A Monte Carlo simulation and setup optimization of output efficiency to PGNAA thermal neutron using ^{252}Cf neutrons *

ZHANG Jin-Zhao(张金钊)[1) TUO Xian-Guo(庹先国)

Applied Nuclear Techniques in Geoscience Key Laboratory, Chengdu University of Technology, Chengdu 610059, China

Abstract: We present the design and optimization of a prompt γ-ray neutron activation analysis (PGNAA) thermal neutron output setup based on Monte Carlo simulations using MCNP5 computer code. In these simulations, the moderator materials, reflective materials, and structure of the PGNAA ^{252}Cf neutrons of thermal neutron output setup are optimized. The simulation results reveal that the thin layer paraffin and the thick layer of heavy water moderating effect work best for the ^{252}Cf neutron spectrum. Our new design shows a significantly improved performance of the thermal neutron flux and flux rate, that are increased by 3.02 times and 3.27 times, respectively, compared with the conventional neutron source design.

Key words: PGNAA, neutron source, thermal neutron, moderation, reflection

PACS: 21.60.Ka **DOI:** 10.1088/1674-1137/38/7/078201

1 Introduction

Prompt gamma ray neutron activation analysis (PGNAA) is a rapid, nondestructive, and powerful multi-elemental technique to analyze large samples of some minor, trace light elements, and it is used in industrial applications [1–5]. In a PGNAA process the nuclear composition of the sample is determined from the prompt gamma rays that are produced through neutron inelastic scattering and thermal neutron capture. Since the inelastic scattering cross section is small, the PGNAA design is based on thermal neutron capture process. Thus, an efficient PGNAA setup requires a high thermal neutron flux available at the sample, along with a high thermal neutron production ratio of neutron source, which is the research focus in the development of PGNAA technique.

Accelerator neutrons, source reactor neutron source, Am-Be neutron source and ^{252}Cf neutron source can be used as PGNAA neutron source [6]. ^{252}Cf neutron source being low-cost and with a high neutron flux density is commonly used in PGNAA setup. However, the ^{252}Cf source is an isotropic neutron source that results in less sample thermal neutrons. Thus, it gives a low utilization rate of the neutron source. In the present study, Monte Carlo simulations were carried out for the design of a ^{252}Cf neutron source moderation setup that was used for the analysis of cement samples [7]. The model of Monte Carlo simulation was verified by performing experiments that were described previously [8, 9]. We demonstrate the improvement of the thermal neutron source yield rate of ^{252}Cf neutron by employing the PGNAA neutron source structure in the design. The calculation results for the new design are compared with the previous reports. Here, we present the results of the thermal neutron flux rate, fast neutron flux rate, and gamma rays yield.

2 The model for Monte Carlo simulations

The two designs of the neutrons moderation setup used in this study consists of a NaI detector of gamma rays, a cylindrical polyethylene sample compartment and a neutron source. The internal and external diameters of the cylindrical polyethylene sample compartment are 20 cm and 30 cm, respectively, along with a height of 20 cm. The neutron source of the first design is wrapped only by a moderator, whereas the second design includes a moderator and a reflector. The reflector is placed below the neutron source and the hemispheric moderator is placed on the upper part of the neutron source. The goal of such a design is to enable the reflection of the neutrons back to the upper part of the lower end of the moderator in order to improve the utilization ratio of neutrons. This is important because different materials have different neutron reflectivity. Fig. 1(a) is a schematic design of the previous PGNAA source-moderator setup that was reported previously [7]. Fig. 1(b) is a schematic illustration of our new design that we present here. We opti-

Received 5 September 2013

* Supported by National Natural Science Foundation of China (41274109, 41025015, 41374112, 11105132, 41104118)

1) E-mail: zhangjinzhao_cdut@163.com

mized the design for best performance by Monte Carlo simulation analysis.

