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Non-destructive analysis of impurities in beryllium, affecting evaluation of the tritium breeding ratio

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Abstract

The non-destructive, pulsed neutron method was used as the most effective way of analyzing the integral effect of impurities in beryllium, relevant to the tritium breeding ratio evaluation. The integral effect was evaluated from time behavior observations of the neutron flux, following the injection of a burst of D–T neutrons into the beryllium assembly. The assembly was constructed from the structural beryllium grade S-200F (Brush Wellman Inc.). Experimental data were compared with the reference data and MCNP-4B calculations. Results show that the measured absorption cross section of thermal neutrons in beryllium blocks is approximately 30% larger than the calculated value, based on the data, specified by the manufacturing company. Impurities in beryllium, such as Li, B, Cd and others, affect the absorption cross section even if the content of impurities is less than 10 ppm.

1. Introduction

Beryllium is a high priority material utilized in the field of fusion technologies. In most conceptual fusion power reactor designs, it is proposed to use beryllium as a neutron multiplier in the blanket [1]. Neutron multipliers are required in order to compensate for parasitic neutron losses, and therefore to enable net tritium production. In addition, beryllium is an excellent neutron moderator with an extremely low absorption cross section for thermal neutrons. In such conditions, tritium production mostly occurs through absorption of thermal neutrons by lithium-6, according to the reaction $^{6}Li(n,\alpha)^{3}H$. However, thermal neutrons can be easily absorbed by other elements as well, if they are presented in beryllium as impurities that increase the absorption cross section. At a maximum, the impurities in beryllium can increase the absorption cross section by up to 30 times [2]. This impurity effect decreases the number of thermal neutrons, and as a result, the tritium production is decreased.

Considering that such impurities can exist in structural beryllium grades, beryllium blocks (S-200F, Brush Wellman Inc.) used for the blanket benchmark experiments were studied with the purpose of qualification and validation. Generally, the inductively coupled plasma mass spectrometry method is suitable for the study, however, due to the wide spectrum and range of expected impurities, and their non-uniform distributions in beryllium, the study would be quite a lengthy and laborious procedure. In such circumstances, it is useful to have a method which is able to quickly, easily and precisely evaluate the integral effect of impurities affecting evaluation of the tritium breeding ratio.

2. Chemical composition of structural beryllium grades and the impurity effect on tritium production

Structural grades S-65C, S-200F and S-200E (Brush Wellman Inc.) are being considered for use in ITER [3]. Manufacturer information concerning the composition of these grades, is shown in Table 1. In typical specifications, the main impurity elements are characterized by

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a high concentration, about 1000 ppm. Minor impurities are commonly not specified in detail. Instead, the total impurity level, of about 400 ppm, is shown in the last row of Table 1. This level is not dependent on the beryllium grade. Very limited information concerning the content of minor impurities is available. Various studies [4–7] have shown that impurities can include elements such as Li, B, Cl, Cr, Mn, Co, Cd, Dy, Th, U and others. Some of the elements tend to have a significant effect on the absorption cross section of thermal neutrons, in spite of the low impurity level, caused by a high value of their absorption cross sections. It is possible to estimate the significance of the effect, using the impure beryllium to pure beryllium ratio of the total macroscopic absorption cross sections, *R*. Results of calculations of *R* for impurities, which are characterized by a high absorption cross section, are presented in Table 2. As indicated in this table, even a small amount of impurities, less than 10 ppm, can significantly increase the total absorption cross section in beryllium.

