

## Comparison between measurement and calculations for a 14 MeV neutron water activation experiment

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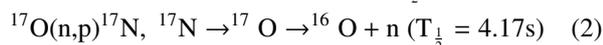
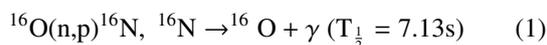
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**Abstract.** The nuclear heat loads due to gamma rays emitted from the decay of <sup>16</sup>N and delayed neutrons from <sup>17</sup>N, generated by the activation of water in cooling circuits, are critical for ITER design. The assessment of nuclear heating from activated water is complex; it requires temporal and spatial dependent transport and activation calculations taking into account variation of irradiation, water flow conditions and cooling circuits' parameters. A water activation experiment has been recently conducted at the 14 MeV Frascati Neutron Generator (FNG) in order to validate the methodology for water activation assessment used for ITER and to reduce the safety factors applied to the calculation results, which have a large impact on the schedule, commissioning and licensing. Water circulating inside an ITER First Wall (FW) mock-up was irradiated with 14 MeV neutrons and then measured using a large CsI scintillator detector. The system consists of a closed water loop where the cooling water, transiting through an ITER FW mock-up, is irradiated by FNG. The induced <sup>16</sup>N activity via 14 MeV neutrons interactions with <sup>16</sup>O via the <sup>16</sup>O(n,p)<sup>16</sup>N reaction is measured in a dedicated counting station via an expansion volume. The water then passes to a much larger holding delay tank, and after several <sup>16</sup>N half-lives decay time, it is then recirculated and exposed again to neutrons in the ITER First Wall (FW) mock-up. The measured <sup>16</sup>N activity is obtained measuring the emitted characteristic 6.13 and 7.12 MeV gamma-rays. Calculations were performed in an accurate model of the FW mock-up using the MCNP Monte Carlo code and FENDL-3.1 nuclear data library to obtain the predicted flux impinging on the water. The EASY-2007 inventory code was used to predict the <sup>16</sup>N activity. In this work, a comparison between measurements and calculations is reported together with associated uncertainty analysis.

### 1 Introduction

The water coolant in ITER components such as those inside the first wall, blanket modules, divertor cassettes and vacuum vessel will become activated by neutrons during DT plasma operations. Two key neutron induced reactions will occur with oxygen producing the nitrogen radioactive isotopes <sup>16</sup>N and <sup>17</sup>N through the following reactions:



Because water coolant is being pumped and transported to other locations, the decay emissions from these nuclides will induce nuclear loads in sensitive tokamak and plant components, e.g. nuclear heat in superconducting magnets, absorbed doses in polymer-based components such as valves, or high dose rates in electronics. The uncertainty in the calculation of radiation maps due to activated water is evaluated to be very large [1], the main sources of uncertainty being due to modelling and nuclear data, and hence safety factors between 8.2 and 4.7 are applied. The motivation for this new experiment is to accurately measure the

water activation in an ITER like environment with the aim to provide a scientific justification to reduce these safety factors. The European Agency Fusion for Energy (F4E) has tasked ENEA to measure the <sup>16</sup>N and <sup>17</sup>N production in water for an ITER first wall (FW) mock-up and to compare the experimental results with calculations to help reduce the uncertainties. ENEA sub-contracted UKAEA the task to conduct the pre- and post-experimental analysis. The results for <sup>16</sup>N are presented in this paper.

### 2 Experimental set-up

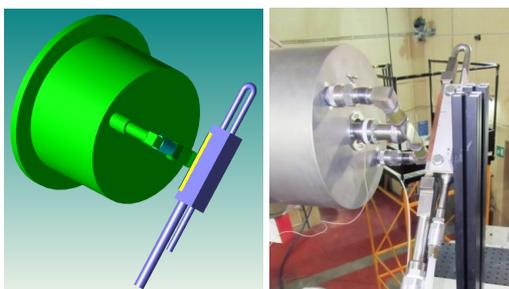
The ITER first wall mock-up component was placed 5 cm from the Frascati Neutron Generator (FNG) 14 MeV neutron source target [2] and connected to a water circuit of about 40 m in length. A high flux-prevalence pump (a model type that is frequently used in artesian wells), immersed in a tank of about 100 litres, sent the water through a plastic tube of internal diameter 2.8 cm to the FW panel mock-up. The water, after circulating in the panel and becoming neutron activated, returns to the tank through a 1.1 cm internal diameter plastic tube. One meter before reaching the tank the water is passed inside an in-line expansion

