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Prototype Mn-solution bath for low activity primary standard neutron dosimetry

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Abstract
In this work, a prototype Mn-bath was designed and constructed in our laboratory. It is composed of a spherical 304 stainless steel vessel with a diameter of 34 cm. A hydraulic system for circulating the ^{55}Mn solution between the bath and the detection system. The detection system is composed of a Marinelli beaker attached to a $3\times3''$ NaI(TI) crystal detector connected to the Nomad analyzer. A ^{252}Cf source with a flux of order 10^3 (n cm^{-2} S^{-1}) was used. The output dose was measured by a standard NM2 neutron monitor. The output flux was calculated from the flux-dose relationship. It was found that the flux delivered by the used ^{252}Cf was 170.56 ± 6.41 n cm^{-2} S^{-1}as calculated from the dose measured by NM2. Different concentrations of ^{55}Mn solution with the molarity of 0.89,1.18 and 1.48 (mol/l) extracted a flux of 194.48, 186.46, and 169.79 n cm^{-2} S^{-1}, respectively. Data of neutron flux obtained from concentrations of 1.18 and 1.48 mol/l was in a good agreement with the flux measured by the standard neutron monitor and the guaranteed value of the used source. Uncertainty of flux measured by Mn bath was enhanced with increased concentration.

Keywords Primary neutron dosimetry · Mn-bath · CF-252

Introduction

Ionizing Radiation Metrology Laboratory (IRML) is one of Secondary Standard Dosemetric Laboratories (SSDL) of International Atomic Energy Agency IAEA and World Health Organization WHO. Neutron dosimetry and calibration of neutron survey meters are one of the IRML activities. A neutron monitor model NM2 is used as a Secondary Standard Dosimeter for fast neutrons in IRML, this monitor was calibrated in the Physics Technical Bureau (PTB) in 2016. IRML promoted its capabilities by the construction of primary standard dosimeter for neutron. Standard laboratories should possess their dosimeter for each activity to spare the need for traceability [1–7]. It is well established that standard dosimeter for neutrons are ionization chamber, Mn bath and neutron monitors as recommended by the International protocols [8, 9]. Since ionization chambers are highly expensive instrument that measures neutron field accurately at one direction, and since neutron monitors are considered secondary standard that measure neutron energy in a wide range with reasonable accuracy, then Mn-bath advantages over both ionization chamber and neutron monitor when it comes to accuracy or cost. Mn-bath is cheap to construct when compared to ion chambers and measures accurately the emission rate of a radio nuclide neutron source in 4π geometry [10, 11]. In addition to low cost an accuracy of Mn-bath, it is easy to self-maintain.

The manganese bath consists of three main parts: bath system, hydraulic system and detection system. It is spherical in shape filled with manganese sulfate solutions of a specific concentration. The hydraulic system is used to maintain circulation of Mn solution between the bath and the detection system [12]. During the design of the Mn-bath, it is necessary to select the dimension of the bath system and the concentration of Mn-solution to be compatible with the activity of the used source [13].

The used neutron source was Am-Be or ^{252}Cf fixed in the center of the sphere. Fast neutrons delivered by the used source are slowed down by a successive elastic scattering with hydrogen nuclei by water used for Mn solution preparation.

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After fast neutron moderation, ^{55}Mn capture the thermal neutron and convert it into ^{56}Mn by the following reaction:

$$^{55}Mn + n_{thermal} \rightarrow ^{56}Mn^{*} \rightarrow ^{56}Fe + \gamma + \beta \quad (1)$$

^{56}Mn decays through beta particles with different gamma rays of energies 846.7, 1810.6 and 2113.04 keV. The activity of ^{56}Mn is obtained by using gamma spectroscopy [14], and then neutron flux (φ) can be calculated by the equation:

$$\varphi = A/\sigma N \quad (2)$$

where A is the activity per liter for ^{56}Mn, σ is the capture cross-section for ^{56}Mn and N is the number density of the manganese atom in one liter. The cross-section of ^{55}Mn was selected at neutron energy of 0.4 eV which is 5.64 barns [15].

The main objective of this work is to design and construct small Mn-bath for measuring low activity neutron source flux. Moreover, to study the optimum conditions needed for accurate flux determination and to calculate the uncertainty of the neutron flux.

