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High accuracy tritium measurement for the verification of the tritium production rate calculations with MCNPX

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ABSTRACT

This paper presents high accuracy tritium production rate measurement results compared with calculations using the MCNPX Monte Carlo particle transport code. The experimental results are regarded as reference values for a new passive technique based on the secondary charged particle activation method developed for measuring the tritium production rate in the test blanket modules of the ITER Tokamak. The $^{16}O(t,n)^{18}F$ reaction, which is one of the possible tritium monitor reactions, was also extensively investigated, and the experimentally determined reaction rates were compared with simulations.

Li_2CO_3 solution was filled and sealed into quartz ampoules which were irradiated in the Training Reactor of the Budapest University of Technology and Economics. The amount of ^{18}F was determined using γ-spectroscopy. Then the precise tritium measurements were carried out in the Hertelendi Laboratory of Environmental Studies using the ^3H–^3He ingrowth method, where the ^3He produced during the storage time is measured by a static noble gas mass spectrometer (VG-5400).

The HT/HTO ratio in the irradiated aqueous solutions was found to be 0.1323 ± 0.0034. Based on the comparison of the measurements and the simulations it was pointed out that the model calculations underestimate the reaction rate of both the $^6Li(n,t)\alpha$ and the $^{16}O(t,n)^{18}F$ reactions by 5–10% and 15%, respectively.

1. Introduction

The tritium production rate (TPR) measurement plays an important role in the investigation and testing of the Test Blanket Modules (TBM) [1] of the ITER Tokamak [2,3]. One of the possible techniques is based on the secondary charged particle activation (SCPA) method [4] where a charged particle is produced in a neutron induced reaction (primer activation) which is able to cause so-called secondary activation. Based on the activity of the product of the secondary reaction (monitor) one can have an estimate on the amount of the produced charged particles. Applying this technique in case of TBM the tritons[1] coming out from the primer activation of lithium cause triton induced reactions. The activity of the monitors produced in these reactions is proportionate to the amount of the produced tritium.

The results of a comprehensive investigation and experimental tests concerning this passive diagnostic method have been already presented in Ref. [5]. The present work aims for both the verification of the TPR calculations and the calibration of the SCPA method in case of the $^{16}O(t,n)^{18}F$ reaction for TPR measurement. Practically the latter means the determination of the ^{18}F/tritium ratio. These efforts are essential for designing high accuracy diagnostic methods.

In the experiments a small amount ($\sim 150\ \mu l$) of aqueous solution of Li_2CO_3 was put and sealed into quartz ampoules and irradiated in the Training Reactor of the Budapest University of Technology and Economics (BME), which is a thermal reactor. Then the amount of ^{18}F (β^+-decay) produced through the $^{16}O(t,n)^{18}F$ reaction was determined with γ-spectroscopy. After that the samples were transported to the Hertelendi Laboratory of Environmental Studies (HEKAL), where the tritium content was determined using the ^3H–^3He ingrowth method [6]. The reaction rate of the $^6Li(n,t)\alpha$ and the $^{16}O(t,n)^{18}F$ reactions were estimated using the MCNPX 2.7.0 [7,8] Monte Carlo particle transport code.

So the whole procedure of the SCPA technique was tracked in a special case using both the experimental and the simulation approach, what was necessary to develop this method to be a reliable diagnostic tool for tritium measurement.

2. Experimental

2.1. Sample preparation

First a 1.01229 ± 0.00099 wt% stock solution of Li_2CO_3 (99.997% purity, Sigma-Aldrich, ref. num.: 203629) and distilled water was prepared. Roughly ~ 100 mg solution was filled and

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[1] In this paper, the term *triton* is used to distinguish the ionized, accelerated ^3H particle from a ^3H atom, tritium.

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sealed into an empty ~ 1.5 g weight quartz ampoule. Approximately 1 cm^3 air was left in the closed ampoule. This type of samples are named as kv_Li_*nn*, where *n* stands for the sample number. The mass of the solution and the ampoules were measured using an analytical balance with an accuracy of 5 μg. For flux determination a small Au foil was packed together with the ampoule into the rabbit (sample holder) prepared for irradiation, and the $^{197}Au(n,\gamma)^{198}Au$ reaction rate was measured after its irradiation. The foils were a few milligram in weight, 5 mm in diameter, 0.1 mm thick, disc shaped Au–Al alloys (0.1:100). Appropriate distance was kept between the foils and the ampoules to avoid shielding effects.

The reaction rate measurements of $^{16}O(t,n)^{18}F$ reaction in the aqueous solution of Li_2CO_3 were compared with the previous investigation presented in Ref. [5], where pellets were used by compressing the mixture of MgO powder (99.995% purity, Sigma-Aldrich, ref. num.: 529699) and the same Li_2CO_3 powder in different ratios. That sample series were assigned as LiMg31–37. The pellets were cylindrically shaped with a diameter of 6 mm and with a height of 0.8–1.6 mm.

2.2. Irradiation and γ-spectroscopy

The samples were irradiated at 10 kW ($\Phi = 4.30\times10^{11}\pm1.25\times10^{10}$ n/(cm^2 s)) and at 100 kW ($\Phi = 4.30\times10^{12}\pm1.25\times10^{11}$ n/(cm^2 s)) power using the rabbit system at thermal irradiation position. The irradiation time, t_{irr}, was between 5 and 30 min, the cooling time, t_c, was between 0.5 and 7 h, the live and real measuring times, t_m^{live} and t_m^{real}, were between 20 min and 2 h (see data in Table A1).