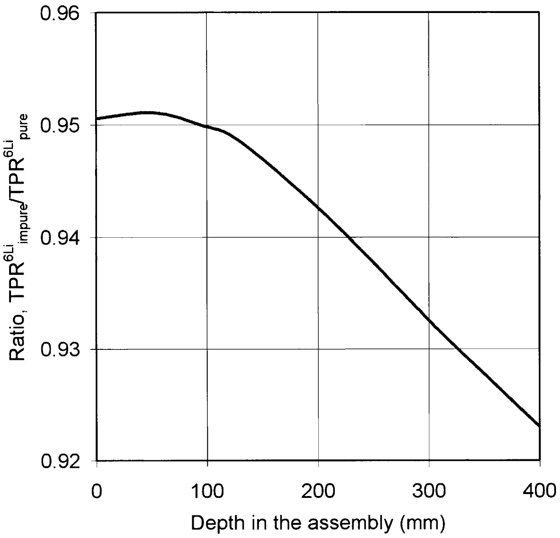

Fig. 1. The calculated effect of the parasitic absorption of thermal neutrons in beryllium blocks on the tritium production rate.

As an example of the influence of the parasitic absorption in the beryllium moderator on the tritium production, an estimation of the tritium production rate on lithium-6, TPR-6, was performed for the beryllium assembly, irradiated by D–T neutrons. The parasitic absorption was simulated by boron at a level of 5 ppm, a concentration corresponding to a 40% increase of the absorption cross section. The ratio of TPR-6 in the impure beryllium to pure beryllium is presented in Fig. 1. The parasitic absorption decreases the TPR-6 by about 5% on the surface of the assembly (Fig. 1). This effect has a tendency to decrease the TPR-6 to a deeper location, due to the moderation and thermalization of neutrons, as a result of the increasing absorption cross section, which follows the 1/*v*-law. Nevertheless, the impurity effect depends on the blanket design, volumetric ratio of lithium and beryllium, and enrichment of lithium.

Table 1
Specification of chemical compositions for structural beryllium grades

Chemical composition	S-65B/S-65C	S-200F	S-200E
Be, min %	99.0	98.5	98.0
BeO, max %	1.0	1.5	2.0
Al, max ppm	600	1000	1600
C, max ppm	1000	1500	1500
Fe, max ppm	800	1300	1800
Mg, max ppm	600	800	800
Si, max ppm	600	600	800
Other max ppm	400	400	400

Table 2
The effect of minor impurities on the ratio of macroscopic absorption cross section of impure to pure beryllium, *R*

Composition			*R*
Be, %	Impurity		
	Element	Level, ppm	
100	–	0	1.00
99.99	B	5	1.43
99.99	Gd	5	2.78
99.99	Cd	5	1.15
99.99	Li	5	1.06
99.99	Cl	100	1.11
99.99	Co	100	1.08
99.99	Mn	100	1.03
99.99	All the above	320	3.63

3. Experimental methods

The non-destructive, pulsed neutron method was chosen with the following constrains in mind. First, it is useful to estimate the total effect of impurities, which increase the macroscopic absorption cross section of thermal neutrons, since the effect directly affects the tritium breeding ratio on lithium-6. Secondly, it is reasonable to measure the integral effect of impurities, in order to avoid a future problem concerning the uniformity of impurity distributions. The effectiveness of this method is based on the following facts:

- all beryllium blocks, used for the benchmark experiments are suitable for estimation of the integral impurity effect;

- the total macroscopic absorption cross section for thermal neutrons can be measured;
- high accuracy of experimental results;
- fast and simple experimental procedures.

3.1. Basic principles

The principle of the neutron pulsed method is described in detail in Ref. [8]. In essence, the method consists of time behavior observations of the neutron flux, following the injection of a burst of neutrons into the moderator. Fast neutrons inside the assembly rapidly slow down to thermal velocities (the slowing-down time in beryllium ~100 μs).

