Shielding studies on a total-body neutron activation facility

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Background: Prompt gamma neutron activation analysis (PGNAA) is known as a non-invasive technique capable of measuring elemental concentration in voluminous samples in a short period of time. Also it is a valuable diagnostic tool for total body elemental measurements. 252Cf and 241Am-Be sources which are usually used in this method, generate not only neutrons, but also emit high-energy and unwanted gamma-rays. Because the patient must be located against the neutron source, patient dose during an analysis is an important concern when using this technique. Materials and Methods: Gamma-rays were attenuated without losing the neutron flux or significant alteration in the neutron spectrum. A relatively safe body chemical composition analyzer was designed with an optimal spherical gamma-ray shield, enclosed to the neutron source. Effects of gamma-ray shielding and optimum radius of spherical Pb shield was investigated and compared with the unfiltered bare source, using MCNP4C code. Then, the gamma ray dose equivalent per source neutron rate (user defined parameter) in the soft tissue is calculated for different radiuses of spherical Pb shield. for both neutron sources. Results: A decreasing flux of gamma-ray was observed when the radius of the spherical Pb shields increased. The value of this reduction was about 94% for ²⁵²Cf source when a lead spherical shield of radius 4 cm was used; while the reduction was about 50% for ²⁴¹Am-Be source with the same spherical shield. For a spherical Pb shield of radius 4 cm, reduction of the gamma dose equivalent per source neutron rate was about 8.44×10-17 Sv when the neutron source was ²⁵²Cf and about 1.24×10⁻¹⁶ Sv when the neutron source was 241 Am-Be. Conclusion: Results show using optimum gamma-ray shield geometry can reduce the patient absorbed dose per incident neutron in a body chemical composition analyzer. Iran. J. Radiat. Res., 2007; 5 (1): 45-51

Keywords: PGNAA, spherical shield, neutron source, MCNP, absorbed dose, dose equivalent.

INTRODUCTION

Determination of elemental compositions for a human body is one of the useful tools for understanding general physiology relationships, diagnosing some disease and cancers, radiotherapy treatments, nutritional disorders, planning treatments for those same diseases and disorders and in quantifying the efficacy of some medicinal therapies. Therefore, a lot of techniques have been used to determine the precise amount of body chemical compositions. Use of neutrons is an ideal approach for conducting these measurements which can be used in three different forms; fast neutron analysis (FNA), prompt gamma neutron activation analysis (PGNAA), and delayed gamma neutron activation analysis (NAA).

Prompt neutron activation gamma analysis technique is a powerful elemental analysis method (1). PGNAA has a wide range of applications, such as bulk material analysis and oil well logging, hidden or buried explosive materials detection (2-4). This method is based on the simple excitation and de-excitation of atom by absorbing a neutron followed by emitting a high-energy (multi-MeV) prompt gamma ray. There are several prompt gamma rays with various intensions for a special active element (5). Ordinarily, the most intensive gamma ray in the spectrum is a characteristic sign for each element. So a gamma spectrum of an active sample can be assumed as a footprint of the existence of special elements in the sample. Since most of nuclei show an increasing in the neutron absorption cross section as the neutron energy decreases, PGNAA is a useful technique when using neutrons with low or thermal energy.

Radioisotope ²⁵²Cf and ²⁴¹Am-Be are neutron sources used for many applications in which compact, portable and reliable neutron sources are required. They are commonly used in the PGNAA method. They

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Fax: +98 511 879 64 16 E-mail: vejdani_ir@yahoo.com provide a high flux and reliable neutron spectrum from a very small assembly. Unfortunately, these neutron sources not only generate neutrons but also high-intensive gamma-rays, 4.438 MeV for ²⁴¹Am-Be and a continuous spectrum for ²⁵²Cf from 0 to 10 MeV.

When using PGNAA method for medical purposes, the sample is the soft tissues of a human body and the gamma-rays can have destructive effects on it. Moreover, the accumulations of these high-energy gamma-rays in the detector volume eventuate simultaneous pulses which can be piled up and distort spectra in the region of interest (ROI). These limitations led us to attenuate these gamma-rays in a reasonable way without losing neutron flux in the vicinity of the sample or production of high-energy and high-rate prompt gamma-ray via neutron interaction by the shield material.

