Validation of calculational determination of ^{18}O(p,n) secondary neutron field

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Introduction

The medical cyclotrons intended to produce medical isotopes are relatively widespread. Nowadays, it is popular to place small and compact accelerators directly to hospitals. This approach simplifies handling with produced radiopharmaceuticals, but it imposes radiation safety measures during production. Radiation protection issues are gaining on importance especially as production increases on current cyclotrons which leads to higher radiation loads than originally designed. For optimal utilization of isotope production cyclotron, the exact knowledge of leakage neutron field is essential due to the deep penetration ability of the high energy neutrons and accompanied secondary radiation production. Our paper presents measurement of neutron leakage spectra in various angles from open target assembly located in the research laboratory of the Czech Academy of Sciences. These spectra are compared with data obtained from a compact medical cyclotron IBA Cyclone 18/9 in the UJV Rez measured previously [4] [5] and also with activation measurements of reactions with different threshold energies. All data were also compared with calculation in different Monte Carlo codes with both models and data libraries. The preliminary results show significant disagreement between experiments and theoretical predictions. These findings could have implications not only to the nuclear data community but also to the production accelerators operators at the licensing stage.

Methodology

Spectra of secondary neutrons produced by the reaction of 18 MeV protons in a water target enriched in ^{18}O were measured at different angles on the cyclotron U-120M at Nuclear Physics Institute of CAS. The data obtained were compared with previously obtained data measured at the IBA Cyclone 18/9 production cyclotron. A comparison of the two target systems modelled in Geant4 is shown in Fig. 1. The neutron spectra were measured by the organic scintillator coupled with fast two-parameter spectrometric system NGA-01 [7] equipped with an active voltage divider. Pulse Shape Discrimination unit is than used to distinguish the type of the detected particle by analyzing the pulse shape, whereas particle energy is evaluated from the integral of the whole response. Acquired recoiled proton spectra are then subjected to the deconvolution by the Maximum Likelihood Estimation.

In addition to the spectra measurements, measurements of the reaction rates in activation foils of aluminium and nickel were also performed. These were irradiated at the same angles at which the spectra were measured, at distances of 100 mm to determine the spectral indices and 269 mm to determine the absolute values of the reaction rates. The activation films were then transferred to a well-characterized HPGe detector whose efficiency is determined by a computational model.

All data were also simulated using the multipurpose Monte Carlo codes Geant4 [1] and MCNP6.2 [6]. In Geant4, two approaches were used to simulate the proton reaction. Both the default model with the Bertini cascade through the Shielding reference physics list and the TENDL-2019 library data [3] implemented using the QGSP_BIC_AllHP physics list were used. In MCNP6.2, the default model CEM03.03 was used.

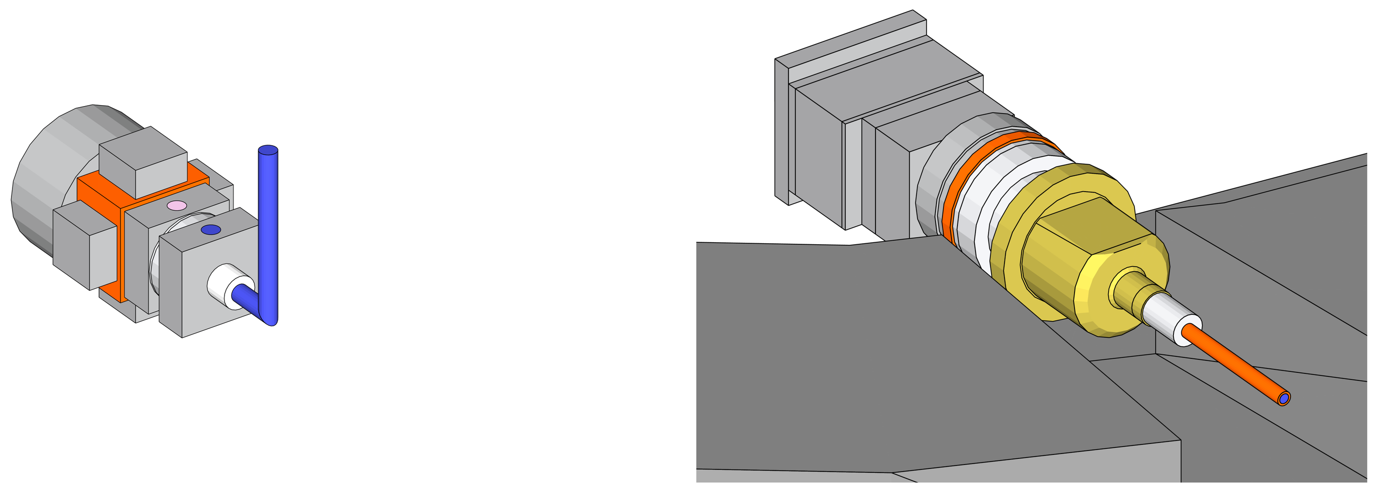

Fig. 1: Comparison of target assemblies at the cyclotron U-120 M at NPI (left) and at the cyclotron IBA Cyclone 18/9 at UJV (right)

