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# Equivalent determination of tritium production in liquid blanket of fusion reactor using lithium isotopic abundance analysis

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## HIGHLIGHTS

- An equivalent method is proposed for measuring the tritium production in the liquid blanket of fusion reactors.
- The theoretical analysis indicates the feasibility of the proposed method.
- A chemical precipitation process is recommended for the sample pretreatment.

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## ABSTRACT

Determination of tritium production in the liquid blanket of fusion reactor is necessary for the nuclear material accounting system. In this paper, an equivalent measurement method is proposed, which is based on measuring the isotope abundance of lithium before and after irradiation. Theoretical analysis of the feasibility of the method is carried out, including complex nuclear reactions of neutron and lithium, the influence of the lead matrix, and the influence of gradient of the lithium isotope abundance before and after irradiation.



## 1. Introduction

Deuterium (D)–Tritium (T) fusion is a key subject of fusion research [1,2]. D-T fusion reactor consumes a large amount of tritium which is very rare in the natural world, and the tritium self-sufficiency in fusion reactors depends on $^{6}$Li proliferation in the blanket. In China, both tritium and enriched $^{6}$Li are nuclear materials. In accordance with the nuclear safeguard technology and nuclear material control requirements, building a nuclear material accounting system is obligatory to use or produce nuclear materials. Thus, it is essential to measure the accumulated tritium in the blanket of fusion reactors.

The liquid lead-lithium alloy is one of the prominent breeder materials for the blanket of fusion reactor [3–7]. Direct measurement of tritium in the liquid lead-lithium alloy is adopted by most of the current methods and tritium should be extracted from the liquid lead-lithium alloy in these methods [8,9]. But tritium proliferated by $^{6}$Li is tightly bound by the liquid lead-lithium alloy, and it is difficult to be extracted [8–12]. Plasma tritium in the core of a fusion reactor may also penetrate into the blanket. Therefore, it is difficult to directly measure the tritium production of the liquid lead-lithium alloy blanket [12]. In addition, tritium has a high diffusion rate for the stainless steel material, and tritium proliferated by $^{6}$Li will penetrate into the first wall through the gas solid interface reaction. Calculation of the typical DFLL-TBM model [13–20] showed that there was about 5% of total tritium resident in the blanket system. Meanwhile, there was about 12% resident in the structural material and about 84% resident in the space of system [21]. The penetration and resident of tritium in the blanket system cannot be quantitatively analyzed due to the difficulty of sampling.

In this paper, an indirect measurement method is proposed, which is based on measuring the isotopes of lithium before and after irradiation. The principle of the method is simple, the quantity of sampling is small, and the accuracy of the method is high. So it is suitable for measuring the tritium production in the liquid blanket of fusion reactors.

## 2. Methods

Based on the reaction of $^{6}\mathrm{Li} + \mathrm{n} \rightarrow {}^{3}\mathrm{H} + {}^{4}\mathrm{He} + 4.79\mathrm{MeV}$, generating a tritium atom must consume a $^{6}$Li atom at the same time. By

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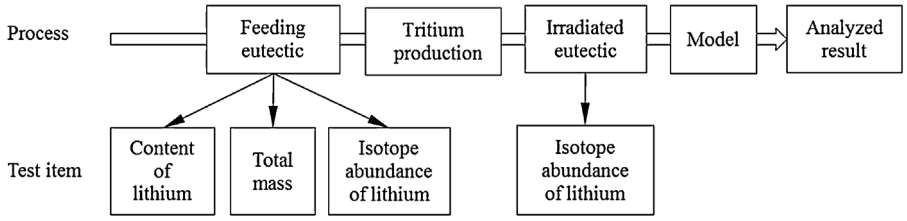