According to the latest recommendations of international bodies, an increasing attention must be paid to the patient protection during cancer radiotherapy and exclusive applications of neutron sources such as ²⁵²Cf and ²⁴¹Am-Be. Therefore the primary purpose of this work was to design a relatively safe body chemical composition analyzer to be used in cancer therapy. In this way a new approach that is employing a proper shield for gamma-rays, and neutrons installed between the sample and neutron source was developed and its effects were evaluated. Since lead (Pb) is a

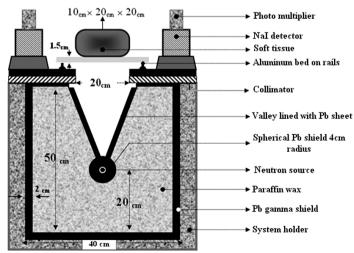
safe and proper gamma shield, a spherical Pb shield (enclosing neutron source) was chosen and modeled in the calculations. Effects of using this gamma-ray shield on the neutron flux and neutron energy spectrum was also evaluated. At the end, this shield in a usual body chemical composition analyzer was applied and consequent results were calculated. To confirm efficacy of this shield, the absorbed dose was measured in a soft tissue phantom, in the presence and absence of the gamma-ray shield. The Monte Carlo N-particle general code optimization studies in PGNAA was used for comprehensive simulation.

MATERIALS AND METHODS

Configuration of the PGNAA facility

To protect personnel from biological effects of neutrons and to reduce background counts, neutron shielding must be considered, but high-speed neutrons are more difficult to shield, because absorption cross sections are much lower at higher energies. So, at first, neutrons must be moderated in a hydrogenous material such as paraffin wax (14.86 % H, 85.14 % C). Since hydrogen has a great absorption cross section for thermal neutrons, the risk of neutrons for personnel vanishes. Although in the capture process (n, γ) by the hydrogen target, potentially hazardous gamma-rays (2.224 MeV) will also be produced. However, it is possible to cover the moderator with a thin layer of Pb or tungsten to filter secondary gamma-rays.

Figure 1 shows a cross-sectional view of the improved body chemical composition analyzer designed at the Ferdowsi University of Mashhad Radiation Research and Measurement Laboratory. Sheets of 2 cm thickness of lead surrounded the paraffin wax to provide radiation shielding for personnel. This setup was an improved one which has originally been installed at Monash Medical Centre (MMC), Melbourne, Australia.



(MCNP) (6) which is usually employed for Figure 1. Schematic diagram of the body chemical composition analyzer.

A sphere of lead with optimum radius is centered at the source position to reduce gamma-ray component of the neutron source. Another part of this configuration is an invert, rectangular, cuneus void cast within a paraffin wax block $(40_{\rm cm} \times 50_{\rm cm} \times 60_{\rm cm})$. To protect patients' body from 2.224 MeV gamma-rays, produced by hydrogenous moderator, the inner wall of the valley-shaped (figure 1) was lined by Pb sheet with a thickness of 2 cm. Then, a rectangular neutron-beam aperture measuring the length of 40 cm (perpendicular to the paper sheet) and 20 cm (width) at the bed level is defined.

The patient is positioned supine, 40 cm above the neutron source, on an aluminum bed which moves directly over the neutron source and through the upwardly collimated neutron beam. Since the final results of the absorbed dose will be similar for most parts of human body and for more simplicity, the absorbed dose in a soft tissue equivalent (TE) $10_{\rm cm} \times 20_{\rm cm} \times 20_{\rm cm}$, of density 1 gm/cm³ (table 1) (7), was evaluated by MCNP code.

 Table 1. Chemical composition of a soft tissue.

Element	Fraction by Weight	
Hydrogen	0.10117	
Oxygen	0.76183	
Carbon	0.11100	
Nitrogen	0.02600	

Source modeling

In the simulation of the optimum shielding investigation and body chemical composition analyzer, ²⁴¹Am-Be and ²⁵²Cf neutron sources were positioned at the center of a spherical Pb filter one by one. A ²⁵²Cf neutron source with 1.61Ci of activity contained in standard X.35 capsules format (code CVN353) and an ²⁴¹Am-Be source with 5Ci of activity contained in standard Amershan X.14 capsules format (code AMN24), were used in the PGNAA configuration system. The final results are calculated in a way to be independent from the source activity. The gamma energy spectrum of ²⁵²Cf was simulated by using the

following empirical energy distribution function:

$$SCf_{\gamma}(E_{\gamma}) = \begin{cases} 375E^{2}e^{\frac{-E}{0.109}} + 0.468e^{\frac{-E}{1.457}} \rightarrow E \le 1.5MeV \\ e^{\frac{-E}{0.851}} \rightarrow E > 1.5MeV \end{cases}$$

This function is based on the experimental data reported by Glässel (8).

