NUKLEONIKA 2012;57(1):61–66

ORIGINAL PAPER

Methodologies to assess uncertainties in the tritium production within lithium breeding blankets

Jesus Salvador,
Oscar Cabellos,
Carlos J. Díez

Abstract. Tritium would have to be produced and controlled in the future fusion facilities. The capture of fusion neutrons by lithium has been proposed as a possible tritium reproduction reaction and lithium blankets for tritium breeding based on this reaction were designed. For the purpose of plant operation and for the safety reasons it is necessary to assess the accuracy with which we can predict the amount of tritium that can be produced. In particular, it is important to assess the impact that the uncertainties inherent in the nuclear data have on the prediced values. By focusing on specific applications and finding specific deficiencies, such studies point to possible directions to improve nuclear data sources. In this paper experimental data on tritium production in a mock-up system are reproduced and their uncertainties assessed in order to identify the reactions that have largest contributions to the total uncertainty.

Key words: uncertainty • nuclear data • lithium breeding blankets

J. Salvador✉, O. Cabellos, C. J. Díez
Institute of Nuclear Fusion,
Universidad Politécnica de Madrid,
c/José Gutiérrez Abascal, 2-28006 Madrid, Spain
and Department of Nuclear Engineering,
Universidad Politécnica de Madrid,
c/José Gutiérrez Abascal, 2-28006 Madrid, Spain,
Tel.: +34 91 336 3108, Fax: +34 91 336 3002,
E-mail: j.salvador@upm.es

Received: 9 July 2011
Accepted: 5 January 2012

Introduction

Due to its physical properties, deuterium-tritium fusion reaction has been chosen for the production of energy in the future fusion machines. Deuterium is an abundant and stable isotope widely available in oceans, but tritium is scarce in nature, so for a continuous operation of the future fusion power plants it would have to be artificially produced. Fortunately, in our planet there are abundant resources of lithium, so tritium could be possibly produced via the neutron irradiation of ^{6}Li and ^{7}Li (7.5% and 92.5% natural abundance, respectively). Their tritium production cross sections, shown in Fig. 1, suggest that an optimized tritium production rate in blankets surrounding the plasma requires enrichment in ^{6}Li and an intense stream of neutrons produced in D-T reactions and subsequently thermalized and multiplied by neutron multipliers.

In the framework of the EU fusion programme two designs of tritium breeding blankets are considered: the helium-cooled pebble bed (HCPB), with lithium ceramic beads as the breeder and beryllium pebbles as the neutron multiplier, and the helium-cooled lithium-lead blanket (HCLL), with a Pb-Li eutectic alloy as both the breeder and the neutron multiplier. It is planned that both types of tritium breeding modules (TBM's) would be installed in ITER to study various aspects of their behaviour [6].

From the point of view of reactor neutronics these preliminary tests would be useful in validating the capability of the neutronic codes and nuclear data libraries

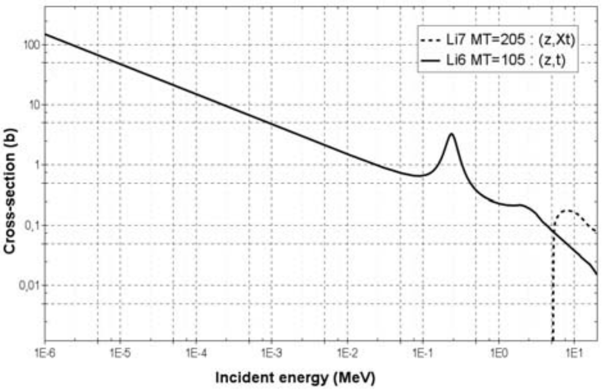

Fig. 1. Tritium production cross section for ^{6}Li and ^{7}Li (JEFF-3.1.1).

to calculate neutron fluxes, energy spectra, neutron multiplication effectiveness of the selected materials, and shielding efficiency, and to predict the tritium production rate. Since the tritium self-sufficiency of the reactor depends on the value of this parameter, the uncertainties in its estimations have to be carefully assessed.