**Fig. 1.** Measurement process of the method.

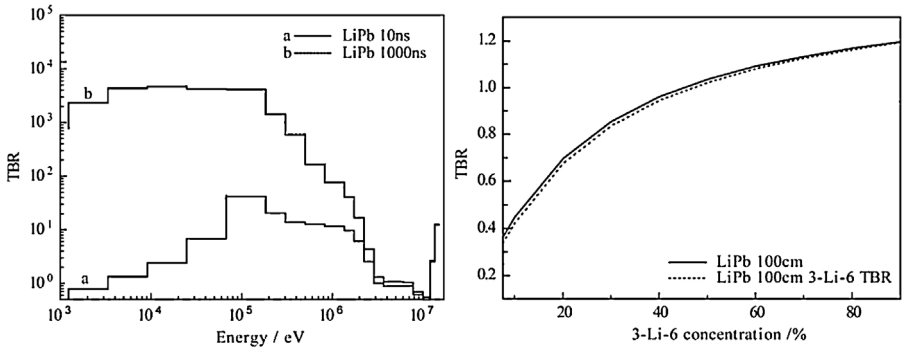


**Fig. 2.** TBR versus energy and TBR versus $^{6}$Li concentration calculated by Yi Weiwei [23].

measuring the total mass and the content of lithium in the liquid lead-lithium alloy, and measuring the lithium isotope abundance before and after irradiation by Thermal surface Ionization Mass Spectrometry (TIMS), the tritium proliferated in the liquid blanket can be calculated eventually. The measurement procedure of the method is shown in Fig. 1.

## 3. Feasibility analysis of the method

### 3.1. Key factors evaluation

The reaction between neutron and lithium atoms is very complex, including both elastic and inelastic scattering, and various neutron capture reactions. The main nuclear reaction and cross sections of neutron and lithium atoms are listed in Table 1 [22]. In the liquid blanket, the content of lead is much higher than that of lithium, and the neutron cross sections of lead is large. Yi Weiwei's research shows that the high energy neutrons will be slowed down rapidly in lead-lithium alloy below threshold of the tritium production with $^{7}$Li [23]. Therefore, the cross section of reactions in the blanket is much closer to the fission spectrum in Table 1. From Table 1, $^{7}$Li (n, n't) and $^{6}$Li (n, p) are the main reactions that may have important effect on measuring tritium production by the indirect method.

The method is based on mass spectrometry, and the tritium production is calculated by the lithium isotope abundance before and after irradiation. The interference of lead and the gradient of lithium isotope abundance before and after irradiation will play an important role in the feasibility of the method. In particular, the gradient of lithium isotope abundance before and after irradiation should be effectively identified by the mass spectrometry measurement.

### 3.2. The effect of $^{7}$Li(n, n't) and $^{6}$Li (n, p)

In Table 1, excepting for cross sections of scattering and $^{6}$Li (n, t), the cross section of $^{7}$Li (n, n't) is the largest which mostly impact the measure method.

$^{7}$Li and $^{6}$Li produce tritium from different reaction channels. Tritium produced by $^{7}$Li is the result of high-energy-neutrons reaction, and the threshold energy is close to 3MeV. Tritium produced by $^{6}$Li is mainly the result of low-energy-neutrons reaction, and the section of tritium production follows the $1/v$ law. The cross section of the tritium production of low-energy-neutrons is much larger than that of high-energy neutrons. With the slowing down of neutrons, the neutron energy spectrum is softer and $^{6}$Li is more likely to produce tritium [23].

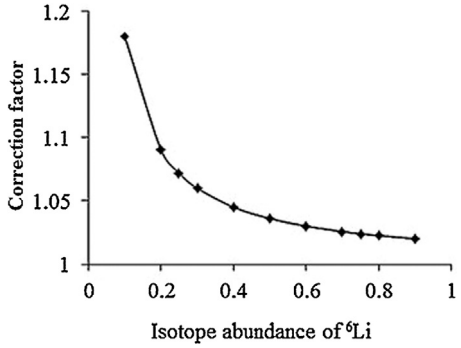


**Fig. 3.** Correction factor of $^{6}$Li/$^{7}$Li contributed by $^{7}$Li (n, n't).

The theoretical calculation results carried out by Yi Weiwei [23] indicated that after the first 10 ns, $^{7}$Li was no longer participating in producing tritium. $^{6}$Li could sustainably produce tritium for a long time. With the same particle density of $^{6}$Li and $^{7}$Li, only 1.75% of total tritium was produced by $^{7}$Li in the lead-lithium alloy, as shown in Fig. 2 [23]. If the flow channel plug and the first wall tile are present, the neutron in the blanket will be further slowed down and the share of total tritium produced by $^{7}$Li will be further reduced. Once the high enrichment $^{6}$Li is used in the liquid blanket, tritium produced by $^{7}$Li can even be ignored.

