

## Design of a Novel Neutron Activation System for Safeguards Purposes using Computer Code Calculations

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**Abstract:** In the aim to design a neutron activation system to verify the nuclear material using neutron source (<sup>252</sup>Cf isotopic neutron source) Monte Carlo simulations were carried out. Several Monte Carlo calculations were carried out using MCNP5 code to estimate the paraffin wax thickness required to shield a <sup>252</sup>Cf isotopic neutron source. Firstly, a point-like source was modeled without shield and the neutron dose rate was estimated. Secondly, the source was shielded in paraffin wax and the simulations were repeated including various thicknesses of paraffin wax cylinders. The calculations performed by means of the Monte Carlo radiation transport code MCNP5 are compared with that optioned experimentally by Wide Energy Neutron Detector Instrument (WENDI). The results obtained were used for design the neutron activation system.

**Keywords:** Neutron Activation System, Shielding Materials, Neutron, paraffin wax, Cf-252 neutron source, Monte Carlo, simulations.

### I. Introduction

Neutron activation analysis (NAA) is a nuclear process used for determining the concentrations of elements in materials. It is very useful as sensitive analytical technique for performing both qualitative and quantitative multielemental analysis. NAA is essentially considered to be a non-destructive method. The method is based on conversion of stable atomic nuclei into radioactive nuclei by irradiation with neutrons and subsequent detection of the radiation emitted by the radioactive nuclei and its identification. The basic essentials required to carry out an analysis of samples by NAA are a source of neutrons, instrumentation suitable for detecting gamma rays, and a detailed knowledge of the neutrons reactions [1].

Neutron activation analysis (NAA) is useful in situations where the identity of the nuclear material is unknown or needs to be verified. For design of a neutron activation system, it is highly desire to chose the best materials for protection against radiation and estimate the thickness required to shield the neutron source.

The transmission of directly and indirectly ionizing radiation through matter and its interaction with matter is fundamental to radiation shielding design and analysis. Design and analysis are but two sides of the same coin. In design, the source intensity and permissible radiation dose or dose rate at some location are specified, and the task is to determine the type and configuration of shielding that is needed.

Radiation shielding involves at placing a shielding material between the ionizing radiations source and the worker or the environment. The radiations which have to be considered are: x and gamma rays, alpha particles, beta particles, and neutrons, each type of these radiations interacts in different ways with shielding material. Therefore, the effectiveness of shielding varies with the type and energy of radiation and also varies with the used shielding material [2].

The best materials for protection against ionizing radiation are mixture of hydrogenous materials (paraffin, polyethylene, water and many plastics) and neutron absorbing elements (B, Li, Bi, Cl, etc.), because they reduce both the intensity of gamma rays and neutrons, indeed, hydrogen slows fast and intermediate neutrons energy via inelastic scattering, and they become thermal neutrons which are absorbed by neutron absorbing elements which have a very high neutron absorption cross-section [2]. Shielding should be light enough to allow for easy movement of the device from one location to other so we have selected paraffin wax as the shielding material as it is enriched with hydrogen and due to its easy availability.

As the fast neutrons get thermalized in the medium, there is a high probability of thermal neutron capture by hydrogen, emitting 2.225 MeV gamma rays. This can be minimized by adding boron into the shielding medium which leads to <sup>10</sup>B (n, 4He) <sup>7</sup>Li reaction giving only 0.478 MeV gamma rays [3].

The most common spontaneous fission neutron source is <sup>252</sup>Cf. Its half-life of 2.65 years is long enough to be reasonably convenient, and the isotope is one of the most widely produced of all the transuranics. The dominant decay mechanism is alpha decay, and the alpha emission rate is about 32 times that for spontaneous fission. The neutron yield is 0.116 n/s per Bq, where the activity is the combined alpha and spontaneous fission decay rate. On a unit mass basis, 2.30 x 10<sup>6</sup> n/s are produced per microgram of the sample. Compared with the other isotopic neutron sources, <sup>252</sup>Cf sources involve very small amounts of active material (normally of the

order of micrograms) and can therefore be made in very small sizes dictated only by the encapsulation requirements [4].

MCNP is a general-purpose, continuous-energy, generalized-geometry, time-dependent, coupled neutron/photon/electron Monte Carlo transport code. It can be used in several transport modes: neutron only, photon only, electron only, combined neutron/photon transport where the photons are produced by neutron interactions, neutron/photon/electron, photon/electron, or electron/photon. The neutron energy regime is from 10<sup>-11</sup> MeV to 20 MeV for all isotopes and up to 150 MeV for some isotopes, the photon energy regime is from 1 keV to 100 GeV, and the electron energy regime is from 1 KeV to 1 GeV. The capability to calculate keff eigenvalues for fissile systems is also a standard feature [5].