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tank located in-front of a CsI gamma-ray detector. The purpose of the thinner tube is, for the same water flux, to reduce the transit time needed to reach the gamma-rays detector while the expansion tank is to increase the total activity that is present in the region of the detector and hence the count rate by increasing the volume of activated water seen by the gamma-ray detector. The volume of the water expansion tank, a cylinder of about 11 cm diameter and 5 cm height was chosen, based on pre-experimental analysis, equal to about 165 cm<sup>3</sup>. The expansion tank material was aluminium with a maximum wall thickness of 0.5 cm to support the water pressure.

### 3 Neutron transport calculations

A highly detailed MCNP5 [3] model of the FW panel mock-up, the FNG target and also of the bunker wall and the main structures present in the hall was prepared. In figure 1 the MCNP model of the FW panel in front of FNG and a corresponding photo is shown. The materials of the



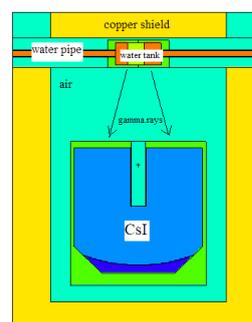
**Figure 1.** The FW panel mock-up in front of FNG. (LHS) the MCNP model, and (RHS) a photo of the real experiment.

FW panel are the following: ordinary stainless steel for the tubes, AISI316LN for the body and CuCrZr for the part facing FNG target. The composition of these materials was provided by F4E. The detailed MCNP model of the FNG and FW panel was used to calculate the neutron flux in the water flowing in front of target. The neutron flux was calculated with an F4 tally across several water volume regions represented in the FW panel model. The activation calculations were performed in the energy range 1.0<sup>-10</sup>-19.61 MeV using the VITAMIN-J 175 neutron energy group structure. The characteristic neutron energy versus angle emission of the D-T fusion reaction, accurately described with the FNG source routine [4] was used during the Monte Carlo calculation. The cross section database used for neutron transport calculation was FENDL-3.1 [5].

### 4 Gamma-rays detector modelling, set-up and calibration

The gamma-rays detector used was a large CsI scintillator of about 25 cm diameter and 20 cm height coupled to a photo-multiplier tube using a glass light guide. In order to increase the volume of the activated water seen by the CsI detector, the cylindrical water expansion tank was

placed in front of the detector with a distance of 13.5 cm between the bottom surface of the tank and the top surface of CsI and this geometry was accurately modelled in the MCNP model. The CsI detector was placed behind a 1 m shielding wall made of ordinary concrete and was located at about 15 m distance from the FNG neutron source and shielded via a large copper cylinder 5 cm thick surrounded by 10 cm of lead. The response to the energetic gamma-rays produced by <sup>16</sup>N decay (most intense gamma ray, 6.13 MeV, branching probability 68.8%) was obtained modelling the CsI detector with MCNP6 [6] and calculating its response using an F8 pulse height tally. The MCNP model, which includes the water expansion tank, is shown in figure 2. The gamma-ray detection structure (geometry