Neutron spectra

In Fig. 1 a comparison of neutron spectra in 1 m for different target configurations can be seen. As can be seen, they all exhibit a similar shape, which roughly corresponds to the evaporation spectrum. Compared to the spectra from the IBA Cyclone 18/9 facility, the open-geometry spectrum at U-120M has a smaller fraction of low-energy neutrons because the target is smaller and there is no shielding where neutrons can be slowed down.

In Fig. 2, the spectra in 0° and 90° measured on the U-120M cyclotron are compared with spectra from the EXFOR database obtained in an experiment at the Japanese TIARA facility [2]. The spectrum in 0° is compared with the spectrum from 15° and is significantly lower, since the target in the second experiment was much less massive and the neutrons are heavily scattered by the water cooling located in 0° direction. At 90° there is already a fairly good agreement between these two spectra. The measured spectrum is also compared with the calculated spectra, where a strong underestimation of the spectrum can be observed for the models in Geant4. A similar pattern for spectra calculated with MCNP6.2 and TENDL-2019 can be seen, which overestimate the spectrum at lower energies and underestimate it at higher energies.

Graphs

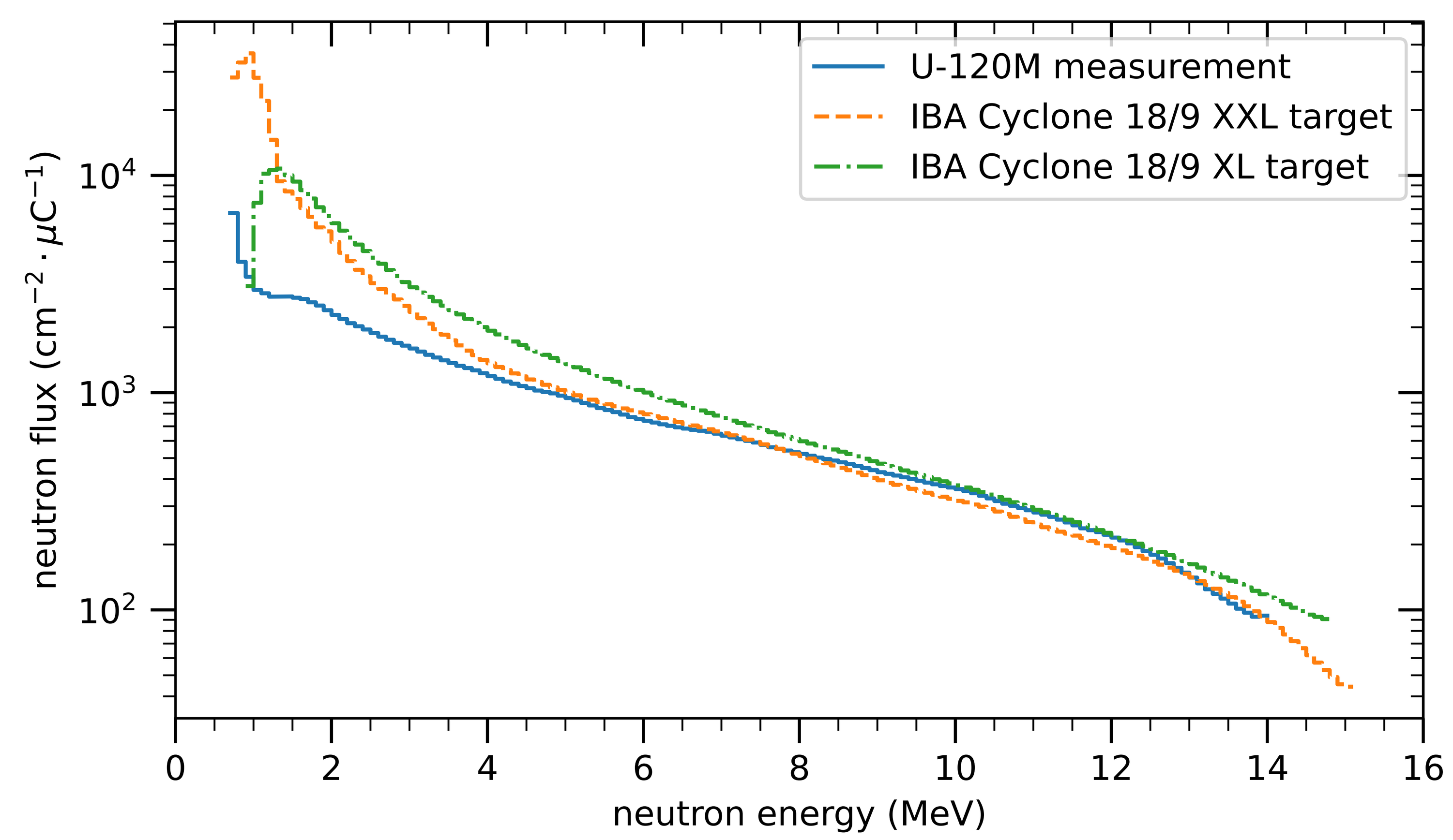

Fig. 2: Comparison of neutron spectra in 1 from the target in straight direction for different target assemblies