We have used a ^{252}Cf spontaneous fission neutron source for the simulation, where the neutron energy spectrum represents a fission spectrum that can be described by Watt distribution. The formula is given below [10]:

$$f(E)=c\mathrm{e}^{-\frac{E}{a}}\sinh\left(\sqrt{bE}\right)a, \qquad (1)$$

E is the neutron energy, a=1.025, b=1.25, c=0.365. The fission neutron energy spectrum of ^{252}Cf can be obtained by MCNP5 simulation.

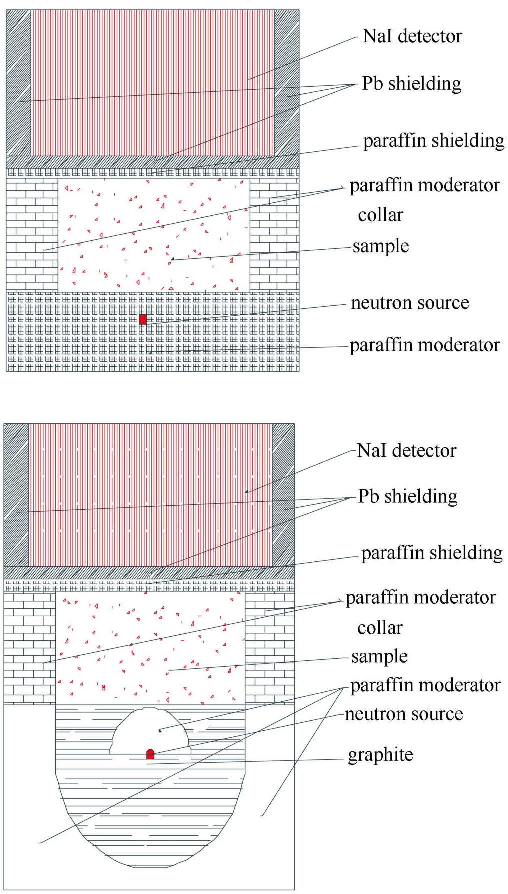

Fig. 1. (color online) A schematic illustration of the previous and new PGNAA source-moderator setup presented in this study.

This study is based on the previous reports [7, 11] of Portland cement composition, which can be considered as a sample for our simulation calculations.

2.1 Moderator material Monte Carlo simulation

PGNAA analysis accuracy depends on irradiation, neutron fluence rate, and neutron capture cross section of nuclides. Radiative capture cross section is the main component for heavy nuclei. In the case of low energy neutrons, the entire cross-section is almost equal to the radioactive capture cross section, which is equal to l/v. Thus, it is important to increase the sample thermal neutron fluence rate while reducing the fast neutron fluence rate in the PGNAA analysis. Therefore, we not only used a moderator for neutrons to improve the thermal neutron fluence rate, but also paid attention to the material used in moderation to decrease the γ background. Hence, we chose five materials (i.e. light water, heavy water, polyethylene, graphite and paraffin) to analyze and compare the results in order to employ the best material. Figs. 2–5 present the Monte Carlo calculation results for these materials.

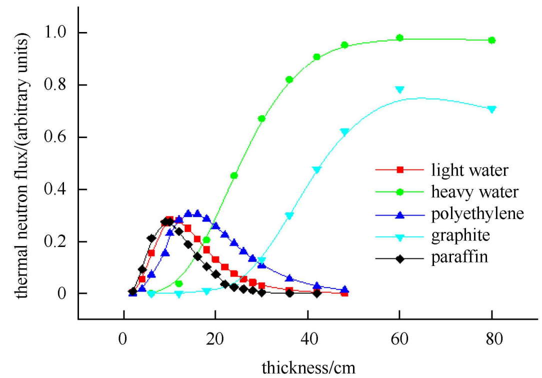

Fig. 2. (color online) Thermal neutron flux of five moderator materials increases with the thickness.