Experimental set-up

Neutron source

The neutron source used in this study was ^{252}Cf manufactured by Amersham Co.UK. The source has an original mass of 50 µg point source in a pressurized sealed stainless-steel cylinder with 5 cm diameter and 10 cm height and of activity 7.24 µCi [16] and flux of 178.67 n cm^{-2} S^{-1}.

Manganese bath system construction

Bath system

The bath system consists of a 304 stainless steel sphere with 34 cm diameter and 2 mm thickness. An upper opening to insert the source through with 10 cm width in addition to two other entry and exit holes through which Mn solution is circulating with aid of the hydraulic system. The sphere on carriage to facility motion.

Hydraulic system

The hydraulic system consists of a pump with different flow rates (from 0.5 to 10 l/mint) to circulate the solution between the sphere and the detection system. Moreover, a set of filters are used to capture impurities within the solution of diameters range greater than 0.2 mm.

Detection system

The detection system consists of a Marinelli beaker of 1-l volume and an inorganic scintillation detector connected to the analyzer. The used detector is a 3 × 3 NaI(Ti) inorganic scintillation manufactured by Canberra with type 2007P. The detector is connected to the power supply with type 428 and an amplifier model 575A with different gain. The output of the amplifier connected to the Nomad analyzer contains a multi-channel analyzer manufactured by Ortec where the final signal is reading of the personal computer system. A schematic diagram of the designed Mn bath is illustrated in Fig. 1a and the a photograph of the constructed Mn-bath and the Gamma spectroscopic system is represented in Fig. 1b.

Results and discussion

Flux of ^{252}Cf measurements

The flux of ^{252}Cf used in this study was firstly determined by the certified NM2 neutron monitor manufactured by Nuclear Enterprises by considering the sensitive depth of the monitor. The flux was determined from the corresponding dose equivalent by using the flux-dose factor [16] where the flux of the used source was 170.5 ± 6.41 n cm^{-2} S^{-1}. Moreover, and to confirm, the neutron flux was calculated by the traditional decay law from the original source activity where a value of 178.67 n cm^{-2} S^{-1} was obtained that reflects the match in the both measured and calculated values.

Efficiency of the gamma spectrometer

The efficiency of the used gamma spectrometer was determined using a standard multi-radio-nuclide source in a Marinelli beaker manufactured by Eckert & Ziegler. This Marinelli beaker contains different isotopes such as ^{137}Cs and ^{60}CO. Its original calibration date is Apr-2009. Gamma-ray emitted from different isotopes in Marinelli measured by using gamma-ray spectroscopy. The photopeak of ^{56}Mn efficiency is obtained by using standard analytics. It is well known, that efficiency of a scintillation detector decreases exponentially with gamma energy, however, in a small window of higher gamma energies (from 662 to 1330 keV of ^{137}Cs and ^{60}Co sources) the efficiency may be linear with gamma energy as represented in Fig. 2. A fitting of efficiency-gamma energy relationship of the data is represented in Fig. 2 has a linear equation takes the form:

$$Eff = aE + b \quad (3)$$

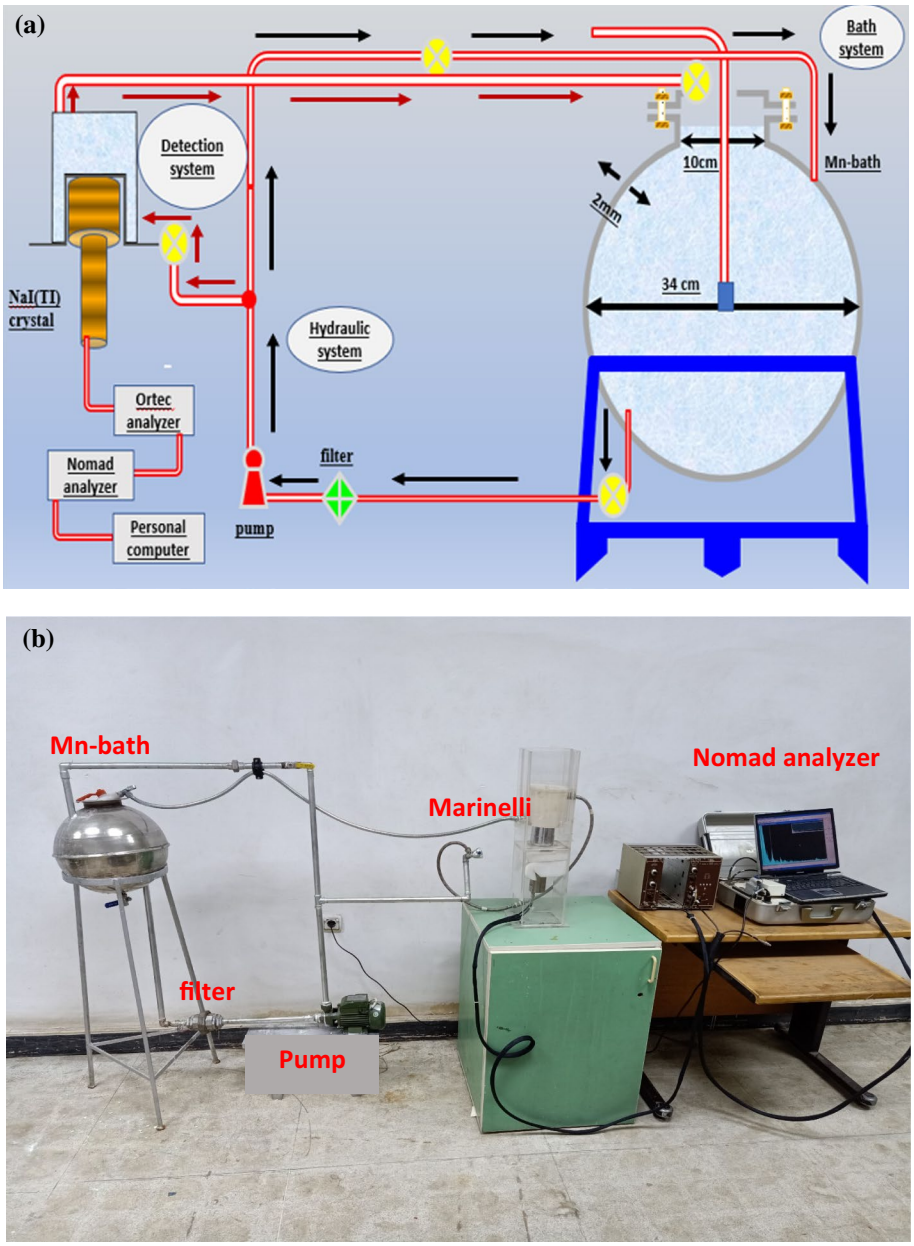

Fig. 1 Shows the designed Mn bath (**a**) and a Photograph of the constructed Mn-bath (**b**)

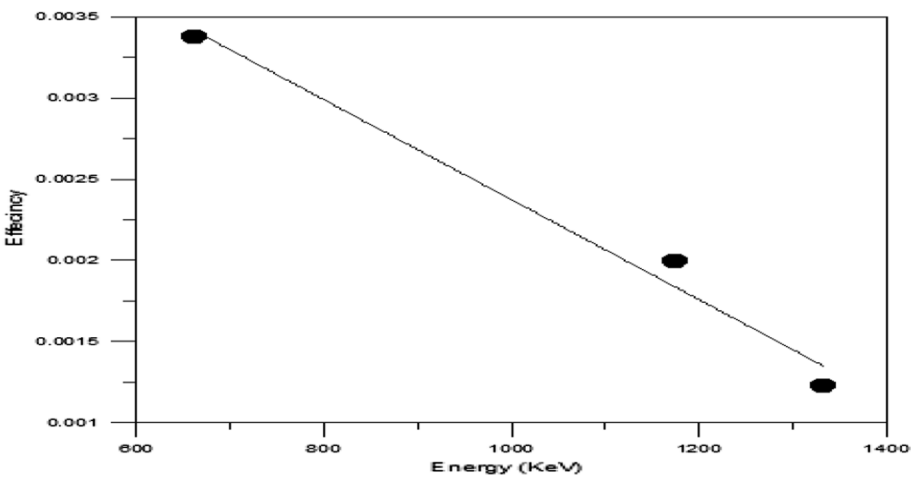

Fig. 2 Efficiency as a function of gamma energy using a standard Cs-137 and Co-60 sources

where a and b are fitting parameters with values of -3.1×10^{-6} and 5.45×10^{-3} respectively and with regression of 98.1% where the uncertainty of this assumption is considered in the uncertainty budget.