At the Neutron Generator Laboratory of the Frascati Research Center, Italy, a TBM-HCLL mock-up consisting on a block made of LiPb eutectic was irradiated in 2008 and 2009 [1]. Eight holders made of lithium-lead were placed at increasing distances from the surface and, after several days of exposure, the tritium activity of the samples was measured [7]. The tritium production at each depth was measured and in this way the dependence of tritium production rate on the changing neutron spectrum was determined. The reported activities are spread over a range of values, which is understood to be a consequence of fluctuations in the Li abundance. From MCNP simulations it follows that a best overall fit for the experimental data [7] is obtained for the composition of 0.62 wt% Li in LiPb and 3.4 at% ^{6}Li in Li (Fig. 2). It is also important to note that different MCNP simulations of the variation of the tritium activity as a function of depth show larger spread close to the surface (where the energy of incident neutrons is high, ~ 7.7 MeV) than deep inside the samples (neutron energies ~ 1.2 MeV). In other words, simulations performed for two different sets of conditions result in a range of values with a spread that decreases as the energy decreases.

Comparing the simulated curves for the tritium activity with experimental data one may in fact determine the actual content of the samples. However, for a correct assessment of the reliability of the predictions one must take also into account the uncertainty arising in the simulation results due to the uncertainties in the available nuclear data themselves. To this end in the section 'Nuclear data uncertainties for the tritium production by lithium irradiation in the EAF/2010 and ENDF/B-VII.0 libraries' we discuss briefly differences between nuclear data libraries used in this work, and we describe two methodologies to propagate uncertainties in the nuclear data to the response functions, applying them to the problem introduced above. Finally some conclusions about our methodologies and the nuclear data libraries needs are presented.

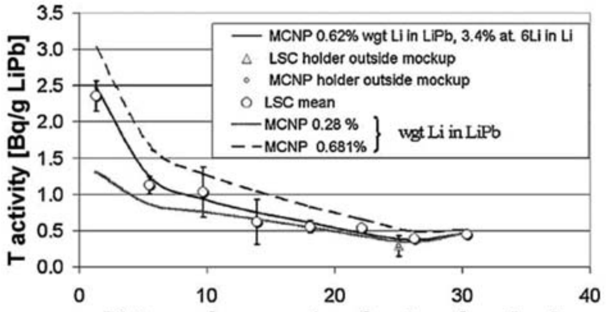

Fig. 2. Tritium activity in LiPb samples, as measured in the Frascati experiment. Lines represent values given by simulations, Ref. [7].

Nuclear data uncertainties for the tritium production by lithium irradiation in the EAF/2010 and ENDF/B-VII.0 libraries

Few of the available nuclear data libraries include detailed information about the uncertainties and covariances that apply to their entire set of nuclides and reactions. It is of uttermost importance to find out which of them are suitable for the actual nuclear engineering applications and to assess the actual quality of their predictions [2]. To study such differences we use EAF/2010 and ENDF/B-VII.0 libraries as an example. The tritium production by the neutron irradiation of ^{6}Li and ^{7}Li and their uncertainties are included in the EAF/2010 library. Figure 3 shows the tritium production cross section for ^{7}Li according to ENDF/B-VII.0 and EAF/2010 and compares them with experimental data. The shaded area corresponds to the EAF/2010 variance and, as can be noticed, its value in the whole energy range represents nearly a 33% of relative error.

It is not so easy to get an idea of the relative error enclosed in the corresponding nuclear information of the ENDF/B-VII.0 library because the tritium production reaction is divided in all its channels and, consequently, the cross section and uncertainties are only reproducible from procedures assuring their proper addition. A well-established strategy to compare nuclear data uncertainties is their inclusion in one energy group with the neutron flux of the corresponding application. For the purpose of this paper it was necessary to make use of the expression for the variance

$$\Delta^2 = \omega^T V \omega \tag{1}$$

where V is the variance matrix of the relative cross section and ω is a weighting factor that involves cross sections and neutron fluxes,

$$\omega = [\frac{\varphi_1}{\bar{\varphi}}\frac{\sigma_1}{\sigma^{\text{eff}}},\dots,\frac{\varphi_g}{\bar{\varphi}}\frac{\sigma_g}{\sigma^{\text{eff}}}]^T \tag{2}$$

where φ_i and σ_i are, respectively, the neutron flux and the reaction cross section in the i-th energy group, and $\bar{\varphi}$ and σ^{eff} are, respectively, the mean flux value and the flux averaged one group cross section. It was also necessary to make use of the neutron spectrum of the fluxes detected at the innermost (SL7) and outermost (SL1) locations of the samples irradiated at Frascati. Table 1 summarizes the relative overall error for one

