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Conventional Fast Neutron Flux Measurement in the Radial Piercing Channel D of the TRIGA Mark II Reactor, Pavia

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ABSTRACT

Measurement of fast flux in four positions of a radial piercing channel of TRIGA Mark II nuclear reactor of Pavia was performed by means of a bare irradiation method and a Monte Carlo simulation both relying on Ni monitors. Reaction rates per target nucleus of Ni, obtained from measurement and simulation, were compared in order to verify how well the two methods behave with respect to each other.

A satisfactory agreement was observed since results were compatible within the stated uncertainties with exception of values related to the position closest to the reactor core, where a discrepancy at ten percent level was highlighted. Consequently, the Monte Carlo code used for the simulation was considered to be validated and thus, intended to be used in the forthcoming structural modifications of the channel D which include neutron shielding and thermal neutron filter.

1 INTRODUCTION

The TRIGA Mark II nuclear reactor operated by the Laboratory of Applied Nuclear Energy (LENA) of the University of Pavia is a $250\,\mathrm{kW}$ light water moderated facility aimed for isotope production, training and general purpose research [1].

One of the research fields conducted at this reactor is focused on the investigation of nuclear reactions induced by fast neutrons. The accessibility to a fast neutron beam allows a broad variety of applications, ranging from the determination of fast neutron cross section data, study of burnup and transmutation in fuel elements and effects of radiation damage in various materials; all these applications might be, in turn, beneficial for what concerns research and development of the upcoming IV generation of fast nuclear reactors [2].

In order to make available a fast neutron beam at the TRIGA reactor, the realization of a new neutron irradiation facility is planned by modifying the so-called channel D, a pre-existing radial piercing channel without reflector material. The channel D will be adapted by introducing filters to remove the neutron thermal flux component and reduce the gamma background, and a beam catcher to assure operator's safety. Characteristics and dimensions of filtering and shielding materials will be modulated according to the neutron flux spectra simulated by means of Monte Carlo Neutron Particle (MCNP6v2) software [3]. The quality of the simulated data is assured by validating the software code using experimental data collected in selected positions along the channel.

In this study, preliminary measurements of the conventional fast neutron flux in four positions of the actual (unmodified) configuration of channel D are reported. The technique adopted to investigate the flux consisted of the bare activations and gamma countings of suitable monitor elements.

2 THEORY

The measurement model, described in [4], relies on monitors that undergo threshold nuclear reactions, e.g. (n,2n), (n,p), (n,α), when irradiated with fast neutrons. Measuring the gamma emissions by the activated nuclide, a value of the intensity of fast flux is obtained. The resulting fast flux, Φ_f, is conventional since it is based on a conventional value of the monitor fission-neutron averaged cross section assuming that the shape of neutron distribution in the fast energy range follows the Watt distribution.

The Φ_f value measured by a flux monitor is obtained using the Eq. (10) of [5], here reported:

$$\Phi_\mathrm{f} = \frac{n_\mathrm{p}\,\lambda\,t_\mathrm{r}\,M}{t_\mathrm{l}\,\left(1-e^{-\lambda t_\mathrm{i}}\right)\,e^{-\lambda t_\mathrm{d}}\,\left(1-e^{-\lambda t_\mathrm{r}}\right)\,m\,\theta\,\Gamma\,\varepsilon\,\bar{\sigma}\,N_\mathrm{A}} \tag{1}$$

where, n_p is the net area of full-energy peak, λ is the decay constant, t_r is the real counting time, M is the monitor molar mass, t_l is the live counting time, t_i is the irradiation time, t_d is the decay time, m is the monitor mass, θ is the isotopic abundance of the target (isotope) monitor, Γ is the γ-yield of monitor emission, ε is the full-energy peak counting efficiency, $\bar{\sigma}$ is the fission neutron spectrum averaged cross section and N_A is the Avogadro constant.

In addition, from Eq. (1) it is possible to calculate the reaction rate per target nucleus of the Ni monitor, R, by multiplying Φ_f times $\bar{\sigma}$:

$$R = \Phi_\mathrm{f}\,\bar{\sigma} = \frac{n_\mathrm{p}\,\lambda\,t_\mathrm{r}\,M}{t_\mathrm{l}\,\left(1-e^{-\lambda t_\mathrm{i}}\right)\,e^{-\lambda t_\mathrm{d}}\,\left(1-e^{-\lambda t_\mathrm{r}}\right)\,m\,\theta\,\Gamma\,\varepsilon\,N_\mathrm{A}} \tag{2}$$

It is worth to note that the R value heavily relies on the adopted monitor but is no longer conventional because it is independent from assumptions concerning the fast neutron flux shape. Moreover, since the R value is one of the outputs of the MCNP software, it will be used to assess the quality of simulated data.