**Figure 2.** MCNP model of the gamma-rays detection system. The lead shield was not modelled.

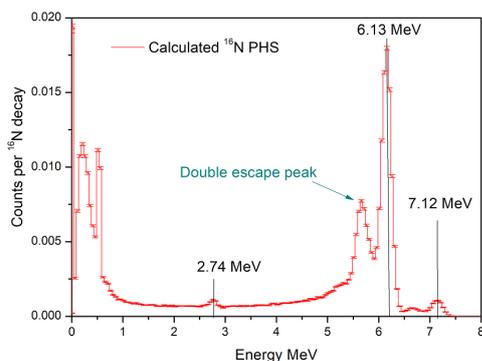
and materials) is well known, allowing a reliable Monte Carlo simulation model to be constructed. The MCNP6 model of the gamma-ray detection system was validated performing a comparison between experimental and computed detector efficiency at different gamma-ray energies. Three different gamma-ray radioactive nuclides were used: a point-like certificated source of <sup>137</sup>Cs (uncertainty ±5%) and two radioactive sources (disks of diameter 15 mm and thickness 1 mm) obtained by 14 MeV neutron activation of Al (<sup>24</sup>Na) and Ni (<sup>58</sup>Co). These three radionuclides were first measured with the HPGe gamma detector available at the FNG laboratory. This HPGe has a 60% relative efficiency and is absolutely calibrated with an uncertainty of ±3%. The activity of the foils counted with the CsI was obtained measuring the total counts in the photon peak area and the CsI efficiency calculated with MCNP6 using a pulse height tally (F8). The activity measured by the CsI detector was compared with the activity measured with the HPGe. Identical values of the gamma lines branching ratio have been used to compute the activity from CsI and HPGe. The results are gathered in table 1. The calculated

**Table 1.** Comparison of radionuclides activity measured with CsI and HPGe detector. The estimated uncertainties, one sigma, are indicated in the brackets

| Radionuclide (γ line)     | Activity CsI-MCNP6 | Activity HPGe | Dev. (%) |
|---------------------------|--------------------|---------------|----------|
| <sup>137</sup> Cs 661 keV | 2990 (5.0%)        | 2830 (2.7%)   | +5.6%    |
| <sup>58</sup> Co 810 keV  | 9569 (5.0%)        | 9230 (3.0%)   | +3.6%    |
| <sup>24</sup> Na 1368 keV | 67700 (5.0%)       | 72700 (4.0%)  | -6.8%    |
| <sup>24</sup> Na 2754 keV | 69700 (5.0%)       | 72700 (4.0%)  | -4.1%    |

pulse height tally of the gamma detector produced per <sup>16</sup>N

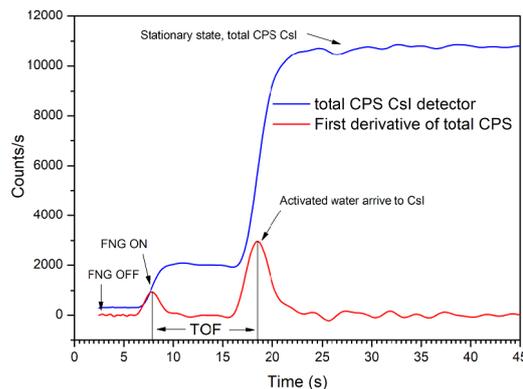
decay is shown in figure 3. The branching ratio of the three emitted gammas was taken from the EAF-2007 database (see later); 2.74 MeV, 0.0893%; 6.13 MeV, 68.8%; 7.12 MeV 5.0% respectively. The experimental measured energy resolution of the CsI was introduced in the calculation with the F8 tally as a Gaussian Energy Broadening (GEB) function. This feature is available in the MCNP code and GEB card fitting parameters were estimated from experimental data.



