$Ni, $^{nat}$Cu, and $^{nat}$Zn targets. The solid lines represent the calculated mass yields. The squares indicate the experimental mass yields compensated with estimated yields of unmeasured products. We were able to obtain the MD for a wide range of product mass number.

Furthermore, we have been measuring the yields of the spallation products from other targets such as $^{89}$Y. Systematic measurements of fixed products, such as $^7$Be, $^{10}$Be, $^{24}$Na, and $^{22}$Na, from various targets are also useful for benchmark. The yields will be reported in the near future, some of them have already been published in [7].

**Figure 4. Mass yields of the spallation products from $^{197}$Au**
**Conclusion**

Applying an activation method, we successfully investigated the spectrum profile, intensity, and attenuation of the neutrons in the concrete shield at KENS. The profile of the neutron spectrum ranging from 10 to 500 MeV was kept and the intensity was exponentially attenuated with an increase in the depth of the concrete, although there was a small build-up of low-energy neutrons at depths below 40 cm. Furthermore, the CDMD formula was demonstrated to be suitable for the neutron spallation at KENS. Using this formula, the charge distributions of the yields and mass yield curves of medium targets were obtained. Furthermore, mass yield curves in an Au spallation are available to the benchmark the residual products of heavy targets. In addition to the data, we will provide the further data in the near future.
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REFERENCES