3 EXPERIMENTAL

Ni was selected as flux monitor in order to exploit the large cross section of ^{58}Ni(n,p)^{58}Co threshold reaction and the conveniently long half-life, $70\,\mathrm{d}$, of its activation product. In addition, solutions of similar volumes were prepared to easily adjust the Ni concentration with respect to the expected neutron flux and optimize the γ-emission rate resulting in an ease of the γ-acquisition process.

A $3.5483(2)\,\mathrm{g}$ mass sample of a Ni foil was dissolved in HNO_3 to obtain a final solution of $29.2931(2)\,\mathrm{g}$ with Ni mass fraction of $0.121\,13(1)\,\mathrm{g\,g^{-1}}$; here and hereafter, values in parenthesis indicate the standard uncertainty and refer to the last digit. Two aliquots of about $0.3\,\mathrm{mL}$ and $0.7\,\mathrm{mL}$ of the Ni solution were pipetted and weighted in two $3\,\mathrm{mL}$ polyethylene (PE) irradiation vials; the resulting masses of such aliquots were $0.300\,45(3)\,\mathrm{g}$ and $0.736\,11(3)\,\mathrm{g}$, respectively. The Ni solution was concentrated by evaporation in order to reach a solution mass of $18.7834(2)\,\mathrm{g}$ with Ni mass fraction of $0.182\,22(1)\,\mathrm{g\,g^{-1}}$. Two aliquots of about $2\,\mathrm{mL}$ each were pipetted in two $3\,\mathrm{mL}$ PE irradiation vials; the resulting masses of aliquots were $3.6157(3)\,\mathrm{g}$ and $3.5813(3)\,\mathrm{g}$, respectively. Deionized water was added to the PE vials to obtain homogeneous volumes among the samples with $1.6\,\mathrm{cm}$ height of liquid. The increasing concentration in Ni mass fractions among the monitor samples was clearly visible from the increasingly darker shade of their corresponding solutions. Moreover, the two most concentrated samples showed signs of crystallization due to $Ni(NO_3)_2 \cdot 6H_2O$, however the change in physical state does not affect the neutron activation process.

The four monitor vials were heat-sealed to avoid loss of content, thus, solutions with increasing Ni concentration were placed at $45\,\mathrm{cm}$, $75\,\mathrm{cm}$, $125\,\mathrm{cm}$ and $195\,\mathrm{cm}$ from the vertical axis of reactor core and fixed in their position with an Al rod. Table 1 reports the relevant information concerning the Ni samples.

Table 1: Values of volume, density and mass of Ni measured for each of the four Ni monitor sample solution; the irradiation position, indicated as nominal horizontal distance from the reactor core, is also reported.

Volume / $\mathrm{cm^3}$	Density / $\mathrm{g\,cm^{-3}}$	Ni mass / g	Distance from core / cm
2.0(1)	0.98(4)	0.0364(1)	45
2.1(1)	0.98(4)	0.0892(1)	75
2.2(1)	1.66(4)	0.6589(1)	125
2.0(1)	1.77(4)	0.6526(1)	195

The irradiation lasted 90 minutes at $10\,\mathrm{kW}$ power because the absence, during the measurement, of most part of the shielding prevented the achievement of the operational $250\,\mathrm{kW}$ power.

4 RESULTS

4.1 Measurement

After a cooling period of six days, gamma spectra of the irradiated monitor Ni samples were acquired using an ORTEC Hyper Pure Ge (HPGe) detection system whose channels were previously calibrated in energy and efficiency. Samples were located in contact with the end-cap of the detector. The net area of the $810.8\,\mathrm{keV}$ full-energy peak of the ^{58}Co was obtained by processing the recorded spectra with the HyperLab software.

