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INVESTIGATIONS OF LITHIUM CARBIDE AS TRITIUM BREEDING MATERIAL FOR BLANKET OF THE FUSION REACTOR

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Comparison of tritium formation values in the course of irradiation by neutrons of fusion reactor of lithium carbide and Li-based ceramics (Li_4SiO_4 и Li_2TiO_3) has been done. The influence of neutron thermalization effect by lithium carbide matrix for tritium yield has been estimated. The yield of tritium during neutron lithium carbide irradiation is 1,5-2 times higher than in the case of Li-based ceramics. The estimated tritium breeding ratio for lithium carbide is about 1.5.

INTRODUCTION

The long lasting campaign of a thermonuclear reactor assumes continuous tritium replenishment in the active zone of the reactor. All projects of the fusion reactor imply presence of tritium breeding zone which is component of blanket. Selection of the effective breeding material which should response different requirements is a serious problem. Breeding material should provide both high yield of tritium through the relevant nuclear reactions and easy tritium release for the fuel preparation system. Analysis of the reports submitted for the International Conference on Fusion Reactor Materials [1], shows that the most frequently used breeder materials are different types of lithium ceramic, such as Li_4SiO_4 and Li_2TiO_3. Lithium metatitanate (Li_2TiO_3) attracts attention due to its chemical stability and high rate of tritium release at relatively low temperatures (200 to 400°C). Lithium orthosilicate (Li_4SiO_4) distinguishes by its low activation, resistance to hydration, and increased yield of tritium due to the relatively high content of lithium.

We have investigated lithium carbide (Li_2C_2) as tritium breeding material for the fusion reactor blanket. The aim of this work is compare the tritium breeding ratio (TBR) with lithium carbide and lithium ceramics (Li_4SiO_4 and Li_2TiO_3) ones. It has been estimated that neutron moderation by lithium carbide results in noticable increasing of tritium yield while irradiation.

PROPERTIES OF LITHIUM CARBIDE

Lithium carbide is colourless, fragile crystal material. Its density at room temperature is 1,65 g/cm^3. Lithium carbide can be received at direct interaction of lithium and carbon ($2Li+2C \rightarrow Li_2C_2$) in vacuum at 650-700°C or while heating of lithium in the current of acetylene or ethylene [2-4]. Industrial production of lithium carbide in the former USSR has been started since 80th years of last century. For synthesis of lithium carbide one can use components of high purity. Implying the simplicity of the synthesis the product of high purity is potentially achievable.

According to the data received on the studying the system lithium — carbon [3], the compound Li_2C_2 can exist in several polymorphic modifications with temperature of transition at 410, 440 and 550°C. Interaction of Li_2C_2 with water causes explosion due to release of hydrogen, the other product of the reaction is elemental carbon [2]. At high temperature lithium carbide intensively dissociates into lithium and graphite. The pressure while dissociation at 925°C is 0,35 kgfs/cm^2 [4].

As concerns the service or disposal of the components of fusion reactor at the end of its life cycle it is important to estimate the induced activity of the breeding materials. The investigations carried out earlier [5] have shown that the total radioactivity of the radionuclides induced while irradiation lithium carbide and lithium ceramics (Li_4SiO_4 and Li_2TiO_3) by fusion neutrons (fluence $2 \cdot 10^{23}$ cm^{-2}) of is quite similar. After 3 years of material cooling there is no other radionuclides excepting tritium. The equivalent dose rate of the irradiated hypothetically pure lithium carbide reaches the safe level (23 μSv/h) for one minute after the irradiation end and this simplifies maintenance the installation. The mass of tritium forming in the lithium carbide is 1,5-2,6 times higher in comparison with lithium ceramics (Li_4SiO_4 and Li_2TiO_3). The rate of tritium recovery is about $\approx 1 \cdot 10^{-6}$ g/s while irradiation of 100 kg of lithium carbide. This value is in a good agreement with data given in work [6].

RESULTS AND DISCUSSION
TRITIUM BREEDING RATIO (TBR)

For sustainable fuel cycle of the fusion reactor the quantity of tritium atoms formed by single thermonuclear neutron (Tritium Breeding Ratio - TBR) has to be more than 1,05 [6]. The structural model of lithium blanket suggested in the work [6] assumes the minimum quantity of lithium about 100 kg. TBR value for such model of blanket is about 1.28. TBR value for the lithium blanket [9] of 2 m thickness (without neutron reflector) is about 2. Such a difference for TBR values for the same material, possibly, can be explained by both different blanket designs and different neutron spectrum. This may be result of missing in the computations the neutrons absorption and leakage.

