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Study of different solutes for determination of neutron source strength based on the water bath

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ABSTRACT

Time required for activation to saturation and background measurement is considered a limitation of strength determination of radionuclide neutron sources using manganese bath system (MBS). The objective of this research was to evaluate the other solutes based on water bath for presentation of the suitable replacement with MBS. With the aid Monte Carlo simulation, for three neutron sources, having different neutron spectra, immersed in six aqueous solutions, i.e., Na_2SO_4, $VOSO_4$, $MnSO_4$, $Rh_2(SO_4)_3$, $In_2(SO_4)_3$, I_2O_5, the correction factors in all nuclei of solutions for neutron losses with different process were obtained. The calculations results indicate that the $Rh_2(SO_4)_3$ and $VOSO_4$ are best options for replacing with $MnSO_4$.

1. Introduction

There are three types of radionuclide neutron sources: (γ, n), (α, n), and spontaneous fission. All of these, which may be defined as standards are available as long-lived activities embodied into small capsules (Knoll, 1989). Available on an international basis, they are widely used to calibrate area survey instruments, neutron detectors and personal dosimeters (ISO, 1998, 2000, 2001). The precision of this method of calibration is dependent on having a detailed knowledge about their strength and neutron energy (Khabaz and Vega-Carillo, 2013). The principal method utilized in a number of metrological laboratories to measure the neutron source emission rate is the manganese bath system (MBS) (Khabaz, 2012a, 2012b). In addition, the MBS produces calibration uncertainties better than 1% (Leite, 2005).

This bath consists (optimally) of a tank filled with an aqueous solution of manganese sulfate, with the source located in the center of the tank. The measurement is on the basis of the slowing down and capture of neutrons by the $^{55}Mn(n, \gamma)^{56}Mn$ reaction. If the bath is large enough, practically all the neutrons sent out by a source, which is in the center, are slowed down to thermal energies and captured by the nuclei of the solution components, i.e., manganese, hydrogen, sulfur nuclei and oxygen (Hwang and Lee, 1988; Park et al., 2005). Using the capture cross sections ratio of manganese and other nuclei, one can calculate the total captures fraction occurring in manganese. Since manganese has only one stable isotope, ^{55}Mn, each capture in manganese results in the formation of a nucleus of ^{56}Mn decaying with a half-life of 2.58 h. This can be readily detected through its γ-rays. If the neutron source is left in the bath until the manganese activity has built up to equilibrium (about 20–24 h), the number of manganese atoms disintegrating in the bath is equal to the number of neutrons captured per second by manganese nuclei (Bittencourt et al., 2010). If the total activity of ^{56}Mn, $A(t)$, can be measured, the neutron emission of the source, $B(t)$, is given by:

$$B(t) = A(t)/[\varepsilon_c F (1 - f_R)(1 - f_S)(1 - f_L)] \quad (1)$$

where ε_c is the system counting efficiency; F is the fraction of thermal neutrons captured by manganese; f_R is the fraction of neutron captured in the (n, p) and (n, α) reactions by the nuclei of the solution components; f_S is the fraction of source neutrons captured in the cavity assembly and neutron source; f_L is the fraction of source neutrons escaping from the bath boundaries.

The dependence of these corrections on various factors suggests the necessity of modeling. Monte Carlo method is suitable for solving complex three dimensional problems, proving impossible to be solved analytically. Therefore, a Monte Carlo simulation has the ability to track neutrons from the source (starting with the appropriate energy spectrum) to a point in the bath where their energy can be considered as thermal. Any (n, α) and (n, p) reactions in oxygen, sulfur and manganese can be also logged by the simulation, providing the calculation of the capture fraction (f_R) (Roberts, 2001).

By replacing the individual evaluations of various correction factors through a direct calculation of the probability of the neutron capture by manganese nuclei (f_P) using the Monte Carlo method, the source emission rate is then given by:

$$B(t) = A(t)/(\varepsilon_c f_P) \quad (2)$$

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Table 1
Comparison of activation elements with manganese (Lide, 2010; Chadwick et al., 2006).

	$_{11}$Na	$_{23}$V	$_{25}$Mn	$_{45}$Rh	$_{49}$In	$_{53}$I
Primary active isotope	^{24}Na	^{52}V	^{56}Mn	^{104m}Rh	^{116m}In	^{128}I
Abundance of primary isotope (%)	100	99.75	100	100	95.7	100
Secondary isotope	None	^{50}V	None	None	^{113}In	None
Appropriate soluble salt	Na_2SO_4	$VOSO_4$	$MnSO_4$	$Rh_2(SO_4)_3$	$In_2(SO_4)_3$	I_2O_5
Solubility in cold water (g/liter)	267	700	520	Very soluble	540	1874
Cost[a] ($/kg)	1	70	3	100	30	50
Half-life of primary isotope	14.965 h	3.743 min	2.5785 h	4.34 min	54.3 min	24.99 min
Cycle time	12 d	1.2 h	2 d	1.5 h	18 h	8 h
Gamma energy (MeV)	1.369 (100%), 2.754 (99.9%)	1.434 (100%)	0.847 (93.9%), 1.811 (27.2%), 2.113 (14.3%)	0.051 (48.2%)	0.417 (27.8%), 1.097 (57.1%), 1.294 (84.4%), 1.508 (9.9%), 2.112 (15.5%)	0.443 (17.0%)
Beta end-point (MeV)	1.390 (99.9%)	2.542 (99.2%)	0.735(14.6%), 1.037 (27.8%), 2.848 (56.3%)	0.622 (0.04%), 1.380 (0.06%)	0.597 (10.4%), 0.869 (34.8%), 1.007 (51.5%)	1.682 (15.0%), 2.125 (77.0%)
Activation cross section[b] (barn)	0.53	5.05	13.42	146.34	202.26	6.19

[a] Approximate price for 99% chemically purity.
[b] Thermal cross section at 0.0253 eV.