The aim of this work is to use computer code to estimate the paraffin wax thickness required to shield a <sup>252</sup>Cf isotopic neutron source to design a novel neutron activation system to verify the nuclear material using neutron source.

## II. Materials and methods

To design the shielding the MCNP5 code was used to calculate the dose rate from the neutron source and the neutron transport. A <sup>252</sup>Cf neutron source with 12μCi of activity (as of 1/3/2010) was used in the calculations. To simplify, the source used in the simulation emits symmetrically and it was treated as a point source in the model. The energy of a neutron emitted from <sup>252</sup>Cf is often modeled as either a Maxwellian or Watt fission spectrum. MCNP manual recommends using a Watt fission spectrum in estimating the fission spectrum of <sup>252</sup>Cf [6]. That is,

$$f(E) = C \exp(-E/a) \sinh(bE)^{1/2} \dots\dots\dots(1)$$

The coefficients a and b vary weakly from one isotope to another.  
For spontaneous fission of <sup>252</sup>Cf,

$$a = 1.025 \qquad b = 2.926$$

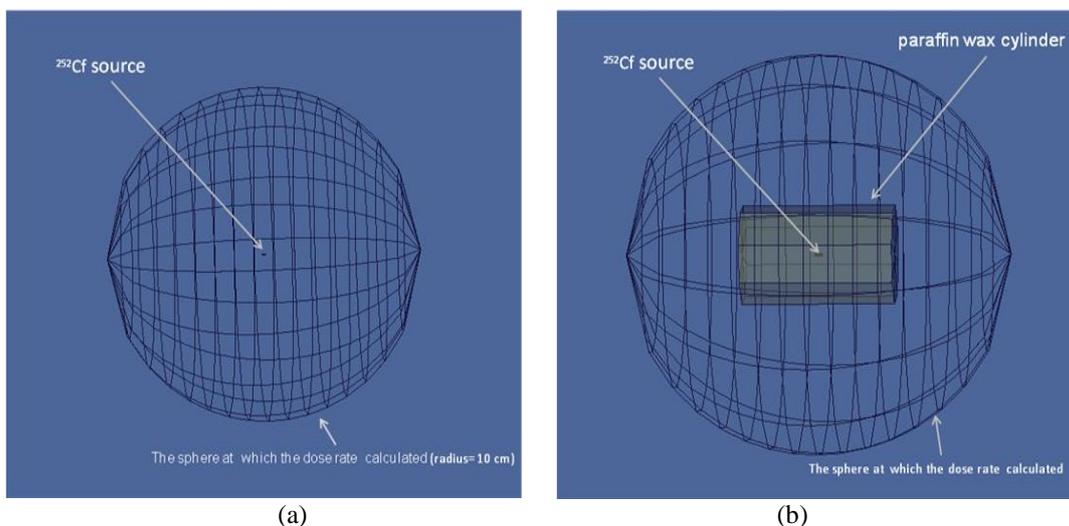
MCNP provides six standard neutron, six standard photon, and four standard electron tallies, all normalized to be per starting particle. Some tallies in criticality calculations are normalized differently [7]. For our problem F2 card (Tally type) was used. The problem has a surface flux tally. MCNP tallies can be modified in many different ways. The DE and DF cards allow modeling of an energy-dependent dose function that is a continuous function of energy from a table whose data points need not coincide with the tally energy bin structure (E card). An example of such a dose function is the flux-to-radiation dose conversion factor given in Appendix H [8]. The flux was converted into dose equivalent rates using the dose factors of ICRP Publication 21 (ICRP 1973).

For purposes of describing the tallies, it is useful to consider a reference sphere whose origin is at the same location of the source. The equator of the reference sphere passes through the origin and is perpendicular to the cylindrical axis of the paraffin wax shield.

Problem cutoff cards are used to specify parameters for some of the ways to terminate execution of MCNP. The mnemonic NPS is followed by a single entry that specifies the number of histories to transport. MCNP will terminate after NPS histories unless it has terminated earlier for some other reason [7]. For our problem we used only the history cutoff (NPS) card. The amount of histories used in each calculation was large enough to reach a Monte Carlo uncertainty less than 1%.

The MCNP5 code was run using a Intel(R) Core(TM)2 Duo CPU (E6750 @ 2.66 GHz 2.66 GHz) with 2 GB RAM under the Windows 7 operating system.