Let us consider an infinite medium of lithium carbide for the calculation of the TBR value for Li_2C_2.

Lithium carbide is assumed to be irradiated by mono energetic 14 MeV neutrons. We suppose that leakage of neutrons does not present, and isotope composition of lithium and carbon corresponds to their natural abundances (7,42% Li^6 and 92,58% Li^7, 98,9% C^{12} and 1,1% C^{13}). It has been previously shown [7] that the main amount of tritium (99.985% of the total concentration of tritium) while irradiation of lithium carbide forms on the nuclei of lithium. The rest amount of tritium (0,015%) is formed on the C^{13} isotope. Nuclear reactions of formation of tritium in the material are given below.

Each neutron which is absorbed by the reaction Li^7(n,n'T)He^4 leads to the formation of tritium and another neutron. This neutron in its turn can react with another isotope of lithium and as a result one may get extra nucleus of tritium. However, the reaction Li^7(n,n'T)He^4 is run in a relatively narrow neutrons energy range: from 3,2 to 20 MeV (Fig. 1, line 2). But for neutrons with energy about 14 MeV, this reaction has a cross-section maximum which is 10 times higher than cross-section of the reaction Li^6(n,T)He^4. The TBR value for isotope of Li^7 (k_{Li-7}) can be written down in the following form [8]:

$$k_{Li-7} = 1 + k', \; where \; k' = 1 - \exp\left\{ -\int_0^\infty \frac{\Sigma_a(u)du}{\xi\Sigma_s(u)} \right\}. \quad (1)$$

Thus,

$$k_{Li-7} = 2 - \exp\left\{ -\int_0^\infty \frac{\sigma_a(u)du}{\xi\sigma_s(u)} \right\}. \quad (2)$$

In the formula (1) Σ_a – macroscopic cross-section of reaction (n,n'T); $\xi\Sigma_s$ — moderating efficiency by lithium carbide; $u=\ln(E_0/E)$ – lethargy, where E_0 – constant, E – energy of a neutron in a result of elastic collisions with nucleus of i-component of the medium (process of neutron moderation in the medium). In the formula (2) σ_a – microscopic cross-section of reaction (n,n'T) and σ_s – microscopic scattering cross-section.

Moderating efficiency of a chemical compound may be presented in the next form:

$$\xi\Sigma_s = \xi_1\Sigma_{s_1} + \xi_2\Sigma_{s_2} + \ldots + \xi_k\Sigma_{s_k} = \sum_{i=1}^{k} \xi_i\Sigma_{s_i} \quad (3)$$

Indices i=1,2,…, k indicates different chemical elements forming the compound; Σ_s — macroscopic scattering cross-section ($\Sigma_s=\sigma_s \cdot N$, where N — number of nuclei in 1 cm^3 of the matter); ξ — parameter of moderation of neutrons, which may be calculated according the following formula:

$$\xi = 1 + \frac{(A-1)^2}{2A} \ln \frac{A-1}{A+1}. \quad (4)$$

Here A – atomic mass of isotope of a chemical element.

Reaction Li^6(n,T)He^4 is effective way for neutrons absorption. This reaction takes place for neutrons having energy in interval $1 \cdot 10^{-5}$ -$20 \cdot 10^6$ eV (see Fig.1, line 1). The value of TBR for Li^6 (k_{Li-6}) may be calculated by the next equation:

$$k_{Li-6} = 1 - \exp\left\{ -\int_0^\infty \frac{\Sigma_a(u)du}{\xi\Sigma_s(u)} \right\}, \quad (5)$$

where Σ_a – macroscopic cross-section of reaction (n,T); $\xi\Sigma_s$ — moderating efficiency of lithium carbide.

The reaction C^{13}(n,T)B^{11} takes place only for high-energy neutrons (13,5-20 MeV). The TBR value for isotope of C^{13} (k_{C-13}) may be calculated by the formula (5).

Table 1

The Tritium Breeding Ratio (TBR) in the combined spherical system Li(50 cm)+C(20 cm)

Reference	TBR
Experimental data [13]	1.13
Calculated data [13]	1.03
JENDL-3.2	1.295
BROND-2.1	1.394
FENDL-2.0	1.288

