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Optimization of Counting Conditions on Tritium Measurement in LSC Considering Volume Effect

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Abstract: The influence of sample volume and cocktail quantity on tritium measurement in liquid scintillation counter (LSC) was investigated experimentally in this paper to optimize the counting conditions. Results indicate that the optimal counting volume is between 7 mL and 17 mL using 22 mL glass vials, which should be mainly attributed to the sensitive character of photon multiplier tubes (PMTs) in LSC. With the obtained optimal volume, we also present the detailed procedure to determine the influence of cocktail quantity, which shows that the ratio of cocktail and sample water should be kept higher than 2.4 to obtain accurate results. In addition, experiments on the influence of light were also performed with pure cocktail and results indicate that 2 h is long enough to eliminate luminescence before counting.

Key words: tritium measurement; LSC; volume effect; counting condition

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1 Introduction

Liquid Scintillation Counter (LSC) is widely used in tritium measurement due to its high sensitivity, especially for tritium in environment and materials [1–4]. However, there are many factors affecting the counting efficiency of measurement, such as vial type, cocktail, components of the sample, etc[5−6].In addition, counting loss caused by volume effect due to different sample volume in a vial is also a crucial factor on counting. Therefore, to achieve accurate measurement a counter must be operated under the optimum counting conditions. The effect of vial type and cocktail quantity on tritium measurement in LSC has been studied[7], but the results were obtained without taking account of the volume effect of the sample. Knoche et al.[8] has specified the effect of volume on ^{14}C measurement with LSC. Cocktail volume effects in $4\pi\beta$ liquid scintillation spectrometry with tritium standard efficiency tracing for low-energy $\beta-$emitting radionuclides, such as for ^{36}Cl and ^{63}Ni, have also been studied[9]. However, volume effect on tritium measurement still remains unspecified. In this article, we firstly studied the influence of light on counting, and then the effect of volume. Finally, with the obtained optimum volume, the effect of cocktail quantity has been examined to optimize the counting conditions.

2 Experiments and methods

To optimize the counting conditions, a set of samples with different volume were prepared to evaluate the effect of volume on counting and to determine the optimal counting volume. Then with optimal volume another experiment is carried out to examine the influence of cocktail quantity on counting. In both experiments, transformed spectral index of the external standard (tSIE) was employed

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to indicate quenching level of samples and quenching correction has been done automatically with a quenching curve, which was obtained by counting a set of samples with different quenching levels. Before all the tests, an experiment on the influence of light is performed firstly to determine the adapted time before counting.

The instrument used in this experiment is Tri-Carb 3100, a commercial product from PerkinElmer. Cocktails modeled Ultima Gold^{TM} from PerkinElmer, which are suitable for both aqueous and nonaqueous samples (2.5 mL deionized water per 10 mL cocktails), are used in the experiments.

2.1 Influence of light

After being irradiated by the light (both visible and invisible) in the laboratory, some molecules will be excited and emit phosphorescence whose wavelength might be appropriate to PMTs of LSC, which will significantly increase counting rates during measurements. Therefore, to prevent the influence of this unwanted emission, an experiment with pure cocktail is introduced to determine the dark adapted time needed before counting after the samples are irradiated by light during sample preparation. Therefore, all the samples are kept in a dark room for certain time reference to the results of this experiment to limit the influence of light to an acceptable level.

2.2 Quench correction curve

A set of 10 standard samples, in which the DPM (Disintegrations per Minute) of each is 286 900 (Jul 24th, 2007), are used to establish the relationship between quenching indicator (tSIE) and counting efficiency (η). Counting efficiency can be denoted by DPM and CPM (Counts per Minute), $\eta = CPM/DPM$. Among those samples, different quenching levels are achieved by adding nitro methane, which gives tSIE range from 44.7 to 756.85.

2.3 Volume effect

A group of 20 samples were prepared to examine volume effects. For those 20 samples, the total volume in each vial varies from 1 mL to 20 mL gradually. The uncertainty of cocktail quantity was limited to be less than 0.1% to minimize uncertainty due to sample preparation.

2.4 Effect of the cocktail quantity

The ratio of cocktail and sample water can be denoted by $\lambda = V_c/V_m$. V_c and V_m are the volume of cocktail and sample water respectively. With the optimum counting volume obtained above, another set of 11 samples with different λ values, which vary from 1.0 to 3.0 gradually, are tested to evaluate the influence of cocktail quantity.

3 Results and discussion

3.1 Influence of light

Fig. 1 gives the results of light influence. Theoretically, the CPM should be around 12 (the background level of the counter). However, it is obvious that the CPM is as high as 75 after about 1 min dark adapted. Counts caused by light significantly decrease in the first 20 min, then gradually down to background level after 1 h. For all the samples were prepared in the same environment, all the samples in the following experiments were dark adapted for 2 h before measurement to prevent luminescence.