From the data and the fitting equation, one can easily obtain the efficiency of gamma line of ^{56}Mn at an energy of 864.7 which has a value of 0.00285 ± 0.00012. The gamma line of energy 864.7 keV was selected due to its higher branching ratio of ^{56}Mn radioactive decay (98.78%).

Flux determination using Mn bath

In order to determine the flux of the ^{252}Cf source using the constructed Mn-bath, a solution of different molarities from $MnSO_4$ was prepared having 0.89, 1.18 and 1.48 mol/l. The californium source was fixed at the center of the sphere that contains the $MnSO_4$ solution and was left to set for 24 h (five folds the value of the half life time) to assure saturation of ^{56}Mn activity and to attain the fulfillment of the build- up.

After activity saturation, the solution was inserted into the Marinelli beaker with continuous stirring the solution. Using the gamma-ray spectroscopy, the activity of the solution was determined by following equation:

$$A = cps/\xi\varepsilon \tag{4}$$

where the cps is net counts per second taken from gamma spectroscopy, ξ is the branching ratio of the ^{56}Mn gamma line with energy 846.7 keV and ε is the photo peak efficiency for ^{56}Mn gamma line.

The flux (φ) is calculated from the relation $\varphi = A/\sigma N\,\frac{A}{\sigma N}$ (see Eq. 2).

The procedures were used for all concentrations. Figure 3 shows the spectrum of ^{56}Mn with different molarities.

The flux measurements were performed for each prepared concentration 4 times and the repeatibility U_{ret} was calculated from the relation [8, 9]:

$$U_{ret} = \sigma/n^{0.5}$$

where σ is the standard deviation and n is the number of measurements.

Figure 4a–c shows the determined flux and its standard deviation with the measurement number. From Fig. 4, the values of the standard deviation are 12.83, 9.44 and 8.1% for molarity of 0.89, 1.18 and 1.48, respectively, that reflects the enhanced standard deviation with the increased concentration.

The mean flux values for each concentration were collected in Table 1 with N_H/N_{Mn} ratio and is represented in Fig. 5 with molarities used in this study. From this figure, it's obvious that the flux measured by Mn-bath is in agreement with the flux measured by the neutron monitor and with that calculated by traditional decay law from the original value of the ^{252}Cf source. Moreover, the upper degree of equivalence showed at molarties 1.18 and 1.48 mol/l.

Uncertainty calculation

The combined uncertainty (U_c) calculated from the statistical sources U_A and uniform sources U_B and is given by [9, 16]:

$$U_c = \sqrt{U_A^2 + U_B^2} \tag{5}$$

In this work, the neutron flux is determined using Eq. (2) based on the ^{56}Mn activity and number density. The activity (see Eq. 4) was determined from CPS and the spectrometer efficiency.

All the uncertainty of these parameters is considered for all concentrations done in this work. An example of the detailed uncertainty calculation for normality 1.48 mol/l is collected in the budget in Table 2.

From the above budget, the combined uncertainty for 1.48 mol/l is 6.68%. By the same method, the uncertainties for the other studied molarities are 8.33% and 7.10% for 0.88, 1.18 mol/l respectively.

General discussion

A prototype Mn-bath was designed and constructed in our Lab for law activity neutron flux determination. The suggested radius was determined as the depth of water needed for fast neutron moderation is 17 cm. Hence, the diameter of the constructed sphere bath is 34 cm. The efficiency of the used gamma spectrometer used for inducing activity determination has the same geometry of the standard multi-radio-nuclei source with the same geometry and volume (1 L). Gamma line of ^{56}Mn with energy 846.7 keV was selected for activity determination due to its higher branching ratio than others which have a value of 98.87% (see Fig. 3). Different concentration of $MnSO_4$ was used to obtain different ratio of hydrogen number density (N_H) to manganese (N_{Mn}). This ratio plays an important role in studying the characterization of any constructed Mn bath. Since the value of (N_H) is responsible of slowing down fast neutrons and N_{Mn} is responsible of thermal neutron capture, thus both activity and flux are affected. The values of N_H/N_{Mn} ratio were 124.82, 93.61 and 74.89 for molarities of 0.88, 1.18 and 1.48, mol/l respectively. All the obtained values of neutron flux of the prepared concentrations were in