**Figure 3.** MCNP calculated  $^{16}\text{N}$  Pulse Height Spectrum (PHS) in the CsI detector

## 5 Experimental procedure

The experiment was carried out in the following way. Irradiations and measurements were performed using nine different water flow rates. The water flow was varied by changing the frequency of an inverter, which supplies the mains to the pump. Firstly, the water circulation was started by turning on the water pump at a fixed frequency. Then, once steady-state conditions were reached, the FNG neutron production was switched on by deviating the deuterons beam from the dump onto the target. The beam was previously prepared to provide the most possible stable neutron rate output. Typical yields used were  $(1.3\text{-}1.5)^{10}$  neutrons/s for a fixed duration of 150 seconds. When the FNG accelerator produces neutrons, a very evident background signal is measured by the gamma detector. However, this background signal is markedly lower with respect to the signal produced by  $^{16}\text{N}$  when the activated water reaches the Water Expansion Tank (WET) in front of CsI detector. The time required by the water to pass from inside the FW panel to reach and fill the WET is derived from the profile of the count rate vs time. The time trace of the count rate (CPS) was recorded with a digitizer CAEN (model DT5724). The methodology to obtain the Time Of Flight (TOF) of the water leaving the FW panel and reaching the WET is indicated in the figure 4. The linear dimensions and the volumes of all the water circuit components were accurately measured, thus the experimental measured water flow rate has been obtained dividing the sum of the volumes of the water from the FW panel by the TOF. These volumes are the plastic water pipe volume (internal diameter 1.1 cm, length 1740 cm) and the WET volume ( $165.4\text{ cm}^3$ ). Using the experimental water



**Figure 4.** Plots indicate the derivative methodology used to obtain the TOF. Blue curve shows the total CPS measured in the CsI detector; Red curve (scale not shown) is the first derivative of the blue curve. The TOF is derived from the time between the two maxima.

flow rate also the transit time of the water in the FW panel was derived with the formula.

$$\text{Transit time} = \frac{1}{\left(\frac{\text{water flow}}{0.322}\right)} \quad (3)$$

The  $322 \pm 2\text{ cm}^3$  value is the measured volume of the water which fills the FW panel mock-up.

## 6 Activation calculation

The activation calculations were performed using EASY-2007 code system [7]. The European Activation System (EASY), developed and maintained at Culham Centre for Fusion Energy, is an international standard for data on activation–transmutation caused by nuclear reactions. EASY-2007, which use the activation cross sections and decay data included in the European Activation File EAF-2007 is not the most recent and updated code system but it is however an extensively validated code system [8]. The inputs which must be provided to the code are: a neutron spectrum, usually in the VITAMIN-J 175 group structure for 14 MeV neutron irradiation; the composition of the irradiated material, water in this case, the irradiation time and the decay time at which it is necessary to know the residual activation. The main output is the activity in Bq/kg of the dominant nuclides. In this experiment the irradiation time is the transit time of the water in the FW panel and the decay time is the time derived by TOF, both dependent on the water flow rate.

## 7 Results and discussion

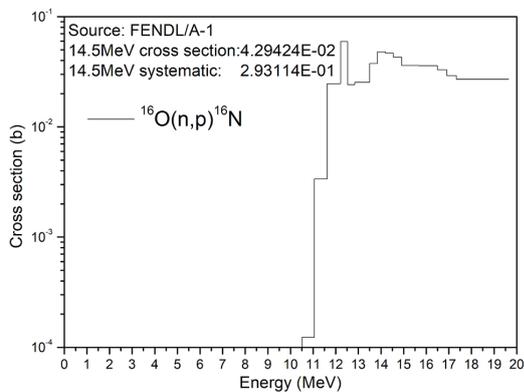
The result of this experiment, which consists of a Calculation versus Experiment (C/E) comparison is gathered in table 2. The “Experimental CsI CPS” in table 2 is obtained from the count rate in the PHS for a region of interest (ROI) which extends from 4 to 7.5 MeV. The “Calculated CsI CPS” is obtained multiplying the EASY-2007 calculated activity of the  $^{16}\text{N}$  radionuclide by the CsI efficiency

**Table 2.** C/E for different water pump speed corresponding to different water flow rates. The various input values that have been used in the EASY-2007 calculations are also indicated.