When the neutrons approach thermal equilibrium with the moderator, they continue to leak slowly out of the assembly or can be absorbed. The thermal neutrons decay according to the law:

$$\Phi(t) \approx e^{-\alpha t}. \tag{1}$$

The constant $1/\alpha$ is the effective thermal neutron lifetime. If the geometrical dimensions of the moderator are characterized by its buckling parameter B^2, the experimentally observed decay constant can be presented in the following way:

$$\alpha = \alpha_0 + (D - CB^2) \cdot B^2, \tag{2}$$

where α_0 is the decay constant which would be observed in an infinite medium and is entirely dependent on absorption ($\alpha_0 = v_0 \cdot \sum_a^{\text{total}}(v)$, for $1/v$ – absorption; $\sum_a^{\text{total}}(v) = \sigma_a^{\text{Be}}(v) \cdot n^{\text{Be}} + \sum_{\text{impurities}} \sigma_a^i(v) \cdot n^i$, σ_a^i – cross section, and n^i – number of nuclei per unit volume); D is the diffusion coefficient; v_0 is the neutron velocity (2200 m/s).

The term – 'CB^2' is a small correction to the diffusion coefficient, which describes the diffusion cooling [8]. In a finite moderator, the neutron equilibrium temperature lies below the moderator temperature, due to the fact, that the leakage rate increases with velocity. Geometrical buckling for the parallelepipedal assembly can be estimated with the following formula:

$$B^2 = \pi^2 \cdot \left(\frac{1}{\tilde{a}^2} + \frac{1}{\tilde{b}^2} + \frac{1}{\tilde{c}^2} \right), \tag{3}$$

where $\tilde{a}$, $\tilde{b}$, $\tilde{c}$ are the 'effective dimensions', $\tilde{a} = a + 1.42\lambda_{\text{tr}}$; a, b, c are the actual dimensions; λ_{tr} is the transport mean free path.

Hence, measurement of the decay constant α of the thermal neutron flux for the fixed size of the moderating assembly and known diffusion parameters for beryllium, will permit determination of the absorption cross section.

3.2. Instrumentation

The equipment consists of a pulsed D–T neutron source (FNS facility in JAERI), a beryllium assembly, a BF_3 neutron detector, and a time analyzer. The fast neutron burst width was 1 μs and the burst repetition time was 5 ms. Standard grade beryllium blocks (S-200F, Brush Wellman Inc.) were used to construct ten cubical prisms with dimensions approximately 30 × 30 × 30 up to 60 × 60 × 60 cm. The pulsed source and the detector were located outside of the beryllium blocks, normally in the middle of the adjacent side planes. This arrangement minimizes the amount of higher harmonics contamination of the thermal neutrons decay. Thermal neutrons diffusing out of the beryllium cube were detected with a BF_3 counter. The decay curve was accumulated in the 256 channel time analyzer using a 20 μs channel width. Each decay curve was treated off-line.

4. Results and analysis

Two independent comparisons were completed, based on the experimental and calculated results. Fist of all, the measured decay curve of thermal neutrons diffusing out of the beryllium assembly was compared with the Monte Carlo calculations using the MCNP-4B code and JENDL-3.2 data set. Manufacturer's information on major impurities in beryllium (C, O, Al and Fe) was

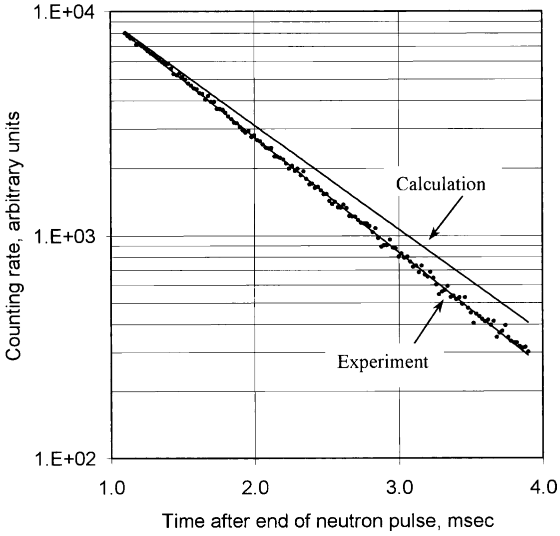

Fig. 2. The decay curve of thermal neutrons diffusing out of beryllium cube, as detected with a BF_3 counter.