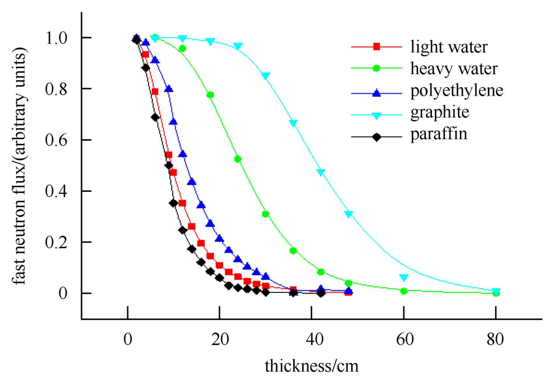

Fig. 3. (color online) Fast neutron flux of five moderate materials increases with the thickness.

As shown in Figs. 2–5, we observe that the thermal neutrons increase rapidly with increasing light water, paraffin and polyethylene with thickness in the range of 9–12 cm and it reaches up to the maximum. However, when employing the moderator, increasing the thickness leads to a decrease of thermal neutrons because a large number of thermal neutrons are absorbed. With heavy water and graphite of thickness 48 cm, the thermal neutron flux reaches up to the maximum. We note that the loss of thermal neutrons is less with an increase in the thickness of the moderator.

For fast neutrons, we observe that the flux of five substances is reduced with the increase in thickness of the moderator substance in the chronological order of paraffin, light water, polyethylene, heavy water and graphite.

We also observe that in a neutron moderator processing, paraffin, light water and polyethylene produce gamma rays, the yields get maximum and reduce subsequently with the increase of the thickness to 22 cm. It is to be noted that graphite and heavy water do not substantially generate γ-rays.

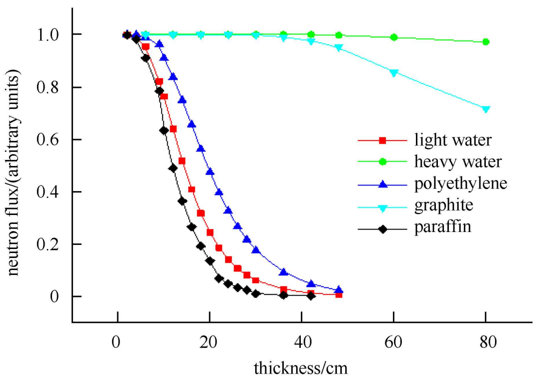

Fig. 4. (color online) Neutron flux of five moderator materials increases with the thickness.

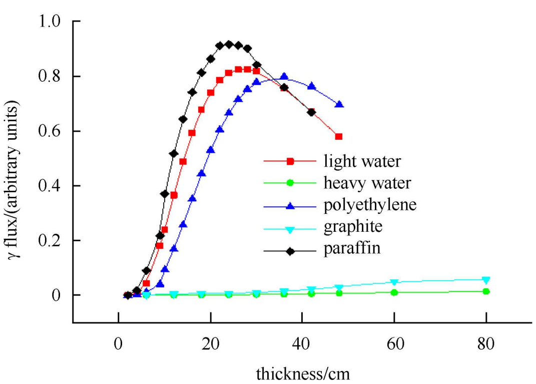

Fig. 5. (color online) γ flux of five moderator materials increases with the thickness.

2.2 Reflective material Monte Carlo simulation

The reflection is determined by the scattering cross section of nuclide, the absorption cross section, surface density of atoms and the thickness of the reflector. We note that the higher scattering cross section results in a smaller absorption cross section, and the higher surface atom density produces a greater thickness of the reflector and a stronger reflection. Figs. 6, 7 show the Monte Carlo calculation results of light water, heavy water, polyethylene, graphite, beryllium and paraffin.

One can see from Figs. 6, 7 that beryllium is the best material for reflective effect on a thin layer followed by paraffin, heavy water, and graphite. Paraffin absorbs neutrons and produces γ-rays. Beryllium, heavy water and graphite on the other hand, produce an extremely low γ-ray flux.