The Φ_f and R results, obtained through Eq.s (1) and (2), respectively, at the experimental $10\,\mathrm{kW}$ power, were scaled by a factor 25 to $250\,\mathrm{kW}$ reactor power. The $250\,\mathrm{kW}$ scaled results ranged from $1.33(4) \times 10^{11}\,\mathrm{cm^{-2}\,s^{-1}}$ and $1.48(3) \times 10^{-14}\,\mathrm{s^{-1}}$, respectively, at $45\,\mathrm{cm}$ distance from the reactor core, to $1.32(4) \times 10^{9}\,\mathrm{cm^{-2}\,s^{-1}}$ and $1.46(3) \times 10^{-16}\,\mathrm{s^{-1}}$, respectively, at $195\,\mathrm{cm}$. The major contribution to the $3\,\%$ and $2\,\%$ relative standard uncertainties of Φ_f and R values were the uncertainty of $\bar{\sigma}$ and the uncertainty of ε (mainly due to sample positioning variability), respectively.

4.2 MCNP Simulation

Since the latest available MCNP modellization of the TRIGA Mark II reactor of Pavia was outdated due to rearrangements of fuel rods, a new kcode was created to take into account these position variations of the rods and the fuel burn up (but not the poisoning due to the burn up). The code was validated in the Central Channel of the reactor. Since the objective of this work was channel D we aimed to validate the new code also in the channel of interest.

Targets with the same properties of Ni monitor samples (volume, elemental composition and density as reported in Table 1) were simulated at the four nominal positions along the channel D. The number of fission neutrons were simulated being a satisfactory compromise between statistical uncertainty and computation time. The quantity returned by the simulation was the production rate for the reaction ^{58}Ni(n,p)^{58}Co for the Ni monitors.

The simulated R results at $250\,\mathrm{kW}$ reactor power, obtained dividing the production rate by the total amount of Ni atoms in the target, ranged from $1.66(3) \times 10^{-14}\,\mathrm{s^{-1}}$ at $45\,\mathrm{cm}$ distance from the reactor core, to $1.7(2) \times 10^{-16}\,\mathrm{s^{-1}}$ at $195\,\mathrm{cm}$. The statistical uncertainty associated with the results increased according to the distance from the reactor core, from $2\,\%$ to $11\,\%$ due to the decreasing amount of simulated neutrons that reached the farthest positions.

4.3 Discussion

Results of experimental measurement and MCNP simulation are reported in Figure 1 and Table 2.

The two orders of magnitude decrease in flux and reaction rate along the $150\,\mathrm{cm}$ horizontal distance was in agreement with the expectations considered in phase of sample preparation and based on previous knowledge of the facility. Moreover, the conventional fast flux at $45\,\mathrm{cm}$ distance is comparable with a previous measurement performed in another irradiation channel (LS-27 in [5]) placed in the close vicinity of that position.

The agreement between the simulated and experimental results was satisfactory with discrepancies ranging from a minimum of $0.8\,\%$ at $75\,\mathrm{cm}$ to a maximum of $18.7\,\%$ at $195\,\mathrm{cm}$ that are justified by the reported uncertainties except for the datum at $45\,\mathrm{cm}$ position.

It is worth to note, however, that all the simulated R values are higher than the experimental R values, thus we cannot exclude the presence of some sort of slight offset due to an