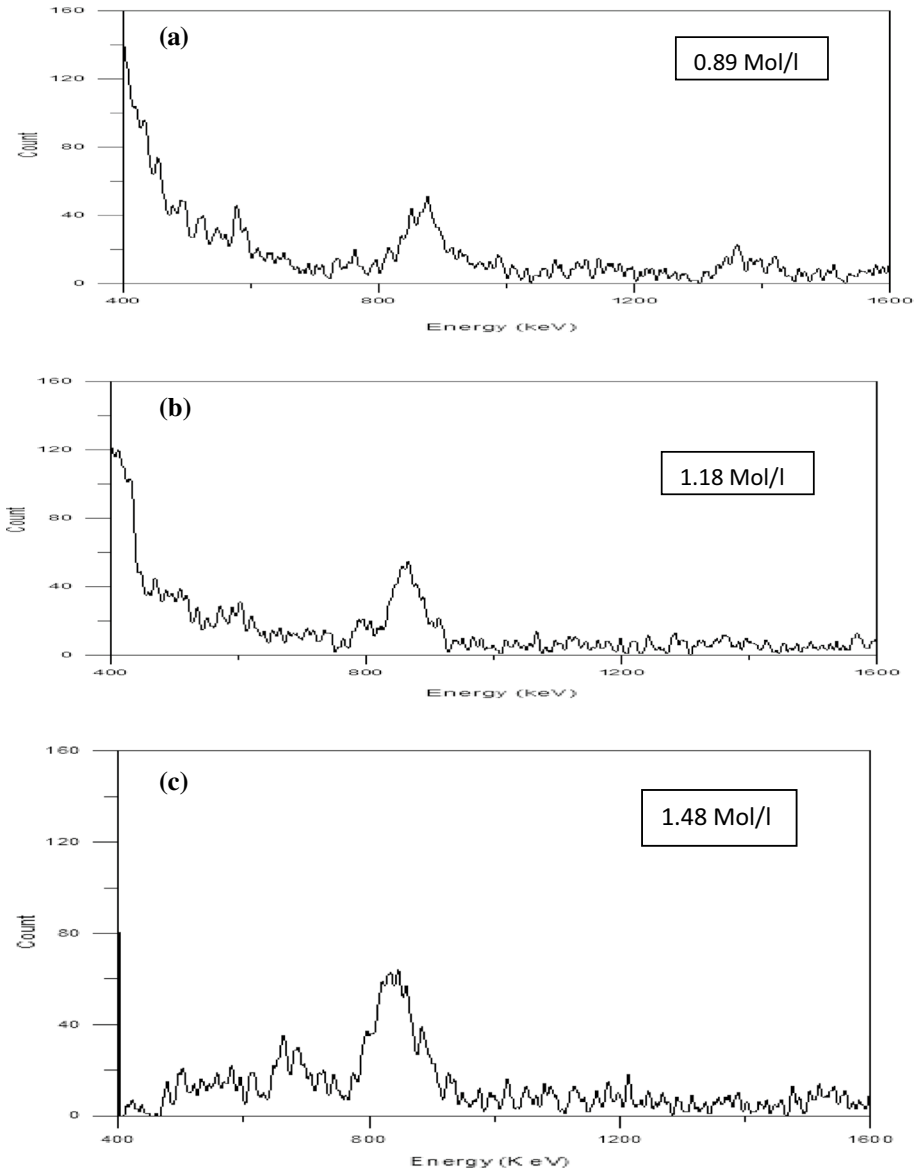

Fig. 3 Spectrum of ^{56}Mn at different concentrations **a** 0.89, **b** 1.18 and **c** 1.48 mol/l

good agreement with that obtained by the standard neutron monitor and that of calculations. However, it was found that both the moralities 1.18 and 1.48 mol/l are of good degree of equivalence with the calculated flux and concentration of 1.48 mol/l had showed lower value of uncertainty which can be attributed to its high number of ^{55}Mn nuclei that enhanced higher thermal neutron capture and hence higher activity. Uncertainty in the determined flux decreases with the concentration increases, which may be attributed to the higher activity obtain with higher concentration increases the accuracy of the determined parameters used for flux determinations. In future work, a large bath will be constructed in the same manner for higher activity sources, enhance the uncertainty value and participated in the inter-comparison programs with the other standard lab [17–19].

