MC simulation of thermal neutron flux of large samples irradiated by 14 MeV neutrons

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Abstract The response of a 14 MeV neutron-based prompt gamma neutron activation analysis (PGNAA) system, *i.e.* the prompt gamma-rays count rate and the average thermal neutron flux, is studied with a large concrete sample and with a homogeneous large sample, which is made of polyethylene and metal with various concentrations of hydrogen and cadmium using the MCNP-5 (Monte Carlo N-Particle) code. The average thermal neutron flux is determined by the analysis of the prompt gamma-rays using the thermal neutron activation of hydrogen in the sample, and the thermal and fast neutron activation of carbon graphite irradiation chamber of the PGNAA-system. Our results demonstrated that the graphite irradiation chamber of the PGNAA-system fairly operates, and is useful to estimate the average thermal neutron flux of large samples with various compositions irradiated by 14 MeV neutrons.

Key words Prompt gamma neutron activation analysis, Large samples, MCNP-5 code

1 Introduction

In Germany, the declaration and balancing of toxic substances in low and intermediate level radioactive waste (LILW) have become obligatory as a result of the plan-approval for disposal Konrad[1]. Depending on its origin, LILW may contain toxic elements (Pb, Cd, Hg, and alkali metals), anions (nitrates, sulphates, chlorides, and chlorates), and toxic organic chemical compounds[2]. Radioactive waste containing such toxic substances must comply with the regulations defined by the German authority and their properties need to be taken into account for a safe disposal.

In order to achieve this, the limiting values for toxic elements and substances were defined by the Federal Office of Radiation Protection (BfS)[3] for the final disposal of radioactive waste packages in the mine Konrad. To determine toxic elements in radioactive waste packages, a non-destructive analytical technique based on PGNAA (Prompt-Gamma-Neutron-Activation-Analysis) with a 14 MeV neutron generator is in development at the Institute of Energy Research-Safety Research and Reactor Technology, Forschungszentrum Jülich, Germany. In the first step, a PGNAA-system consisting of a 14 MeV neutron generator for sample irradiation, an HPGe detector for prompt-gamma ray measurement, and a graphite irradiation chamber was designed for the investigation of large samples with a maximal volume of 50 L.

In this paper, the prompt γ-ray count rate and the average thermal neutron flux in the sample for the PGNAA-system was studied with a large concrete sample and with a homogeneous large sample made of polyethylene (PE) and metal with various H and Cd contents using the MCNP-5 code. To determine the average thermal neutron flux in large samples of various compositions, a new method was proposed using the prompt-gamma rays at 2.22 MeV produced by thermal neutron activation of hydrogen in the sample, and at 4.44 MeV and 4.95 MeV produced by fast and thermal neutron activation of carbon in the graphite chamber and in the sample, respectively.

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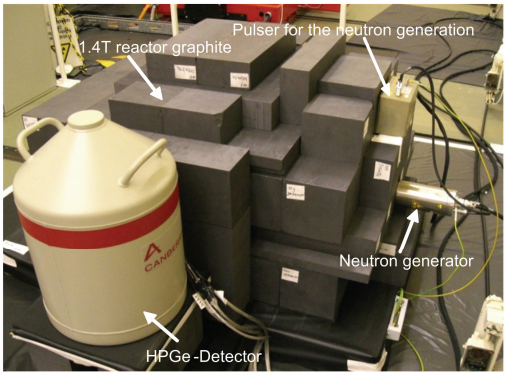

Fig. 1 Side views of the PGNAA-system in the experimental hall with the HPGe-detector in front, the neutron generator on the right side and the graphite irradiation chamber.

2 MCNP5 modeling

The PGNAA-system was modeled according to the experimental setup (Fig. 1). The investigated drum was placed in an irradiation chamber made exclusively of graphite as neutron moderator and reflector. Out of radioprotection purpose, the chamber wall was about 40 cm thick, and the inner volume was 40 cm × 40 cm × 50 cm. Hydrogenous materials like PE or paraffin were prohibited in the irradiation chamber, so as to determine hydrogen content of the drum matrix using the 2.22 MeV γ-ray. The drum was irradiated with 14 MeV neutrons from a D-T neutron generator (GENIE 16GT, EADS SODERN). The deuterium beams irradiate the tritium target located at mid-height of the drum, and a 35-cm distance was kept from its centre. The prompt γ-rays were detected with an n-type HPGe detector (Canberra). The detector was surrounded by ^{6}LiF plates to avoid thermal neutron capture in the Ge crystal. The distance between the detector window and the drum centre was 35 cm so that a 50 L drum was fully analyzed.