Total TBR value for lithium carbide equals $k = k_{Li-6} + k_{Li-7} + k_{C-13}$. Calculated value of TBR for lithium carbide irradiated by neutrons with 14 MeV energy (without consideration neutrons leakage) is 1.52. For comparison, TBR value for lithium orthosilicate (Li_4SiO_4), depending on values of breeder enrichment by lithium-6, varies from 1.069 [10] to 1.147 [11] and for lithium metatitanate (Li_2TiO_3) it is equaled to 1.185 [11]. Thus, the calculated TBR value for lithium carbide exceeds 1.45 times the required minimum value (1.05) for the fuel cycle of reactor. The TBR value for Li_2C_2 is higher than for proposed recently lithium ceramics. Experimental data to determine k value for lithium carbide is absent so far. The paper [12] presents the TBR values in lithium sphere that is 50 cm thick and covered with a layer of 20 cm thick graphite calculated using various programs (see Table 1) and obtained experimentally [13]. Despite the difference of the calculated and experimental data values (from 9% to 23%), the value of the TBR (1.13) obtained in practice in the combined spherical system Li(50 cm)+C(20 cm) exceeds the threshold equal to 1.05 and is in magnitude comparable with the considered lithium ceramics.

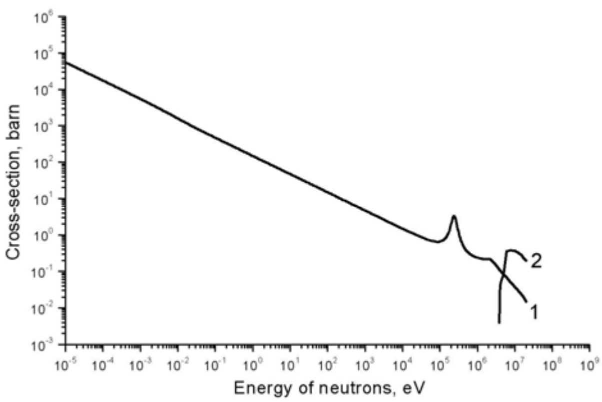

Fig. 1. Dependence of the nuclear reaction cross section for tritium formation from the neutron energy:
1 – reaction Li^6(n,T)He^4,
2 – reaction Li^7(n,n'+T)He^4 [9]

THE ESTIMATION OF INFLUENCE OF EFFECT OF NEUTRON THERMALIZATION BY LITHIUM CARBIDE MATRIX FOR TRITIUM FORMATION

Carbon and lithium are effective neutron moderators, providing thermalization of the neutron spectrum. For example, the authors [12] believe that the TBR value in spherical system Li(50 cm)+C(20 cm) is over two times than the TBR value in the lithium sphere that is 50 cm thick due to the reflected with the graphite layer flux of thermalized neutrons returned to the lithium sphere. The distribution of the moderated neutrons among the energies in the non-absorbing environment (i.e., the environment where the moderated neutrons are not absorbed) can be expressed as the following equation [14]:

$$n(E)=\sqrt{\frac{m_n}{2}}\cdot\frac{q}{\xi\Sigma_s\cdot E^{3/2}}, \quad (8)$$

where m_n – the mass of the neutron (m_n=1 amu), q - the rate of neutrons generation (or neutron flux density before the moderation), E – neutron energy, $\xi\Sigma_s$ - moderating ability of the environment is calculated using the formula (5). The number of lithium (or carbon) atoms (36,8 wt.% Li and 63,2 wt.% C) in 1 kg of the lithium carbide is defined as the following formula:

$$N=\frac{1000\cdot 6{,}02\cdot 10^{23}\cdot n}{M_{Li_2C_2}} \quad (9)$$

In formula (9) n - number of Li (or C) atoms in Li_2C_2 molecule, $M_{Li_2C_2}$ - molecular mass of Li_2C_2.

For the construction of the projected spectrum of moderated neutrons in the lithium carbide we will use the neutron spectrum of the fusion reactor [15] designated as DEMO (Fig.2, continuous part of the line 1). This spectrum does not contain any data on distribution of thermal neutrons. However, in the field of thermal neutrons ($1\cdot 10^{-5}$-1 eV) the cross section of reaction of the tritium from lithium-6 formation differs with the exceptionally high values, making ~10^4-10^2 barn. In this connection, the function describing DEMO neutron spectrum was extrapolated to 0,1 eV energy neutrons value (Fig. 2, dotted part of the line 1). Then, for a number of points of the obtained neutron spectrum, the moderating ability value of lithium carbide was calculated by the formula (3) and the data received was inserted into the formula (8). As a result, the predicted neutron spectrum was obtained in Li_2C_2 taking into account the moderation effect (Fig. 2, line 2). This illustration shows an increase in the proportion of moderated neutrons with the energy less than 1 MeV, which should contribute to the increase of the tritium recovery from $Li^6(n,T)He^4$ reaction. For the numerical determination of the neutron moderation effect on tritium formation using ACTIVA program [16], the integral cross sections of nuclear reactions taking place in the lithium were calculated for the following energy distributions of neutrons: DEMO spectrum and the spectrum of neutrons in lithium carbide based on their moderation (Fig. 2, line 2). The calculated value of the integral cross section of the reaction (n, T) on lithium-6 for the spectrum of the neutrons moderated by lithium carbide is next larger than the integrated cross section of the same reaction for DEMO spectrum. Given that the mass of the resulting radioactive isotope in the material is directly proportional to the cross section of the reaction of this isotope formation, the tritium formation from $Li^6(n, T)He^4$ reaction will be 10 times greater taking into account neutrons moderation in lithium carbide.