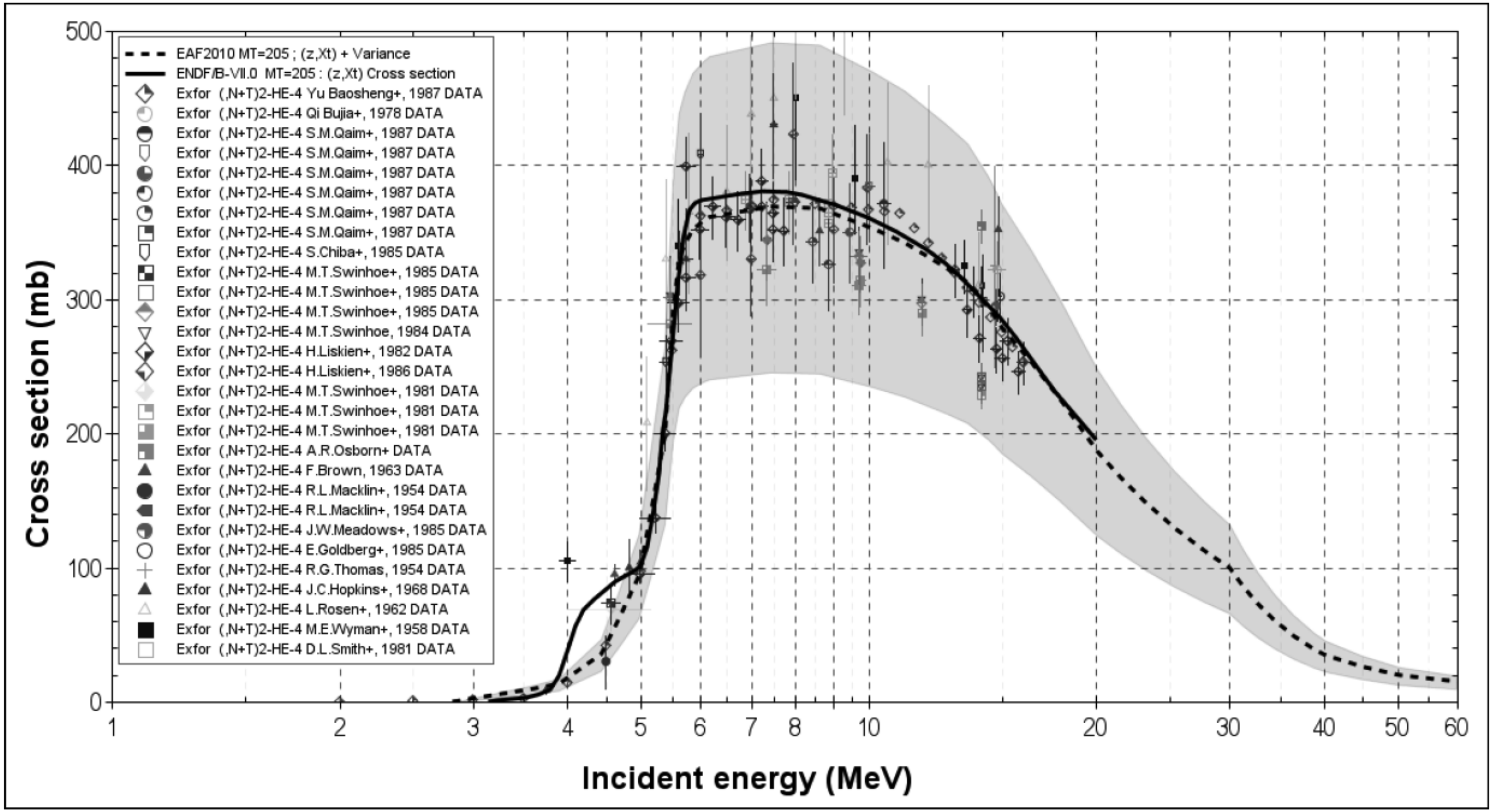

Fig. 3. ^{7}Li(n,T) cross section from EAF/2010 and ENDF/B-VII.0, compared with experimental data.