In the MCNP model, the 14 MeV neutron source was defined as a disc source emitting 10^{8} n·s^{-1}(4π), and the HPGe detector was described as a 8×8 cylinder of natural germanium. To be like with a real HPGe detector, track lengths of the γ-rays were simulated using the so called "pulse height tally" and the Gaussian Energy Broadening function (GEB), which is related to energy resolution of the physical detector. The 50 L steel drum with 1.5 mm wall was filled with dry concrete or a mixture of PE and metal.

Contents of the dry concrete were: O, 51.6%; Si, 37%; Ca, 8.9%; Al, 0.85%; K, 0.7%; Mn, Mg, 0.57%; Fe, 0.45%; H, 0.41%; Mn, 0.0294%; and Cl, 0.0097%. It was about 115 kg, with a density of 2.35 g·cm^{-3}. In order to simulate the samples with higher H content, the silicon content was replaced by hydrogen. Composition of the PE and metal mixture was: O, 24.7 α%; Na, 5.9 α%; Al, 11.8 α%; Si, 28.2α%; Ca, 11.8 α%; Fe, 11.8 α%; and Cu, 5.9 α%, where α = (1–C_{PE}/100) and C_{PE} is the PE content. For polyethylene, the carbon content changes with hydrogen content at a mass ratio of 6/1. And the mixture of PE and metal was 115 kg, with an average density of 2.3 g·cm^{-3}.

In the simulations, Cd content of the samples increased from 0 to 1000 ppm, and hydrogen content from 0 to 4%. Each simulation calculated 10^{9} particles, for a high statistical precision in terms of relative error and variance of variance (vov), within an accuracy interval of 0.2% (1σ).

3 Prompt gamma ray count rates

As an example, the prompt gamma spectrum (Fig. 2) was obtained by MCNP5 with the concrete sample of 500-ppm cadmium. A large number of prompt γ-rays were observed from the fast and thermal neutron activation of the sample, the graphite chamber and the steel drum. The prompt γ-rays at 2.22 MeV produced by thermal neutron activation of hydrogen in the sample, and at 4.44 MeV and 4.95 MeV produced by fast and thermal neutron activation of carbon in the graphite chamber and in the sample were labeled. They were used to monitor the average thermal neutron flux of the samples. The major prompt γ-ray of Cd at 0.558 MeV was also marked in the figure.

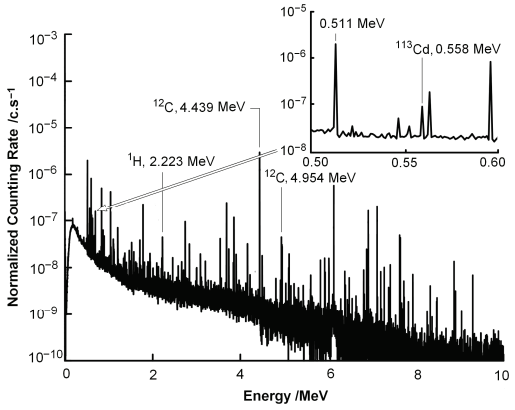

Fig. 2 Simulated prompt gamma spectrum of concrete sample (115 kg) of 500 ppm cadmium. The simulation was performed by MCNP5 with 10^{8} n·s^{-1} of 14 MeV neutrons, which was normalized to 1.

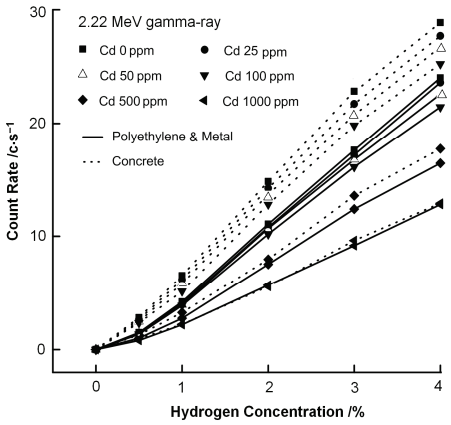

Fig. 3 Count rate of the prompt γ-ray at 2.22 MeV calculated by MCNP5 with thermal neutron activation of H in the sample, as function of the H and Cd concentration in the two materials.