One of the MBS limitations is the time required to activate saturation and subsequent decay to background level; approximately ten half-lives (~ 24 h) are required to exceed 99.9% saturation, and the same amount of time to retreat to background (McGarray and Boswell, 1988). Not only, it is a two-day cycle somewhat time-consuming from an experimental point of view, but it tends to hide the existence of short-term background fluctuations. The latter deficiency is particularly considerable when irradiations are conducted near a reactor or accelerator (Scott, 1970).

This work attempts to substitute other solutes for the bath water as the activation component. An analysis of the periodic table for suitable short-lived substitutes indicates few other candidates. One prime requirement is that the element be nearly mono-isotopic; other related characteristics are cross section, solubility, half-life, cost and availability as well as radiation signature.

Several other elements which can satisfy these general constraints are $_{11}$Na, $_{23}$V, $_{45}$Rh, $_{49}$In and $_{53}$I. The characteristics of these elements are compared with $_{25}$Mn in Table 1.

All elements are mono-isotopic except vanadium and indium. The natural vanadium and indium consist of two isotopes: [^{51}V (99.76%), ^{50}V (0.24%)] and [^{115}In (95.7%), ^{113}In (4.3%)], respectively.

In this work, using Monte Carlo simulation, a spherical bath containing two different concentrations of six aqueous solutions (Table 1) was modeled. In the case of each solution, all correction factors of Eqs. (1) and (2) for three radionuclide neutron sources having different energy spectra were calculated and compared.

2. Materials and methods

MCNP is regarded as the most extensively used general purpose Monte Carlo neutron-photon-electron transport code. Transport equations describe particle transport in media, and also Monte Carlo method is one way to describe neutron transport in media with the use of the results to infer the mean behavior of all particles by the Central Limit Theorem to receive answers by simulating the actual behavior of particles.

It was modeled, in the MCNPX2.6 (Pelowitz, 2008) radiation transport code, a 3-mm-thick stainless steel (316L. type, ρ = 7.93 g/cm^3) (Lide, 2010) bath with inner diameter of 127 cm, containing six aqueous solutions, i.e., Na_2SO_4, $VOSO_4$, $MnSO_4$, $Rh_2(SO_4)_3$, $In_2(SO_4)_3$ and I_2O_5. This bath was filled with two different concentrations belonging to these solutions, i.e., 220 (CI) and 350 (CII) g/(kg of solution), which for Na_2SO_4 was only for CI (saturation concentration for this solute). For concentration CI, the number ratio of hydrogen nuclei to primary nuclei of these aqueous solutions was 27.95, 64.14, 59.43, 97.19, 101.88 and 65.70, respectively. Except for Na_2SO_4 solution, the corresponding values for CII were also 33.61, 31.13, 50.93, 53.37 and 34.41 for these related solutions.

In the simulation, the neutron source capsule was located in the spherical Teflon holder (Ø10 cm), the thickness of whose wall was 4 mm. This holder itself was located at the bath geometric center (Fig. 1).

The radionuclide neutron sources considered for this study were three well-known sources, i.e., ^{241}Am–Li, Po-Be and ^{242}Cm-Be, which have the X.14 (Ø30 mm × 60 mm) stainless steel capsule. The mean energy values of these sources were 0.56, 2.04 and 5.50 MeV, respectively. The Po-Be and ^{242}Cm-Be sources were defined as a cylinder of beryllium (with a density of 1.85 g/cm^3), and ^{241}Am–Li as a cylinder of lithium (with a density of 0.534 g/cm^3) contained in a stainless steel capsule. Fig. 2 shows the neutron energy spectra of these sources, which have been used in MCNP simulation (Griffith et al., 1990).

The correction factors calculations were carried out based on the above-mentioned MCNPX simulation using the ENDF-B/VII.0 cross section data library (Chadwick et al., 2006). Fig. 3 shows and compares

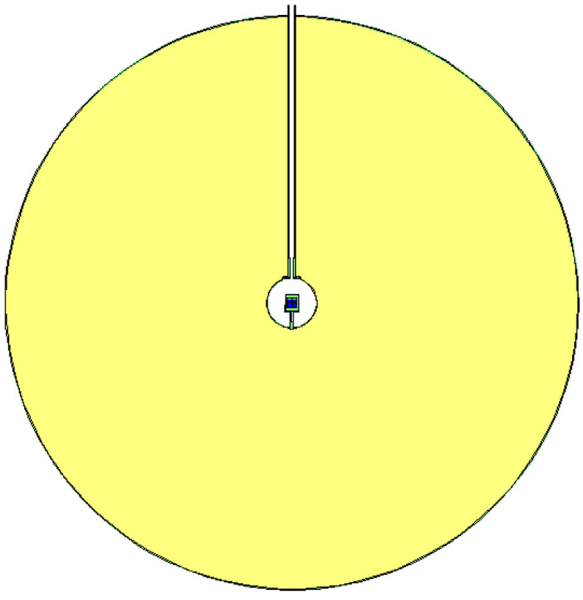

Fig. 1. Schematic diagram of bath simulated by MCNP.