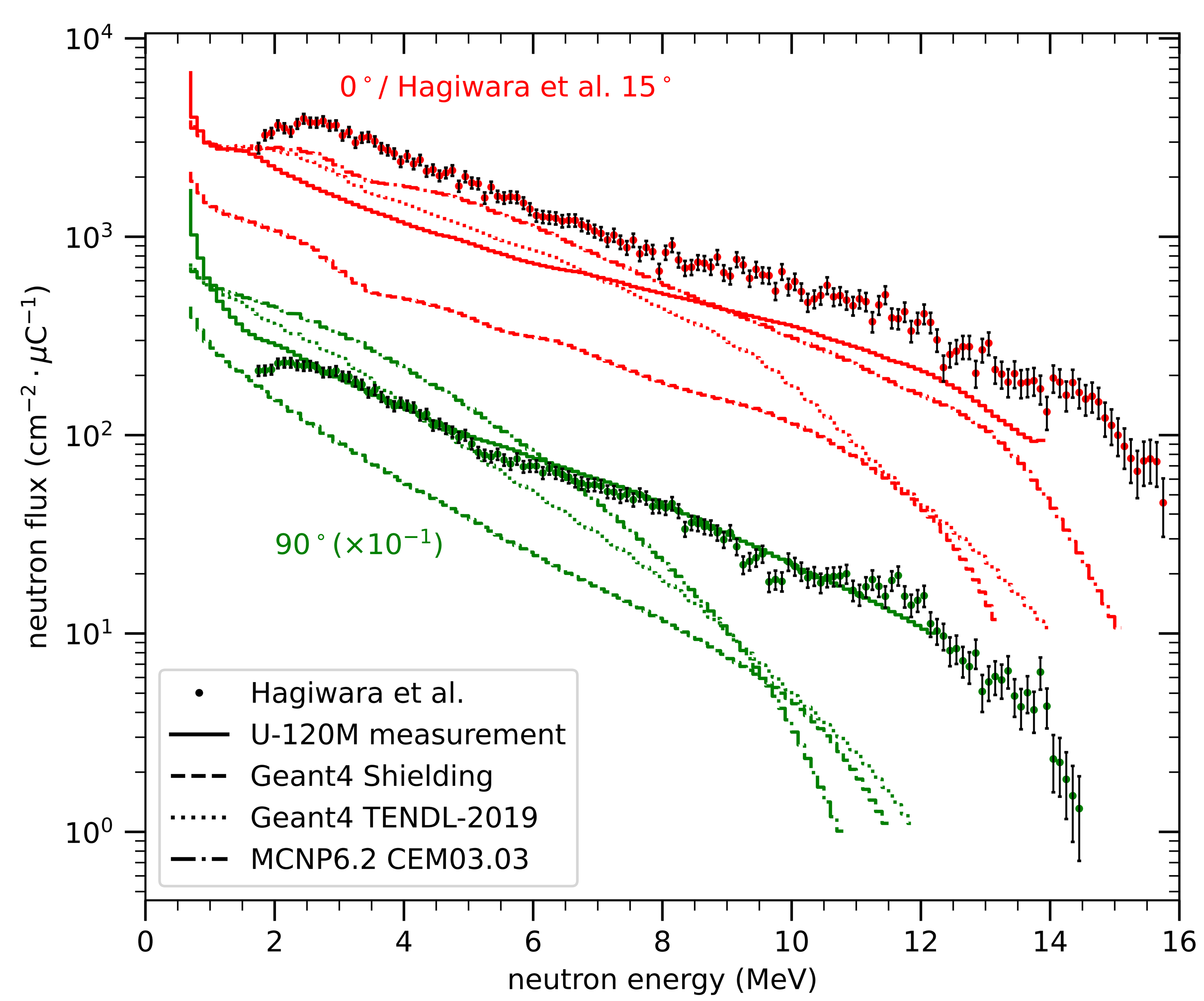

Fig. 3: Comparison of neutron spectra in different angles with EXFOR database (Hagiwara et al.) and calculations in various Monte Carlo codes with both models and data libraries

Reaction rates

Table 1 shows the absolute values of the measured reaction rates and their comparison with the calculated ones. A reasonably good agreement of the calculated reaction rates using the measured neutron spectra can be seen for reactions with different energy thresholds. The Geant4 calculation using the physic models strongly underestimates all reaction rates as was seen in the comparison of the spectra. The calculation using TENDL-2019 predicts the reaction rates of ^{58}Co production reasonably well, but underestimates the reaction rates with higher energy thresholds. The calculation in MCNP6.2 then even overestimates reaction rates with lower energy thresholds and underestimates those with higher ones.

Comparison of reaction rates

Tab. 1: Reaction rates normalized per one proton in distance 269 mm from the target in different angles and their comparison in the mean of C/E-1 with reaction rates calculated in different codes and reaction rates calculated with measured neutron spectra.

angle	reaction	reaction rate	stilbene measurement	Geant4 Shielding	Geant4 TENDL-2019	MCNP6.2 CEM03.03
0°	Ni(n,x)^{58}Co	5.00E-32	2.7%	-62%	-5.0%	25%
	Al(n,x)^{24}Na	4.60E-33	-15%	-77%	-58%	-27%
	Ni(n,x)^{57}Co	2.50E-33	0.1%	-86%	-72%	-12%
90°	Ni(n,x)^{58}Co	4.50E-32	7.9%	-63%	-18%	17%
	Al(n,x)^{24}Na	2.35E-33	-3.7%	-80%	-71%	-71%
	Ni(n,x)^{57}Co	5.28E-34	4.3%	-88%	-74%	-92%

Spectral indices of reaction rates with different energy thresholds can be suitably used to compare the shape of the spectra. The ratio of the reaction rates of formation of ^{58}Co and ^{57}Co in nickel is well suited for this purpose. In Tab. 2, one can see how this ratio increases from 0° to 90° due to