The fission neutron spectrum SCf_n (E_n) of the ^{252}Cf was simulated by using the Watt fission spectrum using coefficients provided with the MCNP-4C code $^{(6)}$. That is:

$$SCf_n(E_n) = 0.30033e^{\frac{-E_n}{1.025}} \sinh(2.926E_n)^{\frac{1}{2}}$$

Where, E is the neutron energy in MeV.

This equation was used as the input neutron spectrum of ²⁵²Cf source in the calculations.

Figures 2 and 3 show energy spectra of both gamma and neutron particles respectively for the $^{252}\mathrm{Cf}$ neutron source as described above. The multiplicity ratio per fission event, R=S_{\gamma}/S_n, for the $^{252}\mathrm{Cf}$ source was about 2.132 $^{(9)}$. Figure 4 shows neutron energy spectrum from the $^{241}\mathrm{Am}\text{-Be}$ source which is adapted from experimental data reported by Kluge and Weise $^{(10)}$.

Flux and absorbed dose calculations

In this work the calculation of the total gamma dose equivalent rate is divided into three parts.

1. H_{S.7} that is the gamma dose equivalent rate

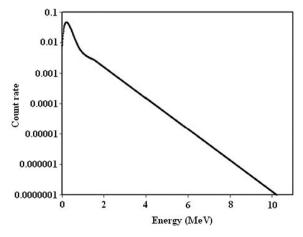


Figure 2. ²⁵²Cf gamma-ray spectrum (per source neutron rate).

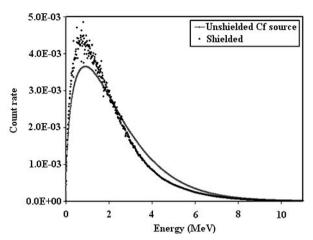


Figure 3. ²⁵²Cf neutron spectrum (per source neutron rate) and the effect of a spherical Pb shield of radius 4 cm on this spectrum.

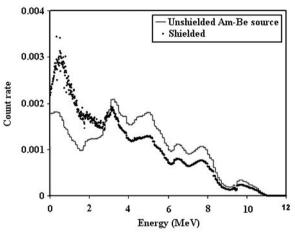


Figure 4. ²⁴¹Am-Be neutron spectrum (per source neutron rate) and the effect of a spherical Pb shield of radius 4 cm on this spectrum.

due to the gamma-ray component of neutron Source, 4.438 MeV for ²⁴¹Am-Be and a continuous spectrum from 0 to 10 MeV (figure 2) for ²⁵²Cf.

- 2. $H_{T,\gamma}$ the gamma dose equivalent rate arising from prompt gamma-rays generated by the interaction between neutrons and filter material, here Pb, as well as the source encapsulation and surrounded materials.
- 3. $H_{F,p}$ the gamma dose equivalent rate due to prompt gamma-rays generated by the interaction between neutrons and the soft tissue.

The total gamma dose equivalent rate in sivert per second (Sv/s), is then $H_{\gamma}=H_{S,\gamma}+H_{T,\gamma}+H_{E,\gamma}$.

In order to be independent from the source activity, a user defined parameter, i.e., gamma dose equivalent per source neutron rate, was calculated. Dose equivalent per source neutron rate is the dose equivalent rate in Sv/s divided by the value of emitted neutrons rate (neutron source activity). When thermal neutrons are captured, in addition to the prompt gamma-ray, residual elements may be active with various half-lives and intensities. In this work the total dose equivalent rate due to delayed gamma-rays were not considered.

For calculations of gamma-ray flux, two separate input files were written. One is to track gamma-rays which were originated from the source position, and the other was to track those prompt gamma-rays which were produced via interaction of neutrons by the source capsule or filter material. The intensity of 4.438 MeV gamma-rays to neutrons, $R=S_{\nu}/S_n$, for the ²⁴¹Am-Be source was about 0.596 (11) and the multiplicity ratio per fission event, R=S₂/S_n, for the ²⁵²Cf source was about 2.132 (8). Final results in the gamma-ray flux calculation were the summation of two parts: 1- Multiplying the gamma-ray flux obtained from tracking the neutron source gamma-ray component by R; and 2-Gamma-ray flux obtained from tracking prompt gamma-rays.

RESULTS

The results are classified into two sections; the first section assesses spherical Pb shield effects and the second section evaluates the effects of employing this spherical shield when used with the body chemical composition analyzer.