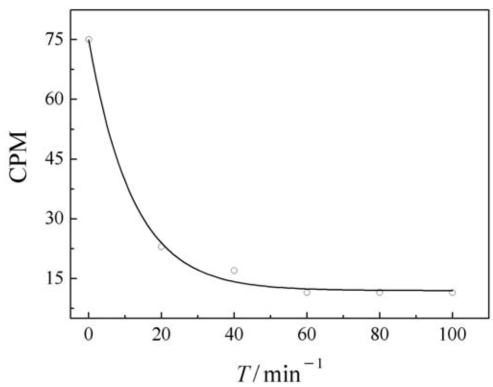

Fig. 1 Results of influence of light on counting.

3.2 Quench correction curve

The results of quenching correction curve with tSIE as quench indicating parameters (QIP) are shown in Fig. 2, which indicates that counting efficiency increases significantly with the increase of tSIE namely the decrease of quench level. With the help of this curve, tritium activity of the sample can be derived and counting loss due to quench effect can also be compensated partly after correction. Tritium activity denoted by DPM can be derived from the following formula:

$$DPM = \frac{CPM}{\eta} \ , \tag{1}$$

where η is the counting efficiency derived from Fig. 2 by linear interpolation method.

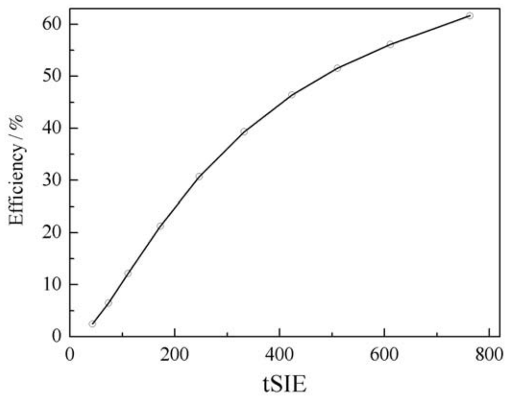

Fig. 2 Quench correction curve with tSIE as QIP.

3.3 Volume effect

Results of volume effect on counting are shown in Fig. 3, in which y-axis is counts per minute per volume of the sample. It shows an increase at volume less than 9 mL, then comes to a plateau from 9 mL to 12 mL, and decreases as volume gets larger than 12 mL. The value at 1 mL is about 20.60% less than the maximum value and counts at 20 mL is about 10.00% less.

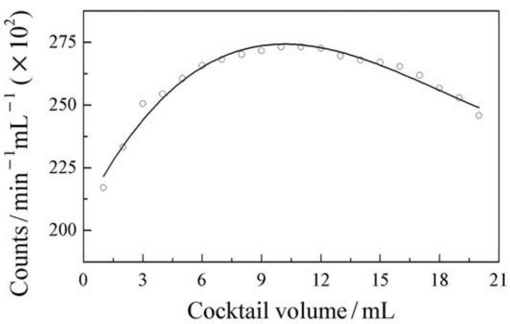

Fig. 3 Results of different volume without quench correction.

Theoretically, for the tritium concentration dispensed into all the samples is the same, it should give a constant response in Fig. 3. However, it shows a significant different one, which might be mainly caused by both quench effect and volume effect. To preclude the influence of quench, quench correction with data provided in Fig. 2 is applied to modify the results. Fig. 4 shows the relationship between tritium concentration and volume after quench correction. It is obvious that, after correction, tritium concentration gives a plateau between 7 mL and 17 mL (within 1%), which is much wider than before. However, tritium concentration at 1 mL is still 3.42% less and 4.15% less at 20 mL.