The count rate at 2.22 MeV with the H and Cd in the two samples is shown in Fig. 3. Under given Cd concentration, the count rate at 2.22 MeV increases with the H concentration, which acted as fast neutron moderator, hence an increase of the average thermal neutron flux. The concrete sample had higher counts than the PE and metal mixture, reflecting a difference in the absorption of thermal neutrons. Given the H concentration, the count rate at 2.22 MeV decreased with the Cd concentration due to the absorption of its thermal neutrons. This was more remarkable in the concrete sample than in the mixture of PE and metal because of the composition difference, and the higher the Cd concentration, and the lower the difference for the two samples. Thus, the count rate at 2.22 MeV may be used to monitor the average thermal neutron flux in a specific sample.

The dependence of the count rate at 4.44 MeV on the H and Cd contents in the two samples is shown in Fig. 4. The value, which was independent of the Cd content, *i.e.* the thermal neutron absorption, only depends on moderation properties of the sample, and decreased linearly with the H concentration. In the mixture of the samples, however, a break of the linearity was observed due to containing 3% to 4% hydrogen except for a large amount of polyethylene. Given the H concentration, the count rate was lower in the concrete sample than in the mixed samples, as the former is a better absorber of fast neutrons than the latter. The macroscopic neutron cross section of 14 MeV neutrons was 0.115 cm^{-1} for the concrete sample, and was 0.0827 cm^{-1} for the mixture without polyethylene, and that was 0.0964 cm^{-1} for the mixture of 24% PE (4% H). Thus the count rate at 4.44 MeV may be used to monitor the moderation and absorption of fast neutrons in the sample.

The count rate at 4.95 MeV varied with the H and Cd contents in the two samples (Fig. 5). The behavior was much more complex than at 2.22 MeV and 4.44 MeV. Given the Cd concentration, the count rate increased for a lower than 1% hydrogen, and reached a constant for hydrogen concentrations of 1% to 2% in the concrete sample, and for that of 1% to 3% in the PE and metal mixture. At the highest H concentrations, the count rate decreased in the concrete sample due to its thermal neutron absorption, and increased in the mixed samples due to the large amount of the carbon. Given the H concentration, the count rate at 4.95 MeV decreased with increasing Cd concentrations due to its thermal neutron absorption, which reflected the moderation of fast neutrons and the absorption of thermal neutrons in the sample.

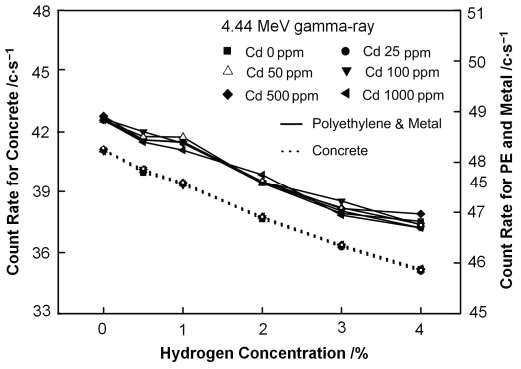

Fig. 4 Count rate of the prompt γ-ray at 4.44 MeV calculated by MCNP5 with fast neutron activation of carbon (graphite cell), as a function of the H and Cd contents in the two materials.

The count rate at 0.558 MeV depended on the H and Cd concentration in the two samples (Fig. 6). Given the Cd concentration, the rate increased non-linearly for a lower than 2 % hydrogen in the concrete sample, and lower than 3% hydrogen in the mixed samples. For high hydrogen and lower cadmium than 500 ppm, the rate decreased due to the thermal neutron absorption by hydrogen. For higher cadmium than 500 ppm, the rate increased. Given the H content, the rate increased with the Cd concentration and reached a saturation value due to the absorption of thermal neutrons by cadmium.