1. The total flux of gamma-rays that originate from neutron sources and those were produced in the inelastic or capture process in the source capsule and filter material are measured for ²⁴¹Am-Be and ²⁵²Cf sources separately. For further simplicity the relative flux to the unfiltered case, bare, were evaluated instead of absolute flux. Figure 5 shows a decreasing flux of gamma-ray when

the radius of the spherical Pb shields increases. According to figure 5, a lead spherical shield of radius 4 cm reduces the gamma-ray flux of the ²⁵²Cf source up to 94% relative to the unfiltered neutron source; while this value for ²⁴¹Am-Be source varied up to 50% with the same radius for spherical shield.

The neutron flux passing through the spherical shield is shown in figure 6 for both types of neutron sources. Under these conditions, the total neutron flux increased relative to the "bare" neutron source while gamma-ray flux decreased as seen in figure 5. This increase arose from (n, Xn) interactions in the lead filter. Where, X can be 2 or 3 depending on the neutron energy, table 2 lists (n, Xn) interaction cross sections for the

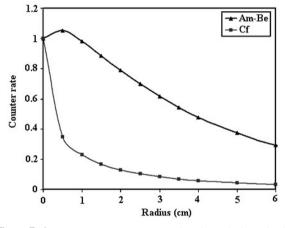


Figure 5. Gamma-rays count rate passing through the spherical shield with several radius (Normalized to an unfiltered neutron source) for both 252Cf and 241Am-Be sources.

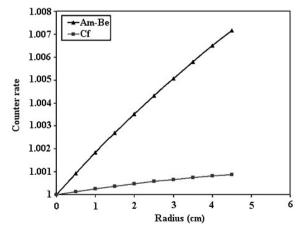


Figure 6. Neutrons count rate passing through the spherical shield with several radius (Normalized to an unfiltered neutron source) for both ²⁵²Cf and ²⁴¹Am-Be sources.

Table 2. (n, Xn) Cross section for natural Pb.

Neutron energy (MeV)	Cross section (Barns)	
(n, 2n)	6.31	4.3523E-03
	7.08	5.85478E-02
	7.90	2.94119E-01
	8.90	7.45645E-01
	10	1.74400E+00
	20	0.00000E+00
(n, 3n)	10	2.60488E-01
	20	0.00000E+00

natural Pb $^{(12)}$. This fact can eliminate the concerning about losing the neutron flux due to the (n, γ) interactions in the filter material and strengthen the idea to use a gamma-ray shield for neutron sources.

Comparisons between filtered (4 cm radius) and unfiltered neutron energy spectrum are shown in figures 3 and 4 for both neutron sources. These figures show that using a lead gamma shield not only increased the neutron flux, but it drifted the whole neutron spectrum to the low or thermal energy region which in turn plays a role in slowing down neutrons. Practically a spherical Pb shield of radius 4 cm is the optimum size, even though spherical shields with larger sizes can be effective.

2. With increasing the radius of the shield, quite little decrease in the total neutron flux colliding to the phantom of soft tissue was resulted (figure 7). This fact showed that although employing a proper gamma-shield the increases outgoing neutron flux (figure 6), but in practical conditions when the spherical Pb shield is not the only shielding material (figure 1); the neutron flux in the vicinity under the soft tissue diminishes. Results shown in figure 7 are presented according to this fact per source neutron rate.

The figures 8 and 9 show the energy spectrum of neutrons, which collide with the soft tissue phantom. These figures indicate that the neutron flux won't have any considerable change with using or not using the spherical shield in the PGNAA setup. Note that 2 cm thickness of Pb shield

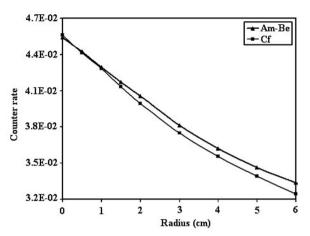


Figure 7. Neutron flux (per source neutron rate) in the vicinity under the sample for several radius of spherical Pb shield for both ²⁵²Cf and ²⁴¹Am-Be.

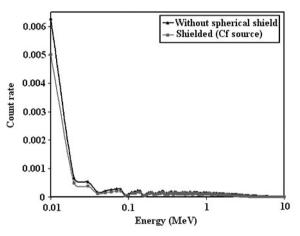


Figure 8. ²⁵²Cf neutron spectrum (per source neutron rate) in the vicinity under the sample.

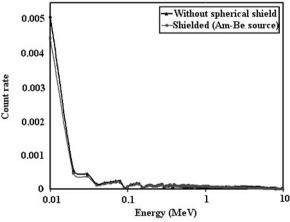


Figure 9. ²⁴¹Am-Be neutron spectrum (per source neutron rate) in the vicinity under the sample.

surrounding the paraffin wax is fixed in all over evaluations.