CONCLUSION

The use of lithium carbide as tritium breeding material for fusion reactor is perspective:

- calculated tritium breeding ratio from of lithium carbide (1.52) with a "margin" exceeds the required (1.05),
- it is an effective moderator, which 10 times increases the yield of tritium on Li-6 isotope.

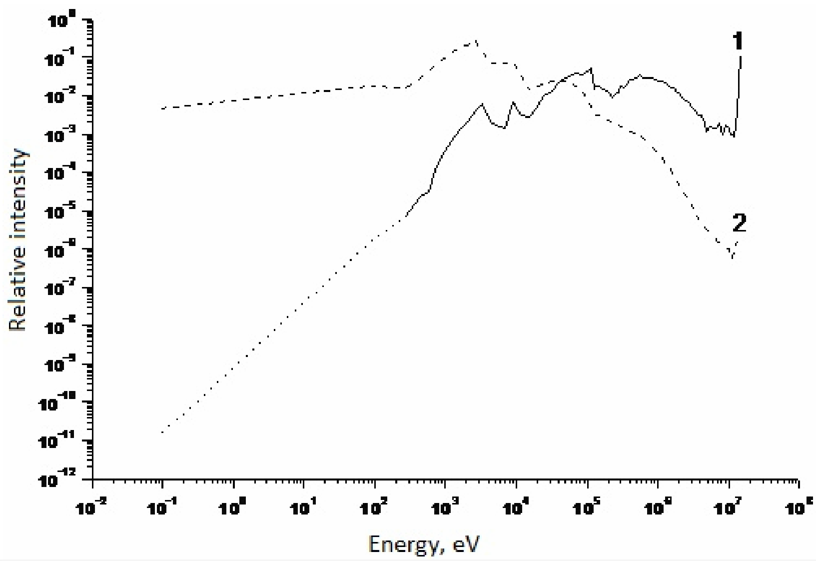

Fig. 2. Spectrum of neutrons: 1 – the neutron spectrum of the fusion reactor (continuous line), 2 – projected spectrum of neutrons in lithium carbide based on their moderation. (Intensities of the spectrum are normalized to 1).

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ИССЛЕДОВАНИЕ КАРБИДА ЛИТИЯ В КАЧЕСТВЕ ТРИТИЙВОСПРОИЗВОДЯЩЕГО МАТЕРИАЛА ДЛЯ БЛАНКЕТА ТЕРМОЯДЕРНОГО РЕАКТОРА

М.В. Аленина, В.П. Колотов, Ю.М. Платов

Проведен расчёт коэффициента воспроизводства трития (КВТ) для карбида лития (Li_2C_2). Расчётное значение КВТ для Li_2C_2 приблизительно на 33…42 % больше, чем для ортосиликата лития (Li_4SiO_4), и на 28 % больше, чем для метатитаната лития (Li_2TiO_3). Образование трития в карбиде лития по реакции $Li^6(n,T)He^4$ увеличивается на один порядок, если в расчётах учесть замедление нейтронов карбидом лития.

ДОСЛІДЖЕННЯ КАРБІДУ ЛІТІЮ В ЯКОСТІ ТРИТІЙВІДТВОРЮЮЧОГО МАТЕРІАЛУ ДЛЯ БЛАНКЕТА ТЕРМОЯДЕРНОГО РЕАКТОРА

М.В. Аленіна, В.П. Колотов, Ю.М. Платов

Проведено розрахунок коефіцієнта відтворення тритію (КВТ) для карбіду літію (Li_2C_2). Розрахункове значення КВТ для Li_2C_2 приблизно на 33…42 % більше, ніж для ортосилікату літію (Li_4SiO_4) і на 28 % більше, ніж для метатитанату літію (Li_2TiO_3). Утворення тритію в карбіді літію за реакцією $Li^6(n,T)He^4$ збільшується на один порядок, якщо в розрахунках врахувати уповільнення нейтронів карбідом літію.