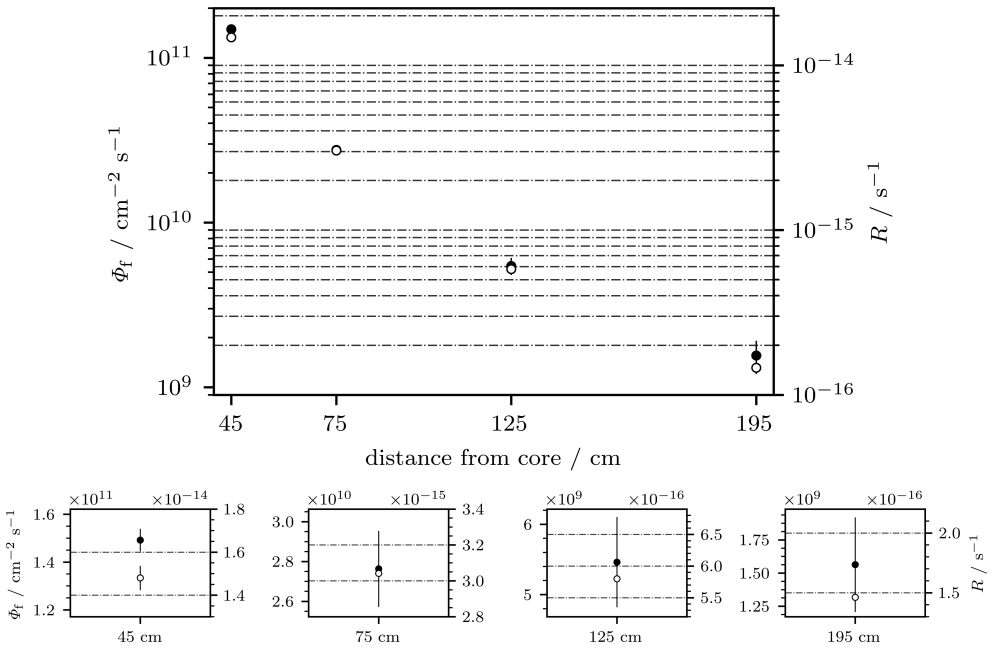

Figure 1: Values of fast flux, Φ_f, and Ni reaction rate per target nucleus, R, scaled at $250\,\mathrm{kW}$ and measured in four positions of channel D with respect to the center of the reactor core. White and black circles represent the resulting values obtained with the experimental measurement and the MCNP simulation, respectively. Expanded uncertainties ($k = 2$) are plotted. The four subplots at the bottom display magnifications of resulting values for each measured position.

overlooked or unknown cause. The offset might derive from possible aspects producing flux perturbations that were neglected. In particular, the presence of the Al rod was not accounted for during the simulation as well as the thermalizing effect of water in the solutions, with subsequent decrease of the apparent fast flux, was overlooked in the experimental measurement model. A further computation is planned to be performed in order to evaluate whether the Al rod might produce sensible flux perturbations by contributing to the offset, and particularly to the discrepancy highlighted for the monitor placed at $45\,\mathrm{cm}$.

In addition, further studies are required to fully understand whether the presence of water is affecting the fast flux and to what extent. Although an experiment was performed to evaluate the self-shielding effect for a target in solution with respect to the metallic foil, the obtained

Table 2: Values of measured conventional fast flux, Φ_f, and measured and simulated reaction rate per target nucleus of Ni, R, recalled at the four investigated positions. the column "R discrepancy" indicates the relative discrepancies of the simulated R values with respect to the measured ones.

Distance from core / cm	Measurement Φ_f / cm^{-2} s^{-1}	Measurement R / s^{-1}	Simulation R / s^{-1}	R discrepancy
45	$1.33(4) \times 10^{11}$	$1.48(3) \times 10^{-14}$	$1.66(3) \times 10^{-14}$	11.9 %
75	$2.74(9) \times 10^{10}$	$3.04(6) \times 10^{-15}$	$3.1(1) \times 10^{-15}$	0.8 %
125	$5.2(2) \times 10^{9}$	$5.8(1) \times 10^{-16}$	$6.1(4) \times 10^{-16}$	4.5 %
195	$1.32(4) \times 10^{9}$	$1.46(3) \times 10^{-16}$	$1.7(2) \times 10^{-16}$	18.7 %

results, albeit satisfactory, were unreliable due to inadequate representation of the study performed in this work. A negligible relative difference in flux, evaluated through the specific count rate, within $3\,\%$ uncertainty level was observed, however, a Au target was used instead of Ni and the activation went through a thermal reaction (^{197}Au(n,γ)^{198}Au) instead of a threshold one. Moreover, a light-water moderated thermal reactor was adopted in this test while channel D is a fast neutron facility without moderator material.

5 CONCLUSIONS

In this work the neutron fast flux was measured with Ni monitors in four positions along the channel D of TRIGA Mark II reactor as well as the production rate per target nucleus for the ^{58}Ni(n,p)^{58}Co reaction; these results were compared with the outcome of a MCNP simulation in order to validate the adopted code.

While for the closest monitor to the reactor core a deeper investigation is required, good agreement was highlighted otherwise yielding a positive outcome concerning the validation of MCNP simulation. The code adopted in this comparison will be useful for further simulation concerning the TRIGA Mark II reactor behavior and in particular, for modeling the future structural modifications on channel D such as redesign of neutron shielding and introduction of the filter for thermal neutrons.

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