The result of mass spectrometry is in a form of $^{6}$Li/$^{7}$Li and the reaction of $^{7}$Li (n, n't) has a direct impact on the value of $^{6}$Li/$^{7}$Li. This impact is mainly determined by the abundance of $^{6}$Li before irradiation, and it can be eliminated through correction. Theoretical value of the correction factor under different initial abundance of $^{6}$Li is shown in Fig. 3. It can be seen that the correction factor

**Table 1**
Nuclear reactions between neutron and lithium.

| isotope | reaction | 14 MeV cross section | fission spectrum mean section | isotope | reaction | 14 MeV cross section | fission spectrum mean section |
|---|---|---|---|---|---|---|---|
| $^{6}$Li | capture | 82.06 μb | 28.05 μb | $^{7}$Li | capture | 1.932 μb | 6.908 μb |
| | (n,2n) | 78.05 mb | 190.5 μb | | (n,2n) | 70.09 mb | 38.53 μb |
| | (n,p) | 6.766 mb | 4.264 mb | | (n,d) | 9.763 mb | 10.98 μb |
| | (n,t) | 28.04 mb | 330.7 mb | | inelastic | 365.0 mb | 184.7 mb |
| | inelastic | 412.0 mb | 142.8 mb | | nonelastic | 444.9 mb | 184.8 mb |
| | nonelastic | 525.0 mb | 478.0 mb | | elastic | 1.025b | 1.660b |
| | elastic | 905.8 mb | 1.422b | | (n, n't) | 302.9 mb | 20.04 mb |

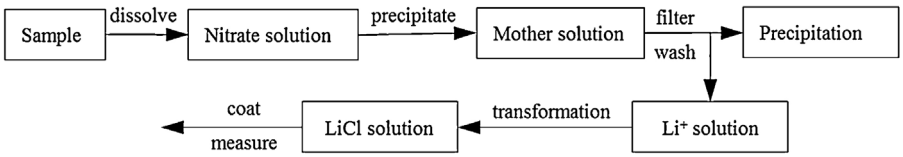


**Fig. 4.** Recommended scheme for separating lead-lithium alloy.

decreases with the increasing initial abundance of $^{6}$Li. If the initial abundance of $^{6}$Li is 90%, the correction factor is 1.02.

$^{6}$Li (n, p) reaction produces $^{6}$He which is an unstable nuclide with a short half-life, i.e. only 806.7 ms and then decays to $^{6}$Li [24]. Therefore, the change of the abundance of lithium isotope caused by $^{6}$Li (n, p) can be ignored.

### 3.3. The effect of lead

Since the relative mass difference between $^{6}$Li and $^{7}$Li is about 17%, the mass fractionation effect is obvious when measuring the isotope abundance by TIMS. This fractionation occurs mainly in the evaporation stage of the sample. The ionized material usually has a large molecular weight and a good thermal stability to reduce the mass fractionation effect and ensure the stable flow of ions, such as LiCl, $Li_2B_4O_7$, $Li_3PO_4$ and any other compounds [25]. The content of lead in the liquid lead-lithium alloy is much larger than that of lithium, the melting point of lead is low and the saturated vapor pressure is high. If the liquid lead-lithium alloy sample is coated directly, a large measurement error will appear due to the fractionation, and the evaporation of lead will contaminate the instrument. Therefore, lithium must be separated from lead before the measurement of TIMS.

Chemical precipitation process is used for separating lead and lithium from the liquid lead-lithium alloy. Kuc [26] studied how to measure tritium in the liquid lead-lithium alloy by the liquid scintillation method. Lead was separated by a chemical precipitation method and the research indicated good separation efficiency. To avoid the oxidation of scintillator, the oxidative activity of the precipitation agent used by Kuc was very weak. For mass spectrometry analysis, some precipitation agent with strong oxidative activity may be selected for better separation efficiency.