Conclusion

A prototype of Mn-bath was designed and constructed in our Lab to be used as a primary standard dosimeter for neutron sources. Different concentrations of $MnSO_4$ were used for a low activity flux determination of ^{252}Cf source. The Flux obtained using different concentrations used was $194.5 \pm 8.33\%$, $186.5 \pm 7.10\%$ and $169.78 \pm 6.7\%$ with molarities of 0.88, 1.14 and 1.48, mol/l, respectively. Moreover, the flux of the used source was also determined by the standard NM2 neutron monitor and calculated by traditional decay law where a value of 170 was and 178.67 n cm^{-2} S^{-1}, respectively was obtained that reflects an agreement between them. The higher degree of equivalence between measured by Mn bath was obtained

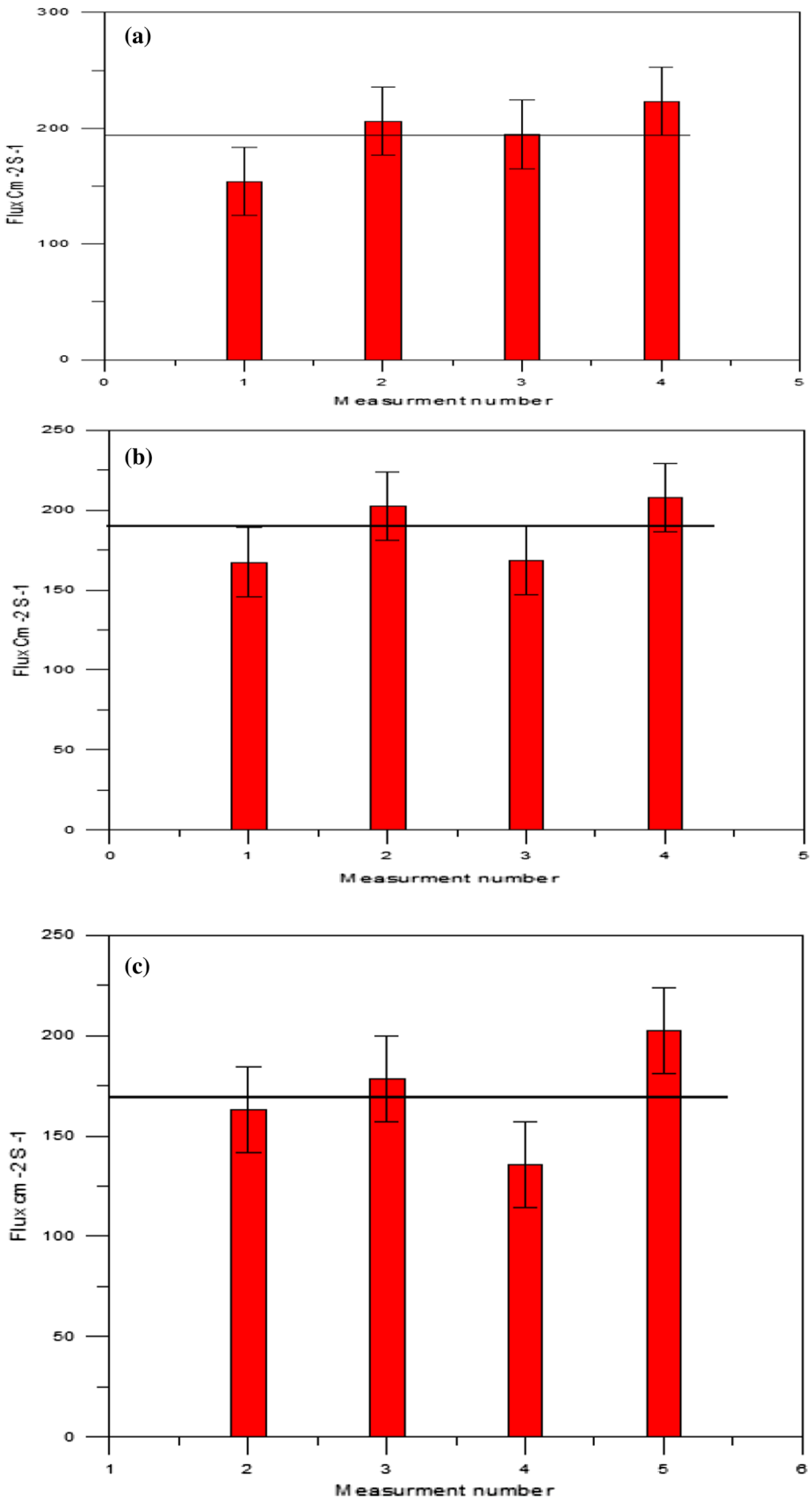

Fig. 4 Shows the flux values and its standard deviation using molarities of 0.89 mol/l (**a**), 1.18 mol/l (**b**) and 1.48 mol/l (**c**)