Figure 10 shows a decrease in the gamma

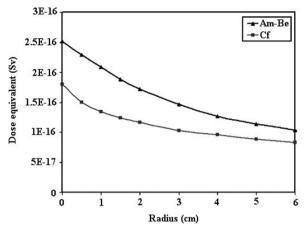


Figure 10. Gamma dose equivalent (in Sv per source neutron rate) measured in the soft tissue plotted against thickness of the shielding for the different sources.

dose equivalent per source neutron rate in the soft tissue equivalent for different radiuses of the spherical Pb shield.

For a spherical Pb shield of radius 4 cm, reduction of the gamma dose equivalent per source neutron rate was about 8.44×10⁻¹⁷ (Sv per source neutron rate) for ²⁵²Cf source and was about 1.24×10⁻¹⁶ (Sv per source neutron rate) when the neutron source was ²⁴¹Am-Be.

DISCUSSION

So far some experiments have been done to measure the body compositions specially by prompt gamma neutron nitrogen, activation analysis method (1). In these experiments industrial systems were used which is originally designed for the real-time elemental analyses of bulk coal on a conveyor belt. Also they have used neutron sources with the high activity regardless to the high intensity of the primary and secondary gamma rays of the neutron sources. Another problem is that the purposed systems don't shield the other body organs from the neutron source. It is clear that the only feasibility of using such system for this determination, even with a high level of accuracy, is not justified since the total absorbed dose by patient is relatively high and important to consider in cancer therapy. Therefore, this method as a diagnostic tool, is not used extensively and going to be

forgotten. At present, six centers make use of this method around the world (13). We have found a way to make this system practical in which it can produce the desirable thermal neutron fluence in the patient body to make the time of exposure shorter and reduce the neutron absorbed dose in other organs using an improved shielded moderator. Also, we examined experimentally and simulated the effects of some materials such as Pb. Bi and Hg as a gamma shield for neutron sources (14). The effects of Pb, presented in this paper, made possible to use these materials in the PGNAA method. These materials are in the category of the spallation materials usually used in spallation neutron sources. Results show that using such materials effectively reduced the total gamma absorbed dose and would act as a moderator.

REFERENCES

- Chichester DL and Empey E (2004) Measurement of nitrogen in the body using a commercial PGNAA systemphantom experiments. Appl Radiat Isot. 60: 55-61.
- Metwally WM and Gardner RP (2004) Stabilization of prompt gamma-ray neutron activation analysis (PGNAA)

- spectra from NaI detectors. *Nucl Instr Meth A* **525**: 518-521.
- Ellis DV (1978) Well Logging for Earth Scientific. Elsevier, New York.
- Pickrell MM (2001) Novel methods for neutron-inducedgamma detection of land mines and buried explosives. Los Alamos National Lab Report. http://www.uxocoe. brtrc.com/TechnicalReps/DoeReport/LosAlamosNatLab. pdf
- NNDC, National Nuclear Data Center, http://www.nndc. bnl.gov/capgam
- Briesmeister JF (2000) MCNP: A General Monte Carlo Nparticle Transport Code, Version 4C, Los Alamos National laboratory, LA-13709-M.
- 7. ICRU (1971) Radiation Quantities and Units. Report 19
- Glässel P, Schmid-Fabian R, Schwalm D, Habs D, Helmolt HUV (1989) ²⁵²Cf fission revisited - new insights into the fission process. *Nucl Phys*, 502: 315-324.
- Palomba M, D'Erasmo G, Pantaleo A (2003) The Monte Carlo code CSSE for the simulation of realistic thermal neutron sensor devices for Humanitarian Demining. *Nucl Instr Meth*, 498: 384-396.
- Kluge H and Weise K (1982) The neutron energy spectrum of a ²⁴¹Am-Be (alpha,n) source and resulting mean fluence to dose equivalent conversion factors. Radiat Protect Dosim, 2: 85-93.
- Croft S (1989) The use of neutron intensity calibrated ⁹Be(alpha,n) sources as 4438 keV γ-ray reference standards. Nucl Inst Meth 281: 103-116.
- OECD/NEA (1997) Joint evaluation file on your PC (JEF-PC-2), OECD Nuclear Energy Agency, France.
- 13. http://www.bcm.edu/bodycomplab/ivnamainpage.htm
- Miri-Hakimabad H, Vejdani Noghreiyan A, Panjeh H (2007) Gamma shielding design studies on ²⁵²Cf and ²⁴¹Am-Be neutron sources. Asian J Exp Sci. 21: 249-258.