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Theoretical study of Jesse effect in tritium measurements using ionization chambers

Zhilin Chen [a,b,*], Shuming Peng [a], Hanghang Lu [a], Zhaoyi Tan [a], Heyi Wang [a], Xingui Long [a], Matsuyama Masao [b]

[a] *Institute of Nuclear Physics and Chemistry, China Academy of Engineering Physics, Mianyang, Sichuan 621900, China*
[b] *University of Toyama, Gofuku 3190, Toyama-city, Toyama 930-8555, Japan*

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ABSTRACT

Jesse effect caused by impurities in helium might enhance the output signal significantly in tritium measurements with ionization chamber, which will lead to overestimation of tritium concentration in experiments. A theoretical method was proposed to evaluate Jesse effect quantitatively. Results indicate that besides Penning ionization, sub-excitation electrons also place very important influence on ionization enhancement by Jesse effect. An experiential expression about the relationship between enhancement factor and impurity concentration was established, in which second order of it fits experimental results very well. Theoretical calculation method in this paper is also applicable to evaluate Jesse effect in other kinds of mixtures besides hydrogen as impurities in helium. In addition, Jesse effects about tritium molecules as impurities have also been investigated.

1. Introduction

Tritium is one of the most important fuels in fusion research, and a large amount of tritium will be used in fuel recycling system for a fusion reactor [1,2]. Therefore, a lot of research have been carried out to find a way to breed tritium in fusion reactor to make tritium self-sustainable [3–5]. During those research, tritium measurements are of great importance to obtain tritium production rate and tritium release behavior for tritium breeding materials. Many kinds of methods have been developed for this purpose [6–8], such as thermoluminiscent dosimeters (TLD), liquid scintillation counter (LSC), secondary charged particle activation (SCPA), and ionization chamber (IC). Among those methods, TLD and SCPA are passive method and are used to measure the amount of tritium. LSC is also used to determine the total amount of produced tritium. IC is used to determine both tritium production and tritium release behavior, because ionization chamber is a real time method and can be used to measure tritium directly. And much research have been done to improve the performance of ionization chambers for tritium measurements [9–11].

In tritium recovery experiments both in-pile and out-of-pile, helium is often used as carrier gas [3,7]. To improve the tritium recovery performance, some additional gases will be mixed into helium, such as hydrogen. Although hydrogen concentration is usually very low (less than 1%), it might lead to significant increase in output signal detected by ionization chambers due to Jesse effect which was observed firstly in experiment with noble gases by Jesse in 1952 [12,13]. Many people have noticed the importance of Jesse effect on tritium measurements in such situations and they have carried out several specified experiments to examine it [14–16]. Among these work, with helium and minor hydrogen as carrier gas, Rodrigo observed in experiments that the enhancement factor at hydrogen partial pressure 10,000 ppm was as large as 1.44 times of signal obtained with pure helium, and Slagle obtained 1.42 at 5000 ppm. However, they only analyzed this phenomenon qualitatively and attributed the signal enhancement to Penning ionization in which meta-stable helium atom (He*) ionized the impurities [17]. Till now in literature, there is no explicit theory to specify the influence of Jesse effect on tritium measurements.

In this paper, influence of Jesse effect was studied in detail. Not only Penning ionization, but also sub-excitation electrons were included into the analysis. An experiential formula was introduced to fit experimental data, which can be used to calculate enhancement factor quantitatively. In addition, the effect of tritium atoms as impurities was also discussed.

2. Theory

For the energy of beta rays emitted by tritium is low, about 5.65 keV in average, tritium in gaseous form should be introduced

* Corresponding author.
E-mail address: zhilinchan@gmail.com (Z. Chen).

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into the sensitive region of an ionization chamber to perform measurements. During measurements, high voltage is applied to the chamber to ensure it works in saturation mode, in which nearly all the ions can be collected and contribute to output signal. Generally, beta ray will transfer its energy to carrier gas by ionizing and exciting. The ionization energy of helium atom is 24.59 eV, and its ground state is 1*s*2 [18]. According to Stone's work [19], there are five main excitation energy levels (He*), 1s2p, 1s3p, 1s4p, 1s5p, 1s6p, and their energies are 21.218 eV, 23.087 eV, 23.7421 eV, 24.0458 eV, 24.2110 eV, respectively. However, the ionization potential for hydrogen (I_{H_2}) is only 15.42 eV [18]. It is obvious that the meta-stable level of He* is much higher than the ionization energy of H_2. As a result, He* will transfer its energy to hydrogen by colliding, and some ions will be produced in this process, which is called Penning ionization [17]. In addition, during the ionizing process by beta rays, many free electrons will be produced, and their energy is lower than the excitation energy of helium atom. These electrons are called sub-excitation electrons [20]. The energy of some sub-excitation electrons is higher than I_{H_2}, and they have some opportunities to ionize hydrogen, too. For carrier gas composed of helium and minor hydrogen, the energy transfer process is shown in Fig. 1.

In Fig. 1, it is obvious that there are four ways to create ion pairs which will contribute to final signal output, marked as (1)–(4). Among those processes, (1) and (2) are produced by beta rays directly, while (3) and (4) are induced by sub-excitation electrons and helium atoms in meta-stable state. Comparing with helium, hydrogen molecule can be disassociated into two hydrogen atoms after absorbing energy.

2.1. *Calculation of total ions created in ionization chambers*

According to analysis above, generally, the total number of ions (N_{tot}) produced in the ionization process should be expressed as

$$N_{tot} = N_{ion-dir-H_2} + N_{ion-dir-H_e} + N_{ion-sub.} + N_{ion-pen.} \quad (1)$$

where $N_{ion-dir-H_e}$, $N_{ion-dir-H_2}$, $N_{ion-sub.}$, and $N_{ion-pen.}$ are number of ions created in processes (1), (2), (3) and (4) in Fig. 1, respectively.