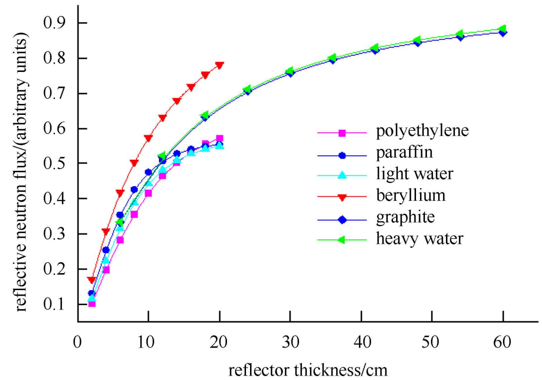

Fig. 6. (color online) The reflective neutron flux with different thicknesses of five kinds of reflective materials.

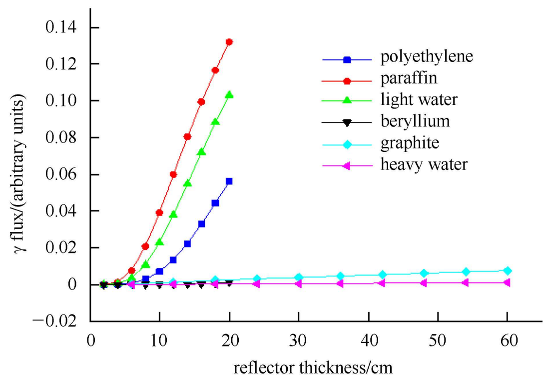

Fig. 7. (color online) The γ flux of different materials and thicknesses.

3 Neutron source setup optimization

Our design for a new neutron source is based on the various considerations of reflection and moderating effects as discussed above. Since the thin beryllium reflector works best, we use a hemispherical wrapped ^{252}Cf fission neutron source for the lower part. As we noted earlier, thick layers of heavy water and graphite have a similar reflectivity and the γ-ray yield is lower than paraffin. However, heavy water is difficult to use in practical applications. Hence, we use a thick layer of graphite as the design device reflector, which is shaped into an hemisphere with a radius of 20 cm and which ensured the reflection to be a total reflection.

The upper part of the neutron source is wrapped by paraffin. Because of the isotropic nature of the neutron source, we consider the geometry of moderation to be a hemispherical. Paraffin is surrounded by graphite. This design is superior compared with the previous design, which uses the parallel plate institutions, because our design not only ensures the full moderation of the neutrons but also significantly reduces the thermal neutron absorption in the moderator material.

The beryllium reflector is optimized through thermal neutrons flux rate of the sample. The calculated flux rate of the sample is plotted in Fig. 10.

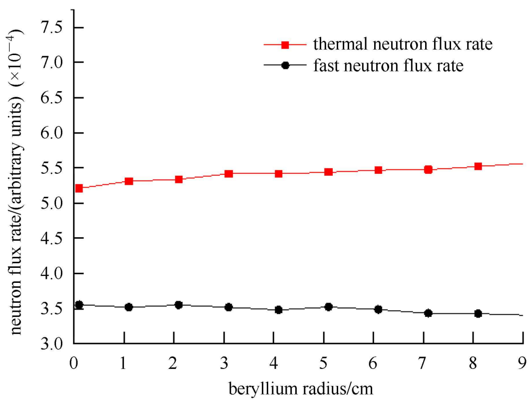

Fig. 8. (color online) Thermal neutrons and fast neutrons flux rate of beryllium with different radius.

It is clear from Fig. 10 that with an increase in the thickness of beryllium, the thermal neutron flux increases while the fast neutron flux decreases. However, we note that the changes in thermal and fast neutron flux are observed to be small. Hence, we chose the reflector to be entirely made of graphite because beryllium did not produce any notable changes in the neutron flux, besides also being quite expensive. The thermal and fast neutron calculated yields as a function of front moderator paraffin radius are shown in Fig. 11. We observe that, with an increase in the paraffin radius, the thermal neutron flux increases while fast neutron flux decreases. The thermal neutron flux rate initially increased with the moderator radius with a maximum value achieved for 6–7 cm radius moderator, followed by a subsequent decrease in the thermal neutron flux rate. The initial increase in the thermal neutrons yield may be due to the neutrons scattering cross section, which is greater than the absorption cross section. The subsequent decrease in thermal neutron yield with further increase in the moderator radius may be attributed to the absorption cross section. The same reason leads to the fast neutrons flux rate reducing linearly with the increase of the paraffin radius. It can be seen in Fig. 11 that the moderating effect of the neutron source slowing-down device works best with a paraffin radius of 7 cm and a graphite reflector.