In the first of calculations the <sup>252</sup>Cf neutron source was modeled in air (without shielding) and the dose rate on the surface of the 25 cm reference sphere was calculated where the source is positioned at its center. After that The MCNP5 calculation was repeated with the source shielded in paraffin wax cylinder its radius is 2.5 cm and height is 8 cm and the dose rate was calculated on the surface of the 25 cm reference sphere was calculated where the source is positioned at its center. Fig. 1 show the <sup>252</sup>Cf neutron source and cylinder containing generated by MCNP Visual Editor for the model.



**Figure 1:** Neutron source and cylinder Containing  $^{252}\text{Cf}$  source Generated by MCNP Visual Editor. (a) Neutron source in air (without shield) (b) Paraffin wax cylinder containing  $^{252}\text{Cf}$  source.

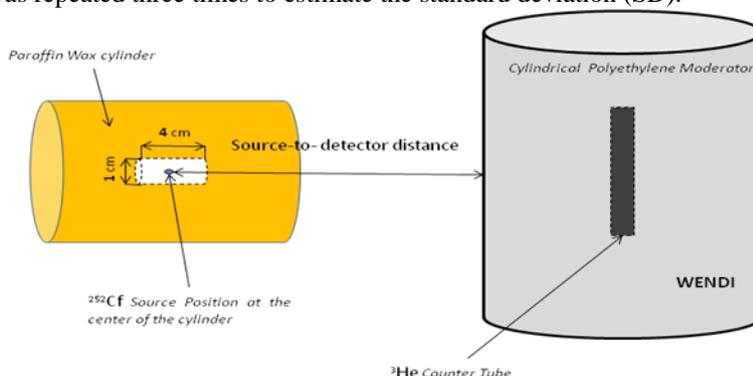
The MCNP5 calculations were repeated with the source shielded in paraffin wax cylinders (the source is positioned at the cylinder center) whose radius was varied from 4.5 up to 16.5 cm and height varied from 12 up to 36 cm and the dose rate on the surface of the 25 cm reference sphere was calculated where the source is positioned at its center. The MCNP5 calculations used  $10^7$  histories which were large enough to reach uncertainties less than 0.1%.

In order to validate the precision of the results calculated with this model, the dose rate from the neutron source in air (without shield) and the dose rate from the shielded source were measured using a Wide Energy Neutron Detector Instrument (WENDI). Four different paraffin wax cylinders were designed to use in this experimental. Dimensions of the cylinders are shown in Table 1.

**Table 1:** Dimensions of the paraffin wax cylinders

Cylinder	Height (cm)	Radius (cm)
C1	8	2.5
C2	12	4.5
C3	16	6.5
C4	20	8.5

Firstly, the dose rate from a  $^{252}\text{Cf}$  neutron source without shield was measured at the source-to-Cylindrical polyethylene of the detector distances  $d=12.5$  cm. Secondly, the source was positioned inside the paraffin wax cylinder (C1) at the centre and the dose rate was measured by positioning the cylinder at the source-to-Cylindrical polyethylene of the detector distances  $d=12.5$  cm where the extended axis of symmetry of the cylinder is perpendicular to the extended axis of symmetry of the detector. The measurements were repeated with the other cylinders (C2, C3, and C4). The experimental setup of the four cylinders was assayed to examine the proposed method. The circular face of the cylinder was facing the detector as shown in Fig.2. Each measurement run was repeated three times to estimate the standard deviation (SD).



**Figure 2:** Experimental setup arrangement to measure the dose rate by Wide Energy Neutron Detector Instrument (WENDI).

### III. Results and discussion

All MCNP tallies are functions of time and energy as specified by the user and are normalized to be per starting particle [9]. So they must be multiplied by the source neutron emission rate. That number can be obtained from the activity in Bq. The flux averaged over the reference sphere surface which calculated by using F2 card was modified according to the dose function. The relative error (error), variance of the variance (VOV) and figure of merit (FOM) of the data are also considered. In Fig.3 is shown the neutron dose rate on the surface of the 25-cm radius reference sphere for the situation when the  $^{252}\text{Cf}$  source is modeled in air and inside the paraffin cylinder shielded.