Table 1. Relative overall error (%) for one energy group, as obtained with SL1 and SL7 neutron fluxes

	SL1		SL7	
	EAF/2010	ENDF/B-VII.0	EAF/2010	ENDF/B-VII.0
^{7}Li(n,T)	33.33	1.17	33.33	1.34
^{6}Li(n,T)	3.33	0.26	3.33	0.13

energy group for both libraries using the neutron spectrum at both distances from the surface of the mock-up. It should be noted that the relative errors provided by ENDF/B-VII.0 and EAF/2010 for the ^{7}Li(n,T) reaction show a clear difference: compared with Fig. 3, ENDF/B-VII.0 gives relative errors that are visibly lower than the experimental errors taken from EXFOR.

Other reactions that were reported in this experiment could be analyzed in a similar fashion, but their contribution is irrelevant for the final purpose of this study and would not change the conclusions that we come to. Huge differences between the relative errors inherent in EAF/2010 and ENDF/B-VII.0 libraries would certainly have significant effect on the uncertainties in the results of tritium production calculations. To estimate these uncertainties we introduce the uncertainty assessment methodologies.

Uncertainty assessment methodologies

A correct assessment of uncertainties in calculated physical predictions is of utmost importance in all applications. The evolution in time of a set of M different nuclide concentrations under the neutron irradiation is given by the expression:

$$\frac{dN}{dt} = AN = [\lambda]N + [\sigma^{\mathrm{eff}}]\phi N \tag{3}$$

where A is the transition matrix depending on $[\lambda]$ (the M × M matrix involving decay constants), $[\sigma^{\mathrm{eff}}]$ (the matrix involving the decay values) and ϕ (the integrated total neutron flux in energy and space [3]). We present two methodologies to estimate how the uncertainties in nuclear data affect the calculated nuclide concentration: a deterministic one (sensitivity/uncertainty) and a stochastic one (Monte Carlo method). We take into account in our study only the uncertainties in cross sections, i.e. we neglect the uncertainties introduced by the rest of the relevant physical parameters.

Sensitivity and uncertainty methodology

The deterministic assessment of the uncertainty in a response function due to the uncertainties in nuclear data is based on the sensitivity analysis, that is, on the analysis of variation induced in the response function by the variation of the parameters on which it depends. A first order Taylor expansion of the response function around the nominal values of the cross sections (σ_0) leads to the expression

$$\frac{N_i(\sigma) - N_i(\sigma_0)}{N_i(\sigma_0)} \approx \sum_{j=1}^{m} \frac{\sigma_{j0}}{N_i(\sigma_0)} \left[\frac{\partial N_i}{\partial \sigma_j}\right]_{\sigma_0} \frac{(\sigma_j - \sigma_0)}{\sigma_{j0}} \tag{4}$$

where the left hand side represents the relative error in the nuclide concentration, N, due to the variations in cross sections, denoted in the following as e_i, and the expression on the right hand-side term represents the sum over all the cross section sensitivity coefficients $\left(\rho_{ij} = \sum_{j=1}^{m} [\sigma_{j0}/N_i(\sigma_0)][\partial_{\sigma_j} N_i]_{\sigma_0}\right)$ multiplied by the relative errors in these cross sections, ε_j. We may write

$$e_i = \rho_{i1}\varepsilon_1 + \rho_{i2}\varepsilon_2 + \ldots + \rho_{im}\varepsilon_m \tag{5}$$

and then

$$\mathrm{Var}\,[e_i] = \rho_{i1}^2\Delta_1^2 + \rho_{i2}^2\Delta_2^2 + \ldots + \rho_{im}^2\Delta_m^2 \tag{6}$$

Monte Carlo methodology

The Monte Carlo methodology relies on sampling of the nuclear data information (mean values, variance and covariance values) according to a selected probability density function (PDF). For each value obtained from the sampling, a calculation of the response function is performed. Since each sampling provides new values of the parameters, after a large number of stochastic samplings and their corresponding response function calculations, a population of values for these response functions is obtained and then a statistical study over this data set is carried out (Fig. 4). In our case, we use repeated simultaneous random sampling of PDF's for all the cross sections involved in the problem, we calculate the amounts of nuclides and calculate statistics estimators. This procedure enables to study the global effect of the complete set of cross section (σ) uncertainties on nuclide concentrations (N).