Table 1 Values of neutron flux as calculated using different concentrations

Molarities mol/l	N_H/N_{Mn}	Flux cm^{-2} S^{-1}
0.89	124.82	194.48 ± 8.33%
1.18	93.61	186.46 ± 7.10%
1.48	74.89	169.79 ± 6.68%

using concentrations of 1.18 and 1.48 mol/l where a ratios between the calculated to the measured was 95.80% and 1.05. The uncertainty decreases with the concentration increases.

Table 2 Uncertinty budget for the molarity of 1.479 mol/l

Parameter for 1.48 mol/l	U_A (%)	U_B (%)
Repeatability	4.05	0
Number density	0.264	0.004
Photo peak efficiency	2.8	0.0035
Geometry efficiency	0.01	0.065
CPS for 1.48 molarity	0.01	4.5
U_c^2	U_A^2	U_B^2
	24.31	20.25
U_c	$(U_A^2+U_B^2)^{1/2}$	
	6.68%	

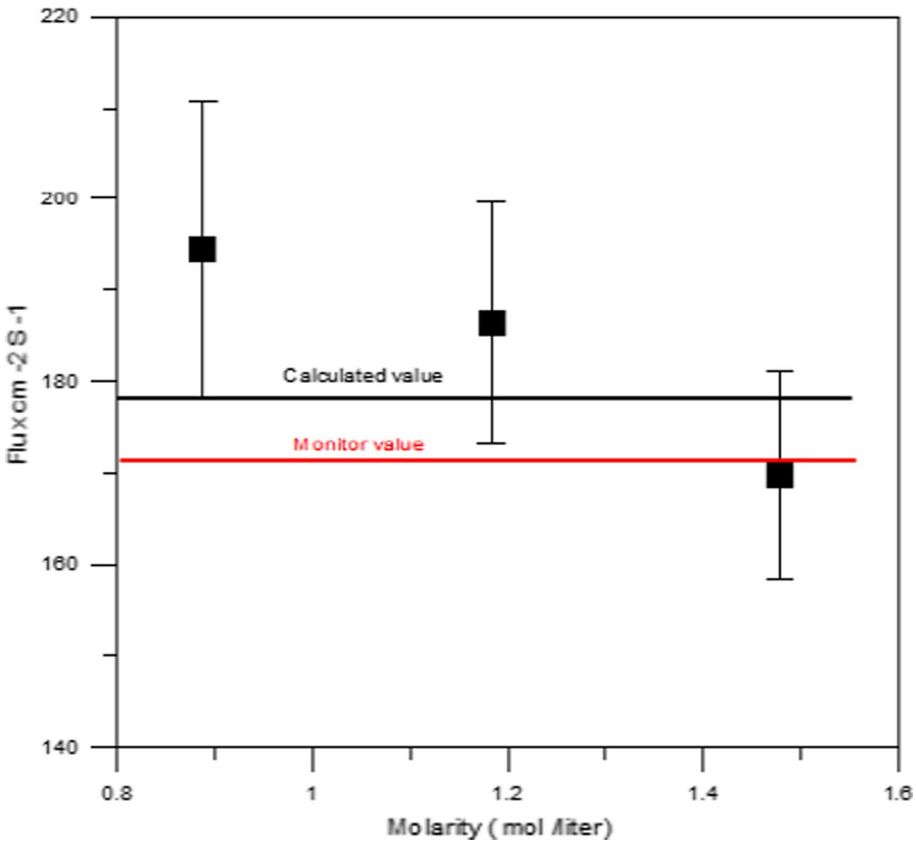

Fig. 5 Flux values as a function of molarity

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