A part of tritium produced during the irradiation remained in the aqueous solution of Li_2CO_3, and formed tritiated water, mainly in HTO form. The other part of tritium diffused into the remained air of the sealed ampoule in HT form. Some hot atom reactions and chemical reaction products caused by ^{18}F nuclides coming from $^{16}O(t,n)^{18}F$ reaction were also expected but not investigated, such as HF and LiF.

A BE3830P type HPGe detector and a DSA1000 spectrum analyser from Canberra were used for recording the γ-spectra, and the Genie2000 [9] program was used for evaluating them. The solution in each ampoule was shaken down to the bottom of the ampoule in order to form a drop having a diameter of 5–6 mm. In this case if the ampoule is placed far away from the detector in upright position, the solution can be regarded as a point source. All of the samples were measured in the same geometry: point source at a distance of 7.5 cm above the detector, on its symmetry axis.

One of the investigated nuclides, ^{18}F, is a β^+ decaying nuclide with no γ-line besides the annihilation peak. Therefore any other possibilities resulting the 511 keV peak in the γ-spectrum must be excluded. For this purpose the decay curve belonging to the peak around 511 keV was measured and its decay constant was determined. First a quartz ampoule filled with distilled water was irradiated without adding any Li_2CO_3 but no γ-lines were found around 511 keV. After that both a pure Li_2CO_3 powder and its aqueous solution were irradiated, and the half-lives of $T_{1/2} = 109.40\pm0.41$ min and $T_{1/2} = 109.7\pm1.8$ min with 95% confidence bounds were determined, respectively. From one hand the half-life of ^{18}F according to the literature is $T_{1/2} = 109.77\pm0.05$ min. From other hand there is no any other nuclide having half-life between 107 and 112 min, and producing γ-line only between 510 and 512 keV. These facts prove that only ^{18}F can be the origin of the annihilation peak.

Note that if silicon is irradiated in a thermal reactor, a significant amount of ^{31}Si ($T_{1/2} = 157.3$ min) is produced through the $^{30}Si(n,\gamma)^{31}Si$ reaction, which has a strong 1.5 MeV energy β^- radiation and only a very low intensity γ-line. The quartz ampoules had a few GBq activity after their irradiation due to the silicon content. Therefore their treatment was restricted due to radiation protection considerations. Nevertheless the β^- radiation caused an intense background in the spectrum through the bremsstrahlung effect and sometimes high dead time ($\sim 10\%$).

2.3. Tritium measurement

The irradiation and the HPGe measurements at BME were followed by the determination of the tritium amount in HEKAL in Debrecen, Hungary. The tritium measurement procedure based on the $^3H-^3He$ ingrowth method, where the ^3He produced during the storage time is measured by a static noble gas mass spectrometer (VG-5400), is presented in the following for one irradiated sample. This procedure was repeated for each irradiated sample.

1. Four ampoules were put into a vacuum system. The first one was the irradiated sealed quartz ampoule containing the aqueous solution of Li_2CO_3 and some air. The second one was an open empty ampoule. The third one was open, too, and contained some CuO and activated carbon. And finally the fourth one was a closed ampoule with ~ 10 cm^3 H_2.
2. The system was placed under vacuum and then hermetically closed.
3. The quartz ampoule was opened and the tritiated water was frozen into the empty ampoule with liquid nitrogen.
4. Then the H_2 containing ampoule was opened and the H_2 gas, as a carrier gas, was let into the system. Then the H_2 with tritium in gaseous form were frozen into the activated carbon with liquid nitrogen.

The activated carbon can adsorb more than the 99% of H_2 at -196 °C, but some small amount is still remaining in gaseous form. Therefore, before removing the ^3He impurity the rest of the H_2 need to be adsorbed, otherwise the H_2 would be removed too. The following steps aim this purpose.

5. After the H_2 had been frozen into the activated carbon, the ampoule was closed and heated up to 500 °C. At this temperature H_2 burns in the presence of CuO catalyst to form water.
6. This water had been frozen by liquid nitrogen. Then the ampoule was opened and the ^3He was pumped away (cf.: Section 4.1).
7. The empty ampoule, in which the water fraction was filled and frozen, was vacuumised too, so the ^3He content was also set to zero here (cf.: Section 4.1 second paragraph).
8. Then the water and the hydrogen fractions were stored separately. The amount of ^3He produced by the decay of tritium in the fractions was measured by the mass spectrometer.

Before measuring with the mass spectrometer the samples were frozen with liquid nitrogen and degassed for avoiding the cross contamination and allowing multiple measurement of one sample.

Tritium measurement errors less than 3.5% have been achieved using the above described procedure. Although the ^3He ingrowth method has been proven to be a precise tritium measurement method using a noble gas mass spectrometer, not all laboratories can provide such an accurate tritium analytical results with this type of instrument. HEKAL's newly developed ^4He isotope dilution technique [6] led the laboratory to achieve a very good absolute accuracy and sensitivity which is substantial for environmental water samples. For testing its precision the laboratory