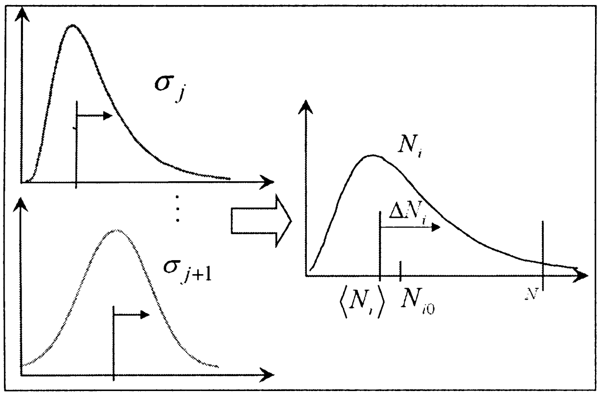

Fig. 4. Monte Carlo sampling of nuclear data to assess how the uncertainties in the nuclear data propagate into uncertainties in the response functions.

Table 2. Tritium content uncertainties at SL1 and SL7, estimated using EAF2010/UN and ENDF/B-VII.0 libraries, for depleted ^{6}Li 3.4% in Li

	SL1		SL7	
Sensitivity coefficient, ρ (%)				
^{6}Li(n,T) ^{4}He	0.11		0.91	
^{7}Li(n,na) T	0.83		0.09	
	EAF/2010	ENDF/B-VII.0	EAF/2010	ENDF/B-VII.0
Uncertainty				
^{6}Li(n,T) ^{4}He	3.33	0.26	3.33	0.13
^{7}Li(n,na) T	33.33	1.17	33.33	1.34
Sensitivity/Uncertainty, ρΔ (%)				
^{6}Li(n,T) ^{4}He	0.37	0.03	3.03	0.12
^{7}Li(n,na) T	27.66	0.97	3.00	0.12
Total S/U	27.67	0.97	4.26	0.17
Monte Carlo	32.99	1.16	5.74	0.23

Results

Both methodologies were applied to the Frascati measurements using the EAF/2010 and ENDF/B-VII.0 cross section and uncertainty libraries. The aim was to quantify their global uncertainty and the main contributing nuclides and reactions. First of all, it was necessary to combine the uncertainties of the reactions involved in one energy group as previously explained. Calculations were performed with the ACAB code [8]. As can be seen in Table 2, for a sample of the composition that provides a best fit to the experimental values (0.62 wt% Li in LiPb and 3.4 at% ^{6}Li in Li), we find that at energies around 1.2 MeV (SL7 position) the sensitivity coefficient of tritium production from ^{6}Li exceeds the coefficient calculated for the ^{7}Li reaction: 0.91% vs. 0.09%. At energies of 7.7 MeV (SL1 position), the sensitivity coefficient of the former is only 0.11% against the 0.83% of the latter.

Comparing the contributions of each of the reactions to the total relative error in the tritium production in the low energy range of the neutrons that reach the SL7 depth we find that they are close to each other: 3.03% due to ^{6}Li and 3% due to ^{7}Li obtained with EAF/2010 library and 0.12% for both reactions according to ENDF/B-VII.0. At the highest energies, which correspond to the SL1 depth, the differences between their contribution increase significantly: 0.37% of the total relative error is due to ^{6}Li reaction and 27.66% due to ^{7}Li in the calculation scheme based on EAF/2010; 0.03% due to ^{6}Li and 0.97% due to ^{7}Li, when ENDF/B-VII.0 is used.

The sensitivity/uncertainty method estimates the total relative error in the tritium prediction around 27.66% for SL1 and 4.26% for SL7 samples, with the EAF/2010 library. Regarding to the results obtained from the ENDF/B-VII.0, the total relative error is around 0.97% for SL1 and 0.17% for SL7. Both libraries suggest that the main contributor to the uncertainty in the high energy range is the ^{7}Li(n,T) reaction.