2.2. *Ions produced directly by beta rays in helium and hydrogen mixture*

In Eq. (1), the first two terms are produced by beta rays directly. For helium and hydrogen mixture (hydrogen as impurity), the probability for beta ray to induce an ion pair depends on the average ionization cross-section (σ_{ave}) of them, and σ_{ave} can be

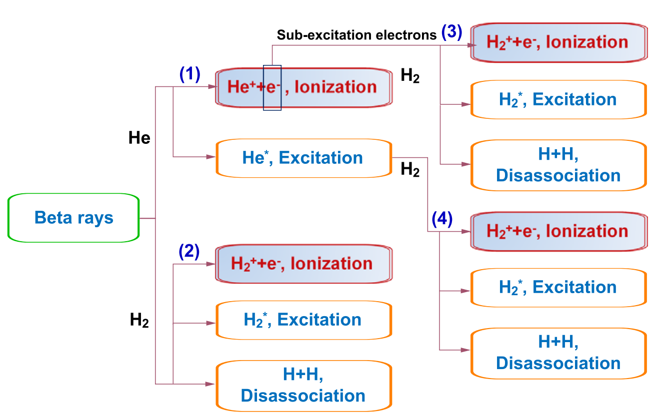

Fig. 1. Schematic of energy transfer process in helium and hydrogen.

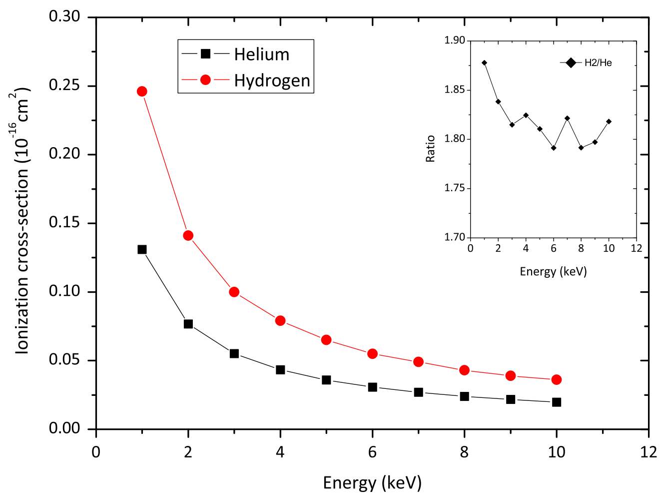

Fig. 2. Comparisons of ionization cross-section for H_2 and H_e.

calculated by the following equation:

$$\sigma_{ave} = \frac{p_{H_2}}{P} \cdot \sigma_{H_2} + \frac{p_{H_e}}{P} \cdot \sigma_{H_e} \quad (2)$$

where p_{H_2} and p_{H_e} are the partial pressure of H_2 and H_e, respectively. P is the total pressure in the chamber. σ_{H_2} and σ_{H_e} are the ionization cross-section per molecule for H_2 and H_e, respectively.

In Eq. (2), for an ionization chamber operated at given pressure, it indicates that two parameters are in charge of the final probabilities, partial pressure (p) and ionization cross-section (σ). For mixture of hydrogen as impurity in helium, the probabilities for a beta ray to ionize a hydrogen molecule can be denoted as

$$\frac{\sigma_{tot-H_2}}{\sigma_{tot-H_e}} = \frac{p_{H_2}}{p_{H_e}} \cdot \frac{\sigma_{H_2}}{\sigma_{H_e}} \quad (3)$$

The ionization cross-section (σ) depends on the energy of beta rays. For beta rays emitted by tritium (average energy of its beta ray is 5.65 keV), details are shown in Fig. 2 [21].

In Fig. 2, ionization cross-sections for both helium and hydrogen decrease gradually as energy increases. However, the ratio of cross-sections for hydrogen and helium ($\sigma_{H_2}/\sigma_{H_e}$) varies only a little, from 1.8 to 1.9. On the other hand, the partial pressure of hydrogen is very small comparing with helium in the mixture. For instance, using helium and hydrogen mixture as carrier gas in tritium recovery system, the concentration of hydrogen is usually lower than 1%. Taking both factors into consideration, it is obvious that in such situations, ions created by beta rays ionizing hydrogen directly, $N_{ion-dir-H_2}$, are negligible comparing with $N_{ion-dir-H_e}$. For partial pressure of hydrogen which is 10,000 ppm, ions devoted by ionizing hydrogen directly are less than 2% of N_{tot}. As a result, beta rays of tritium will transfer almost all of their energy directly to helium atoms. Therefore, only the latter three items in Eq. (1), ions produced by H_e^+, H_e^* and sub-excitation electrons will be included in the following calculations.

3. Calculation of the enhancement factor *K*

Taking all the three parts into consideration, the total number of ion pairs (N_{tot}) created by one beta particle can be denoted as follows:

$$N_{tot} = N_{ion-dir-H_e} + N_{ex} \cdot \eta(p) + N_{sub.} \cdot \beta \cdot \eta'(p) \quad (4)$$

where N_{ex} is the number of H_e^* (H_e atom in excitation state). $\eta(p)$ is the probability for a H_e^* to produce an ion pair by transferring its energy to hydrogen. $N_{sub.}$ is the number of sub-excitation electrons, and it is equal to $N_{ion-dir-H_e}$. β is the probability for a sub-