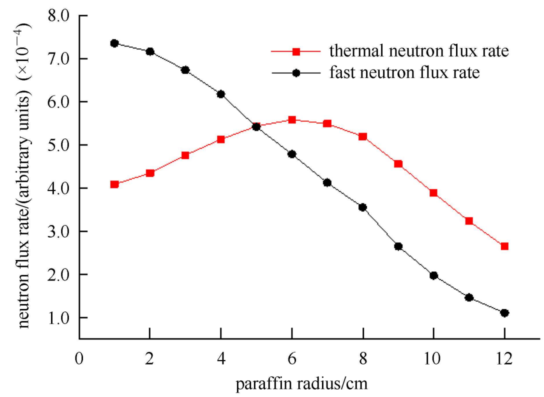

Fig. 9. (color online) Thermal neutrons and fast neutrons flux rate of different radius of moderation.

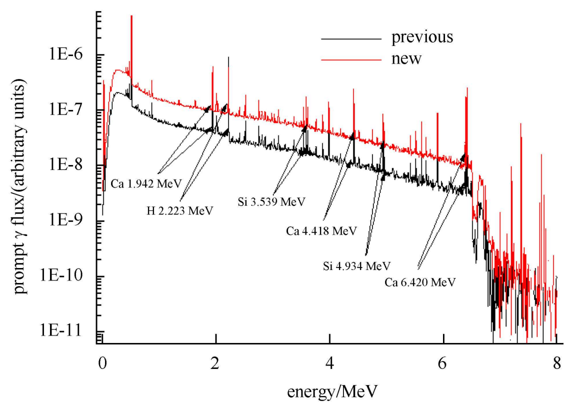

Fig. 10. (color online) New and previous designs for gamma rays yield from prompt γ-ray neutron activation.

As shown in Table 1, we demonstrate that the amounts of the thermal neutrons output and fast neutrons show a significant increase in the present design, which consists of a neutron source moderated device, when compared with the Saudi device [7]. We observe that the thermal neutrons output grows by 3.02 times together with the fast neutrons output that grows by 3.27 times. However, we notice that the percentage of fast neutron ratio increased to a lesser degree. Furthermore, since it is desired to reduce the proportion of fast neutrons, reducing neutron inelastic collisions may be useful

Table 1. The results of two designs compared with the absence of sample.

	reflection structure	conventional structure	times
thermal flux rate	5.49555E-4	1.81875E-04	3.02E+00
fast neutron flux rate	4.12318E-4	1.26135E-04	3.27E+00
γ ray flux rate	1.03028E-04	3.18849E-05	3.23E+00
the rate of thermal, fast neutron output flux	1.33E+00	1.44E+00	9.24E-01

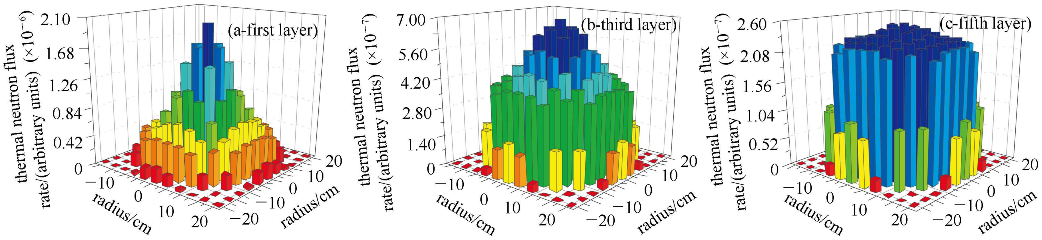

Fig. 11. Previous design of thermal neutron flux rate of the layer.