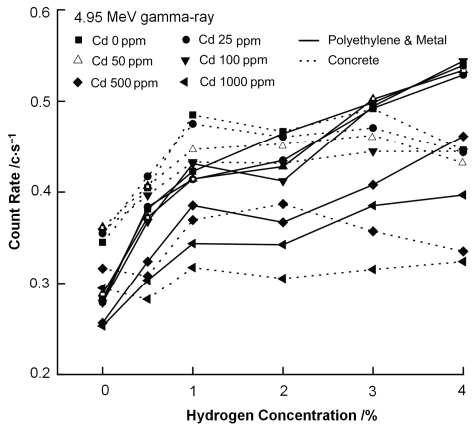

Fig. 5 Count rate of the prompt γ-ray at 4.95 MeV calculated by MCNP5 with thermal neutron activation of carbon(graphite cell) as a function of the H and Cd contents in the two materials.

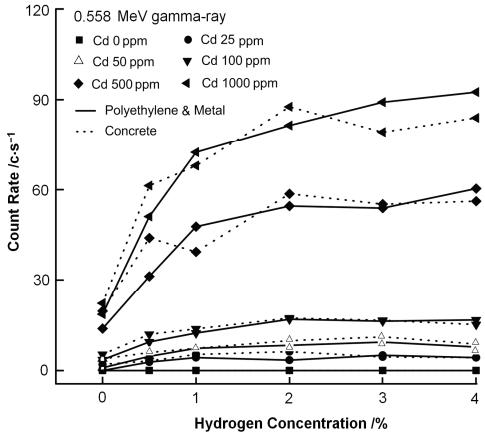

Fig. 6 Count rate of the prompt γ-ray at 0.558 MeV calculated by MCNP5 with thermal neutron activation of cadmium in the sample as a function of the H and Cd concentration in the two materials.

4 Thermal neutron flux

Figure 7 shows dependence of the average thermal neutron flux on the H and Cd concentration in the two samples. Given the Cd concentration, the average thermal neutron flux increased non-linearly with the H concentration, whereas it decreased non-linearly with increasing Cd concentration under given H content.

In order to find a general analytical expression of the average thermal neutron flux based on the count rates, the average thermal neutron flux, Φ_{th}, was calculated by MCNP5 without cadmium in the two materials, *i.e.* the strong thermal neutrons absorber was first fitted by function of the prompt γ-ray count rate of hydrogen at 2.22 MeV, Z_H, using Eq.(1),

$$\Phi = \Phi_0 + \Phi_{max} Z_H / (C + Z_H) \quad (1)$$

where, Φ_0 was the average thermal neutron flux without hydrogen, Φ_{max} was its maximum value, and C was sample composition dependent corrective factor after taking into account the moderation of the fast neutrons. In the concrete sample, fitting the Φ should lead to Φ_0=3138 ± 355 n·cm^{-2}·s^{-1}, Φ_{max}=15063 ± 488 n·cm^{-2}·s^{-1}, and C=4.11± 0.49 counts·s^{-1} with a regression coefficient of 0.998. In the PE and metal mixture, Φ_0 = 2173 ± 242 n·cm^{-2}·s^{-1}, Φ_{max}=15352 ± 547 n·cm^{-2}·s^{-1}, and C = 7.44 ± 0.49 counts·s^{-1} with a regression coefficient of 0.999. The Φ_0 values are in good agreement with of that of the MCNP5 calculation (3162 and 2021 n·cm^{-2}·s^{-1}). Taking into account the errors, the two materials have the same Φ_{max}, averaged at 15207 ± 204 n·cm^{-2}·s^{-1}, which was used for the following calculation.

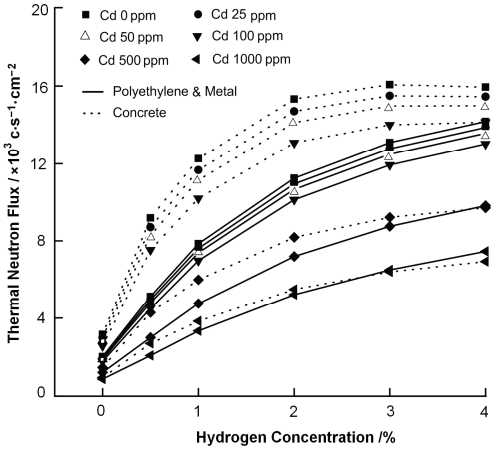

Fig. 7 Average thermal neutron flux calculated by MCNP-5 varied as function of the hydrogen and cadmium concentration for the two materials.