Regarding the application of the Monte Carlo methodology, it predicts under the same conditions a relative error of 32.99% in SL1 and 5.74% in SL7 with the EAF/2010; and 1.16% in SL1 and 0.23% in SL7 with the ENDF/B-VII.0. In all cases the obtained values are higher than those obtained from the sensitivity/uncertainty analysis. This suggests that the Monte Carlo method is able to take into account a higher number of small tritium production channels, whatever their individual importance is, which altogether contribute meaningfully to the final relative error.

Conclusions

The tritium production in the future fusion reactors cannot be avoided in order to maintain a continuous exploitation. Breeding blanket concepts are under development and study. The quality of nuclear data is of paramount importance for their design, since the tritium production has to be predicted as accurately as possible. Making the most of experimental programmes, nuclear data and calculation methodologies can be improved. Two methodologies of uncertainty assessment have been applied to the TBM HCLL mock-up irradiated in Frascati and help us to critically asses the quality of nuclear data libraries.

The results of the sensitivity/uncertainty analysis show a clear deficiency for the uncertainty of ^{7}Li reaction in the EAF/2010 nuclear data library in the high energy range of the experiment. Calculations based on ENDF/B-VII.0 library provide relative errors that are in agreement with those estimated in previous studies [4, 5]. However, when compared against other nuclear data evaluations, like EAF/2010, and the experimental spread collected in EXFOR, these uncertainties of ^{7}Li reaction in ENDF/B-VII.0 seem to be much too low.

It is also important to highlight the combined potential of these methodologies: the sensitivity/uncertainty analysis provides us with an idea of the main contributors to the uncertainty of a response function, which shows the way in which nuclear libraries may be improved; the Monte Carlo methodology allows us to take into account the complete set of nuclear reaction channels that contribute to the uncertainty and to include in the calculation their global impact.

We plan further tests of nuclear data libraries in order to identify their shortcomings and asses their usefulness. We also plan a deeper study of the effect of choosing different probability density functions. The methodologies explained here can be applied not only to tritium production in the breeding blankets but also in other plasma-facing materials.

Acknowledgment. We want to thank the Organising Committee of 10th Kudowa Summer School "Towards Fusion Energy" for including this topic in the students presentations programme. We are grateful to Władyslaw Pohorecki from the AGH University of Science and Technology (Kraków, Poland) for providing us with the basic data needed to perform calculations reported here.

References

1. Batistoni P, Angelone M, Carconi P *et al*. (2011) Neutronics experiments on HPCB and HCLL TBM mock-ups in preparation of nuclear measurements in ITER. Fusion Eng Des 85:1675–1680
2. Fischer U, Batistoni P, Forrest R *et al*. (2007) Development needs of nuclear data for fusion technology. In: Proc of the Int Conf on Nuclear Data for Science and Technology, 22–27 April 2007, Nice, France, DOI: 10.1051/ndata:07494
3. García Herranz N, Cabellos O, Sanz J, Juan J, Kuijper JC (2008) Propagation of statistical and nuclear data uncertainties in Monte Carlo burn-up calculations. Ann Nucl Energy 35:714–730
4. Leichtle D, Fischer U, Kodeli I *et al*. (2010) Sensitivity and uncertainty analyses of the HCLL mock-up experiment. Fusion Eng Des 85:1724–1727
5. Leichtle D, Fischer U, Perel RL, Serikov A (2011) Sensitivity and uncertainty analysis of nuclear responses in the EU HCLL TBM of ITER. Fusion Eng Des 86:2156–2159
6. Li Puma A, Poitevin Y, Giancarli L, Rampal G, Farabolini W (2005) The helium-cooled lithium lead blanket test proposal in ITER and requirements on test blanket modules instrumentation. Fusion Eng Des 75/79:951–955
7. Pohorecki W, Kuc T, Ostachowicz B, Bilski P (2011) Novel methods of tritium production rate measurements in HCLL TBM mock-up experiment with liquid scintillation technique. Fusion Eng Des 86:2429–2432
8. Sanz J (2000) ACAB activation code for fusion applications: User's manual UCRL-MA-143238. Lawrence Livermore National Laboratory, USA