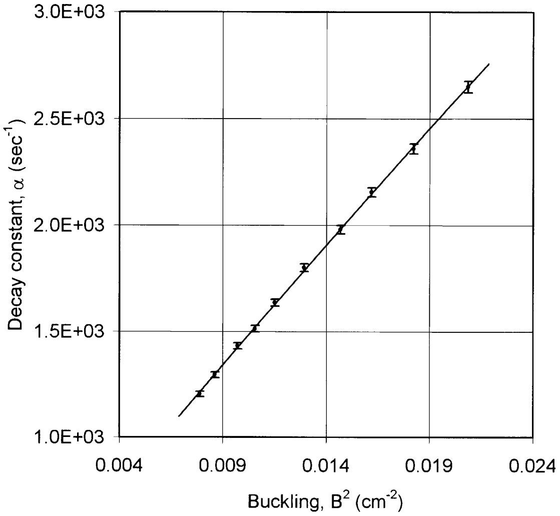

Fig. 3. Dependence of decay constant of thermal neutrons diffusing out of beryllium assemblies on the size given by the geometrical buckling B^2.

taken into account. The comparison example for a beryllium assembly with a size of about 60 × 60 × 60 cm is shown in Fig. 2. It is evident that the measured decay curve was faster than the calculated curve. Similar results were obtained for all ten assemblies, indicating, that the thermal neutrons are absorbed by the minor impurities present in the beryllium blocks. In addition, by analyzing the dependence of the experimental decay constants for assemblies of various sizes on the geometrical buckling, the total macroscopic absorption cross section was evaluated. Fig. 3 shows a plot of the decay constant as a function of geometrical buckling. According to function (2), a parabola has been fitted to experimental points by the method of least squares. Thus, the decay constant was evaluated as $\alpha_0^{\exp} = 278 \pm 25\ \mathrm{s}^{-1}$. This value is about 30% higher than the calculated value, $\alpha_0^{\mathrm{cal}} = 219\ \mathrm{s}^{-1}$, based on the absorption cross section data taken from the JENDL-3.3 library [9] and chemical composition of beryllium provided by the manufacturing company.

5. Conclusions

Structural beryllium (S-65C, S-200F, S-200E from Brush Wellman Inc.), which is considered for use in ITER, contains unspecified impurities with a total amount of less than 400 ppm. The composition of these impurities contains elements, such as the Li, B, Cd and others, which can affect the tritium breeding ratio, due to parasitic absorption of thermal neutrons in the beryllium material, even if their content is less than 10 ppm. In order to evaluate the integral effect of these impurities relevant to the tritium breeding ratio evaluation, a non-destructive, pulsed neutron method was used. The measurements demonstrated the usefulness of the pulsed neutron method, as a tool ideal for fast and simple determination of the integral effect of impurities. It has been experimentally proven that the effective absorption of thermal neutrons in structural beryllium (S-200F) is approximately 30% higher than the calculated value, based on the data specified by the manufacturing company. For the pure beryllium assembly, such effect will decrease the tritium production rate by at least 5%. In the blanket of a fusion reactor, the effect will depend on the blanket design, volumetric ratio of lithium and beryllium, and enrichment of the lithium. The estimation should be completed in each case.

6. Additional remarks

Acquiring the impurity information is not only important for evaluation of the tritium breeding ratio, it is also significant for studies concerning the activation and transmutation behavior of beryllium in future fusion power plants. The activation behavior of beryllium in a fusion reactor depends strongly on the presence of impurities in the metal, thus affecting the choice of conditioning methods for beryllium waste from fusion reactors. Estimations [10] show, that the radiotoxicity, and the actinide inventory of the fusion beryllium waste, are strongly associated with the initial concentration of uranium and thorium.

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