| Pump Freq. (Hz) | Transit time mock-up exp. | TOF (s) Unc. ( $\pm 0.1$ ) | Water Flow Exp. (l/min) | Calc. Activity $^{16}\text{N}$ (Bq) | CsI efficiency         |      | C/E   | Error $\pm$ |
|-----------------|---------------------------|----------------------------|-------------------------|-------------------------------------|------------------------|------|-------|-------------|
|                 |                           |                            |                         |                                     | Energy range 4-7.5 MeV | CPS  |       |             |
| 10              | 1.894                     | 10.7                       | 10.2                    | 6.31E+04                            | 3.71E-02               | 3172 | 0.990 | 0.119       |
| 15              | 1.186                     | 6.7                        | 16.3                    | 6.03E+04                            | 3.71E-02               | 3030 | 1.009 | 0.120       |
| 20              | 0.885                     | 5.0                        | 21.8                    | 5.38E+04                            | 3.71E-02               | 2706 | 1.037 | 0.121       |
| 25              | 0.708                     | 4.0                        | 27.3                    | 4.79E+04                            | 3.71E-02               | 2406 | 1.043 | 0.122       |
| 30              | 0.566                     | 3.2                        | 34.1                    | 4.16E+04                            | 3.71E-02               | 2094 | 1.022 | 0.123       |
| 35              | 0.496                     | 2.8                        | 39.0                    | 3.81E+04                            | 3.71E-02               | 1914 | 1.027 | 0.124       |
| 40              | 0.443                     | 2.5                        | 43.7                    | 3.51E+04                            | 3.71E-02               | 1765 | 1.009 | 0.125       |
| 45              | 0.389                     | 2.2                        | 49.6                    | 3.18E+04                            | 3.71E-02               | 1599 | 1.000 | 0.127       |
| 50              | 0.354                     | 2.0                        | 54.6                    | 2.96E+04                            | 3.71E-02               | 1487 | 1.016 | 0.129       |

and dividing by the total branching ratio of the 6.13 MeV and 7.12 MeV gamma-ray emissions, i.e. 73.8%. The uncertainties considered for the evaluation of the errors in the C/E are the following:

- $\pm 10\%$  as reported by the output of EASY-2007 (cross section and half-life of  $^{16}\text{N}$ );
- $\pm 5\%$  uncertainty in efficiency for the gamma-rays detector calibration and modelling;
- $\pm 4\%$  for FW panel neutron flux evaluation, due only to FNG yield uncertainty, since the Monte Carlo statistical errors are negligible;
- $\pm 0.9\%$  up to  $5\%$ , depending on the water circulation speed, for the determination of TOF due to the fixed time resolution uncertainty of  $\pm 0.1$  seconds used to analyse the experimental data;
- $\pm 0.6\%$  uncertainty from the FW panel water volume.



**Figure 5.**  $^{16}\text{O}(n,p)^{16}\text{N}$  cross section value used in EASY-2007 for our calculations.

All of these uncertainties are at  $1\sigma$  and have been summed in quadrature. The statistical error of the counts recorded with CsI is negligible since all the irradiations last for 150 seconds with typical count rate  $> 1000$  cps. The FNG neutron yields were all around  $(1.3-1.5) \cdot 10^{10}$  n/s but the reported results are all normalized to  $1.0 \cdot 10^{10}$  n/s so that comparisons can be made. All of the C/E results are very close to 1 with deviations less than the overall estimated uncertainties. This overall result indicates the good quality of the measurements performed and of the data in the EAF-2007 database. In figure 5 a plot of the  $^{16}\text{O}(n,p)^{16}\text{N}$

cross section used in EASY-2007 is reported. This cross-section has a Quality score of 6 in EASY-2007 [8]. The Quality score is a value from 0 to 6 indicating the degree to which the EAF data are backed up by experiment. 6 = agreement between differential and integral data (validated).

## 8 Conclusions

The experiment described permits to determine the accuracy in the prediction of the  $^{16}\text{N}$  radionuclide by 14 MeV neutron activation of water in a medium complex configuration, where the neutron transport in a FW mock-up has been modelled and calculated. The result of the C/E comparison indicates the very good quality of the comparison since the values of the C/E, determined in 9 different conditions, are all close to unity. This represents an interesting result also in view of the evaluation of the “safety factor” to be introduced in the ITER design to mitigate the effect of the water activation produced by the 14 MeV neutrons.

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