the loss of fast neutrons originating from the reaction kinematics. It can be seen that the spectral index calculated using data from the EXFOR database is quite strongly underestimated, indicating a lower yield of high energy neutrons. With the measured spectra, the spectral indices are in agreement within 10%, which is a very good result. As seen with the spectra comparison in Fig. 3, in addition to the overall underestimation of the spectra, Geant4 also underestimates the yields of the highest energies, as seen with the overestimation of the spectral indices. As for MCNP6.2, it shows a strong anisotropy of the high energy neutron production, while for 0° it overestimates the spectral index by only 40%, for 90° it is already over 1200%.

Tab. 2: Spectral index ^{58}Co/^{57}Co calculated in 100 mm from the target and its comparison in the mean of C/E-1 with calculations in different codes and with measured neutron spectra.

angle	spectral index	Hagiwara et. al	stilbene measurement	Geant4 Shielding	Geant4 TENDL-2019	MCNP6.2 CEM03.03
0°	21.0	-39%	-2.3%	134%	276%	40%
90°	80.4	-56%	9.6%	215%	215%	1277%

Remarks

Characterization of the secondary neutron spectrum produced during the production of the radionuclide ^{18}F at two different angles has been performed using a stilbene spectrometer with accompanying measurements of the reaction rates. The possibility of measuring such spectra with the NGA-01 spectrometer set has been demonstrated. Furthermore, the inconsistency of the EXFOR data with the measured reaction rates and their overestimation of high energy neutrons was shown. Discrepancies were also shown for data calculated by simulation programs, where none of the models provide satisfactory results for all energies and angles. Among other things, a strong underestimation of the neutron yield for the physics models in Geant4 was shown.

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