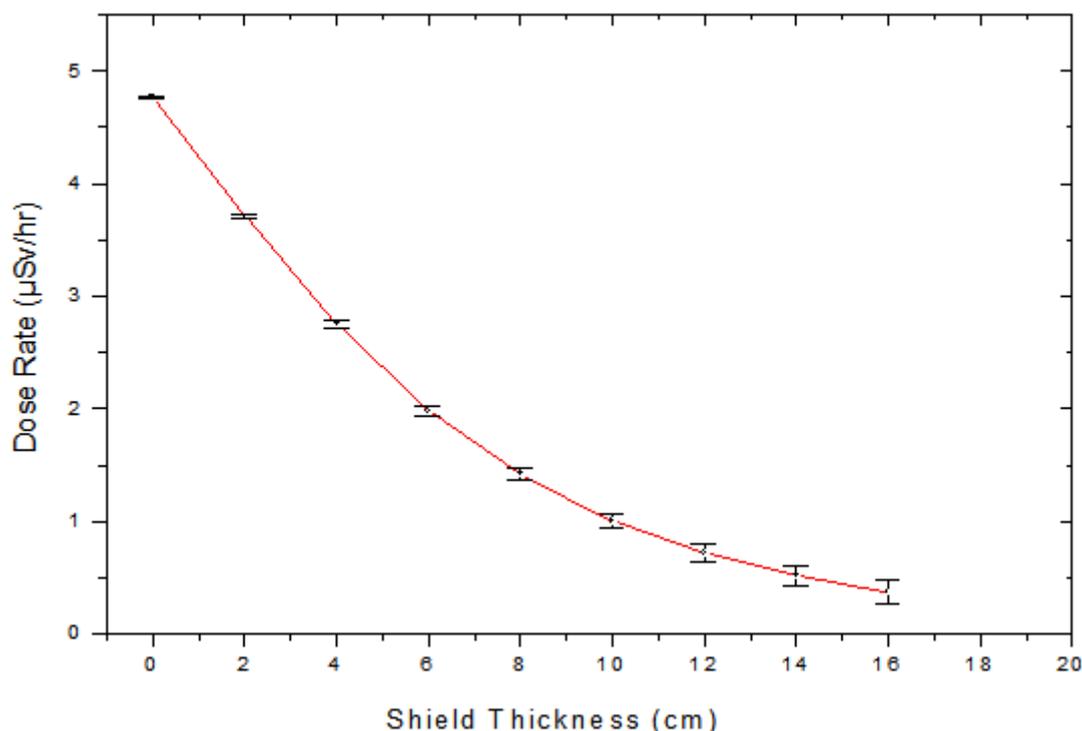
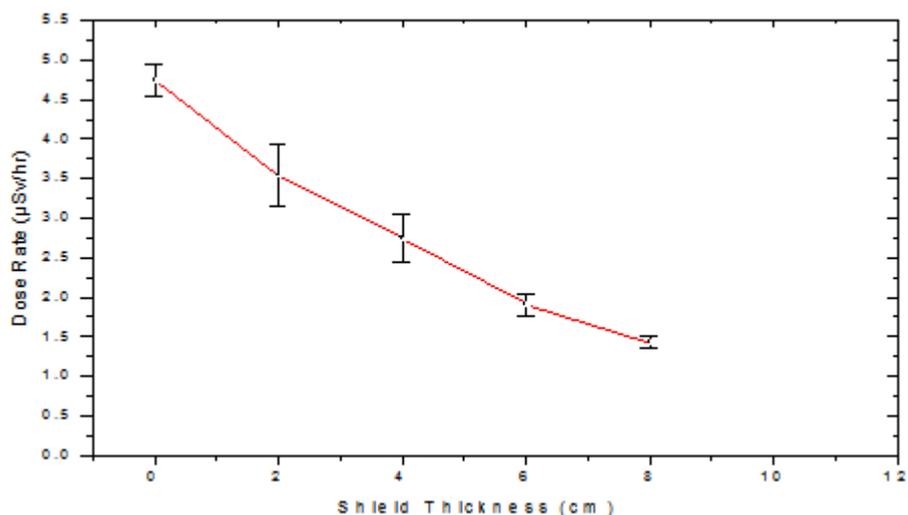


Figure 3: Neutron dose rate calculated by MCNP5 model.