A recommended scheme for separating lead-lithium alloy is shown in Fig. 4. Lead-lithium alloy samples are dissolved at a low temperature by adding dilute nitric acid, and then concentrated ammonia is added to precipitate lead. After filtering and washing, the filtrate and wash solution become solid through heating, and then it is dissolved by hydrochloric acid. The LiCl solution is coated and measured by mass spectrometry. Since the high solubility of LiOH and low charge of $Li^+$, the co-precipitation of lithium can be neglected. In addition, the mass number of lead is much high than that of lithium, the residue of lead cannot have an important effect on measuring the lithium isotope abundance.

**Table 2**
The thick blanket scheme of HiPER.

| PbLi volume | PbLi density | PbLi compose | $^{6}$Li abundance | TBR |
|---|---|---|---|---|
| 475 m$^3$ | 9.150 g/cm$^3$ | 15.8 Li at.% | 25% | 1.1 |

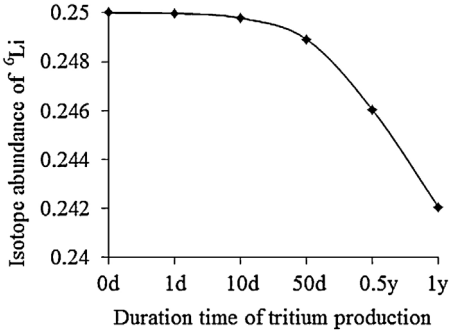


**Fig. 5.** Variation of $^{6}$Li abundance during tritium production.

### 3.4. The effect of the gradient of lithium isotope abundance before and after irradiation

The gradient of lithium isotope abundance before and after irradiation is closely related to the total load of the lead-lithium alloy, the composition of the lead-lithium alloy, the neutron flux and the duration time of tritium production. Table 2 shows the thick blanket scheme of HiPER [27], and the neutron flux in the blanket is characterized by the Tritium Breeding Ratio (TBR). Based on the data in Table 2, the relationship between the lithium isotope abundance and the duration time of tritium production is shown in Fig. 5.

From Fig. 5 it can be seen that the $^{6}$Li abundance decreases to 24.9% after 50 days, and decreases to 24.1% after 1 year. Further calculation shows that the $^{6}$Li abundance will fall to less than 15% after 10 years. The measurement of $^{6}$Li abundance by TIMS is highly sensitive. In GJB 5168, LiCl is chosen for the ionized media, the extension uncertainty of the method is 0.100% (k = 2), and the measurement range of $^{6}$Li abundance is from 4% to 99% [28]. According to this precision, the gradient of $^{6}$Li abundance can be detected accurately after tritium production for more than 50 days.

Since the nuclear material accounting cycle of tritium is very long. If it is 1 time per year, this equivalent measurement can be used for the nuclear material accounting for tritium in the blanket of fusion reactors.

## 4. Advantages and disadvantages

This method has the following advantages theoretically:

a. The mass spectrometric analysis has many advantages, such as small sample size, high accuracy, low protection requirements for the sample pretreatment, etc.;
b. It does not need to extract tritium from the liquid lead-lithium alloy, therefore the problem of the combination between tritium and lead-lithium alloy is avoided, since tritium is indirectly measured in this method;
c. Lithium atoms do not diffuse in the structure material, and the measurement value is the ratio of isotope abundance. The resident of lithium in the blanket has no effect on the measurement of tritium production;
d. Concentrated $^{6}$Li is also a nuclear material. According to the nuclear material accounting requirement of lithium in a fusion reactor, the isotope abundance of lithium is also need to be measured. So this method does not cause the additional measurement;
e. $^{6}$Li and $^{7}$Li are both stable isotopes, and the measurement of lithium sample is not limited by time.

The disadvantages of the method including:

a. Real-time measurement cannot be realized;
b. Mass spectrometry analysis has much higher requirements about instruments.

## 5. Summary

It is important for the nuclear material accounting system to measure the tritium production in the blanket of fusion reactors. To avoid extracting tritium from the liquid lead-lithium alloy and to reduce the influence of tritium retention in the blanket, an equivalent measurement method is presented in this paper. By measuring the total load of the lead-lithium alloy and the gradient of $^{6}$Li abundance before and after irradiation, the tritium production in the blanket can be obtained. Theoretical analysis of the method is also carried out. The result shows that this method is feasible and it can be applied to the nuclear material accounting system of fusion reactors.

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