In a next step, a factor taking into account the relative effect of the concrete sample on the moderation of the fast neutrons was defined using the prompt γ-ray count rate at 4.44 MeV, $Z_{C,fast}$, which was produced by the fast neutron activation of carbon as,

$$\alpha = Z_{C,fast} / Z^{ref}_{C,fast} \quad (2)$$

where, $Z^{ref}_{C,fast}$ is the average count rate at 4.44 MeV in the concrete samples without cadmium, to be free from Cd influence on fast neutron moderation (Fig. 4). The count rate at 2.22 MeV, Z_H, can be calculated by:

$$Z^{ref}_{C,fast} = a_0 + a_1 Z_H + a Z_H^2 \quad (3)$$

with a_0= 41.08 ± 0.06, a_1= –183.88 ± 8.55 and a_2 = 889.77 ± 41.37 the coefficients of the fit with a regression coefficient of 0.999.

A factor considering the relative influence of the concrete sample on the absorption of the thermal neutron without cadmium was derived from the prompt gamma-ray count rate at 4.95 MeV, $Z_{C,thermal}$, which was produced by the thermal neutron activation of carbon as follows:

$$\beta = Z_{C,thermal}/Z^{ref}_{C,thermal} \quad (4)$$

where, $Z_{C,thermal} = 0.453 \pm 0.035$ counts·s^{-1} is the mean count rate at 4.95 MeV in the cadmium-free concrete samples.

Finally, the average thermal neutron flux, Φ_{th}, can be calculated for any samples with the lower than 4% hydrogen, and with the lower than 1000 ppm cadmium, as well as other elements, which showed a strong absorption of thermal neutron using the following semi empirical formula:

$$\Phi_{th} = \alpha \beta^2 [\alpha^2 \Phi_0 + \Phi_{max} Z_H/(\alpha^{5/2} C + Z_H)] \quad (5)$$

where, $\Phi_0 = 3138 \pm 355$ n·cm^{-2}·s^{-1}, $\Phi_{max} = 15207 \pm 204$ n·cm^{-2}·s^{-1}, and $C = 4.11 \pm 0.49$ counts·s^{-1}, obtained from the analysis of the average thermal neutron flux without Cd in the concrete sample. The values of Φ_{th} obtained by Eq. (5) were compared with that of the MCNP5 calculation, both were in good agreement within a relative error of 16% (Fig.8), and the deviation for the two data sets was $-11 \pm 23\%$.

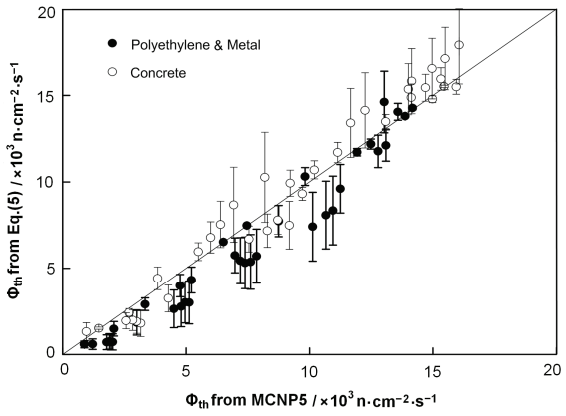

Fig.8 Comparison of the average thermal neutron flux calculated by mean of Eq.(5) by MCNP5 for the two materials.

According to the PGNAA system, therefore, Eq.(5) led to a fairly good estimation for the average thermal neutron flux in a homogeneous large sample of 50 L, irradiated by a 14 MeV neutron source emitting the 10^8 n·s^{-1} (4π). Any modification of the PGNAA system and a change of the sample volume should impact on the prompt gamma-ray count rates, and was necessary for a new parameterization.

5 Conclusion

In this work, the demonstration that the average thermal neutron flux in large samples irradiated by 14 MeV neutrons may be estimated from the prompt gamma-rays produced by the thermal neutron activation of hydrogen in the sample, and by the thermal and fast neutron activation of carbon in the sample and the graphite irradiation chamber has been proved based on the simulation by MCNP-5. The analytical expression to calculate the average thermal neutron flux is specific to the PGNAA-system described in this work *i.e.* to the amount and thickness of graphite surrounding the sample and to the relative position of the gamma-ray detection system to the 14 MeV neutron source.

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