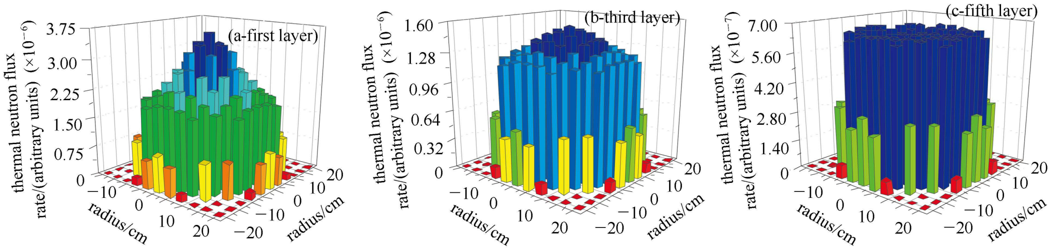

Fig. 12. (color online) New design of thermal neutron flux rate of the layer.

and this leads to nuclides produced by γ-rays, where γ-rays minimize the background. On the other hand, an increase in the proportion of thermal neutrons improves the thermal neutron capture spectra of the sample measuring the spectral contribution of γ-rays, which eventually results in simplified lines and improves analysis accuracy. The calculated yield of the gamma rays from prompt γ-ray neutron activation is plotted in Fig. 12, which illustrates how the new structure can be leveraged to improve the prompt γ count rate that also simultaneously does not result in complicated excitation spectrum. We note that the same neutron source improves the utilization rate of the neutron with the new structure.

The samples are divided into 4 mm×4 mm×4 cm grids. Figs. 11 and 12 show the thermal neutron distributions of the two designs. The new design not only provides a significantly improved thermal neutron flux structure but also results in a more uniform distribution of thermal neutrons in the sample. This means that the activation of the sample thermal neutrons is uniform, which is more conducive to the analysis of large and heterogeneous samples.

4 Conclusion

Monte Carlo simulations have been carried out to design a PGNAA thermal neutron output setup to increase the thermal neutron output efficiency. The new design of PGNAA moderation neutron source device produces 3.02 times higher flux of thermal neutrons and 3.27 times higher neutron flux rate when compared with the previously reported device. We envision that our new design of PGNAA offers significant potential to improve the efficient utilization of the fission neutron source.

The authors wish to acknowledge the support of the Applied Nuclear Techniques in Geoscience Key and State Key Laboratory of Geohazard Prevention & Geoenvironmental Protection, Chengdu University of Technology.

References

1 Gardner R P, Guo P, WANG Y Y et al. Applied Radiation and Isotopes, 1997, **48**(10–12): 1355–1372
2 Borsaru M, Berry M, Biggs M et al. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 2004, **213**: 530–534
3 Naqvi A A, Nagadi M M, Al-Amoudi O S B. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 2006, **569**(3): 803–809
4 Naqvi A A, Garwan M A, Nagadi M M et al. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 2009, **607**(2): 446–450
5 Oliveira C, Salgado J, Gon C C Alves I F et al. Applied radiation and isotopes, 1997, **48**(10): 1349–1354
6 Ghassoun J, Mostacci D. Applied Radiation and Isotopes, 2011, **69**(8): 1138–1142
7 Naqvi A A, Abdelmonem M S, Al-Misned G et al. Applied Radiation and Isotopes, 2006, **64**(12): 1631–1636
8 Oliveira C, Salgado J. Applied Radiation and Isotopes, 1998, **49**(12): 1685–1689
9 Naqvi A A. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 2003, **511**(3): 400–407
10 Team M C. Book MCNP-A General Monte Carlo N-Particle Transport Code Version. 2003, 5
11 Al-Jarallah M I, Naqvi A A, Fazal-Ur-Rehman et al. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 2002, **195**(3–4): 435–441