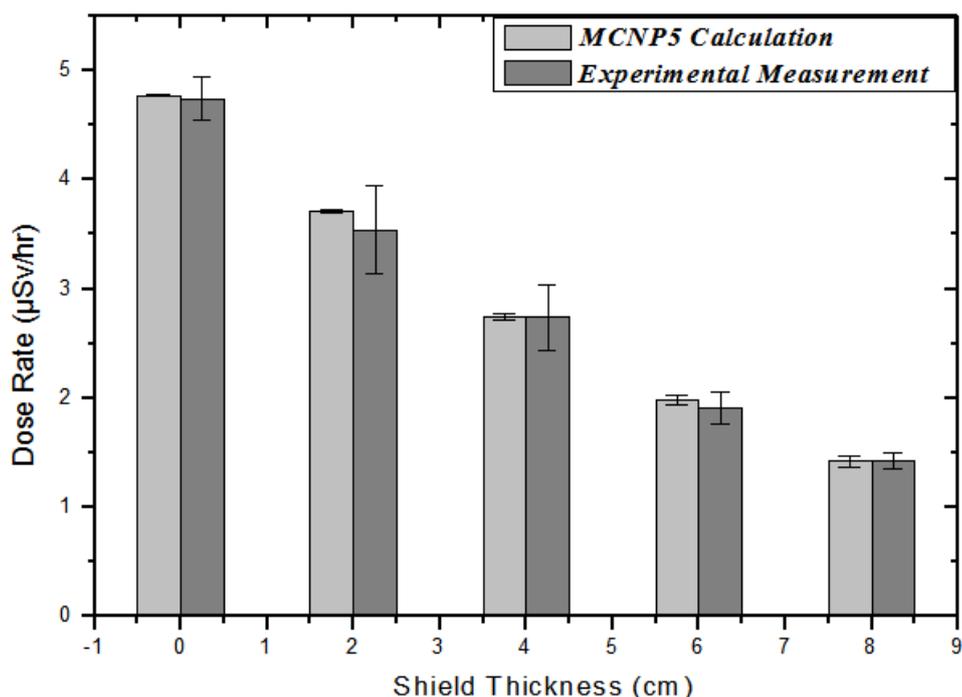
As can be seen from these figures, the calculated dose rate values at reference sphere surface were decreased exponentially with increasing the paraffin wax cylinder thickness, this is mainly due to the attenuation of neutron through the shielding material is generally explained by a high moderating ratio due to the light element contents of the materials composition. The number of light element was increased with shielding materials thickness increased. When paraffin radius increases, the thermal neutrons flux increases, fast neutron flux decreases. The thermal neutron flux rate initially increases with the moderator radius, then began to decrease. The initial increase in the thermal neutrons yield may be due to neutrons scattering cross section greater than the absorption cross section. Then subsequent decrease in thermal neutron yield with further increase in the moderator radius may be due to the absorption cross section than scattering cross section greater due to increasing thermal neutrons flux rate with increasing moderator radius. The fast neutrons flux rate reduces linearly with the increase of the paraffin radius. All estimated uncertainties due to MC calculations for all setup configurations were less than 0.1%. Also, it can be notice from the figure that the 16 cm-shield is widely fulfill the requirement shielding design.

The results obtained from measurement dose rates with WENDI are plotted in Fig. 4 with their uncertainties. It is clear from the figures the decreased of the dose rate exponentially with increasing the shield thickness. All estimated uncertainties due to measurements for all setup configurations were less than 0.1%.



**Figure 4:** Neutron dose rate measured by Wide Energy Neutron Detector Instrument (WENDI).

Fig.5 compares the MCNP code calculated dose rates with those obtained from the measurements. The measured dose rates range from 4.74 µSv/h for source without shielding to 1.34 µSv/h at 8cm paraffin thickness. MCNP calculated results range from 4.77 µSv/h for source without shielding to 1.42 µSv/h at 8cm paraffin thickness. The MCNP results agree with the measurements within 10% for all the measured locations. It is clear that the estimated dose rate using both methods is in agreement within the uncertainties.



**Figure 5:** Neutron dose rate calculated by MCNP5 model in comparison with that measured by Wide Energy Neutron Detector Instrument (WENDI).

#### IV. Conclusion

MCNP uses a per particle history average to score tallies. By modeling and simulating the production, path, and interactions of millions, or as in this case, billions of particles, MCNP can accurately estimate the dose to 52 individuals in radiological events. Utilizing MCNP therefore is an excellent method for simulating and observing the dose rate from a known source at an instant in time.

The Monte Carlo simulation code MCNP5 is employed to determine the paraffin wax thickness require for shielding a <sup>252</sup>Cf neutron source with 12µCi of activity. Calculations performed for a cylindrical defined configuration from paraffin wax, a series of cylinders with radius between 2.5 cm and 16.5 cm in 2 cm increments are modeled.

The reliability of the model is assessed by comparing the calculated results for four cylinders (it is radius are 2.5, 4.5, 6.5 and 8.5cm) with the experimental results obtained from WENDI for the same cylinders.

The results obtained from this calculation will be used for design a neutron activation system to verify the nuclear material using the  $^{252}\text{Cf}$  isotopic neutron source.

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