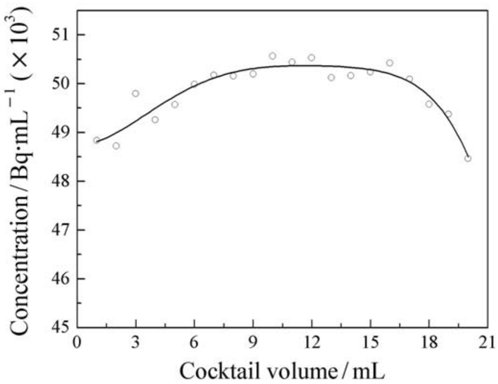

Fig. 4 Results of different volume after quench correction

In fact, considering the quench effects have been compensated, counts loss in Fig. 4 should be attributed to geometrical volume effect, which is greatly affected by the character of PMTs in LSC. There are two PMTs used in this measurement system and they do not give uniform response to photons on their surface, more sensitive at the center of the surface while weaker outside. Considering this character of PMT, counts loss due to volume effect can be illustrated in Fig. 5, where the sample volume in Fig.5 (a) is small, and the major number of photons emitted from the sample can not reach the most sensitive region of both PMTs (central part in the figure), which directly lead to the counts loss of a big portion, as shown in Fig. 4. As volume increases, the major part of photons gradually cover the sensitive region, and then counts reach its maximum value, shown in Fig.5 (b). However, once volume excesses a certain level, there come two problems which will contribute to the loss of photons. One part of photons will reach the surface above the sensitive region and another part will reach the cap of the vial, which will significantly decrease the number of photons received by PMTs, as shown in Fig. 5(c).

Therefore, to avoid the influence of volume effect, proper volume should be chosen in the measurements.

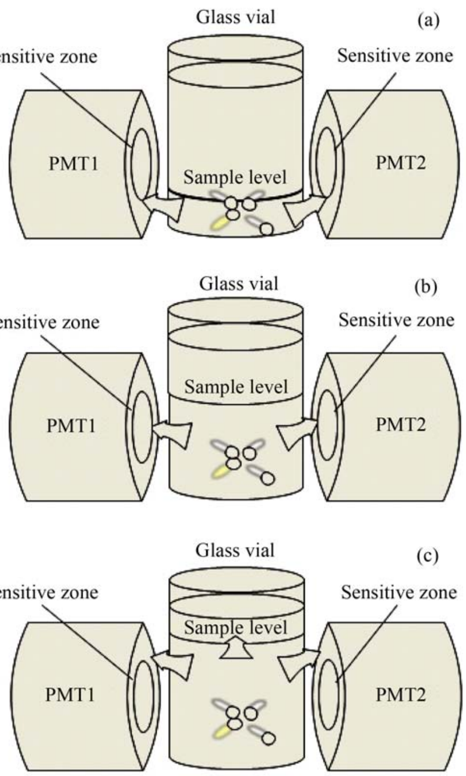

Fig. 5 Schematic diagram of count loss due to response character of PMTs.

3.4 Effect of the cocktail quantity

Taking account of volume effects, sample volume of 12 mL was chosen to evaluate the influence of cocktail quantity on tritium measurements, and results are shown in Fig. 6, where the tritium concentration increases as λ increases, especially at low values, then gradually becomes nearly constant. In Fig. 6, tritium concentration increases greatly from 1.0 to 2.4, as much as 26.60%, while after λ getting higher than 2.4 it varies less than 1%. Although tritium concentrations are obtained after quench correction, the lower values shown in Fig. 6 should be caused by the limited quench correction ability of quench correction method at low λ values, as explanation in reference[7].

Comparing with results obtained without taking account of volume effect[7], λ should reach 2.4 to meet the optimum counting condition, other than 2.0 in former experiments. The difference should be mainly attributed to the fact that values of tSIE vary at different volume due to volume effect.

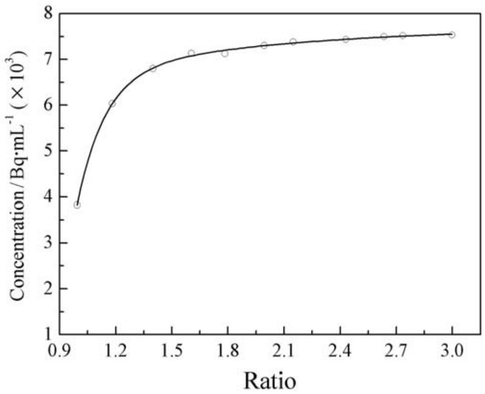

Fig. 6 Effect of cocktail quantity on measurement with optimal volume.

4 Conclusion

Results of the volume effect experiment indicate that sample volume should be limited within 7 mL and 17 mL to avoid counting loss mainly due to the defects of PMTs. Considering volume effects, the ratio of cocktail and sample water must reach 2.4 to optimize counting. By the way, a certain time should be dark adapted before counting to prevent luminescence according to light irradiation conditions. However, all the optimizations noted above might vary a lot between different counting systems. Therefore, to perform accurate measurements, experiments should be done with methods introduced in this paper to obtain its own optimum counting conditions.

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考虑体积效应的液闪测氚计数条件优化研究

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摘要： 采用外标转换谱指数(tSIE)作为淬灭校正因子，通过研究闪烁液及样品总体积、闪烁液用量的影响获得液闪测氚时的最佳计数条件。结果表明，对于22 mL标准玻璃计数瓶，最佳计数体积为7～17 mL，体积过少或过多可导致高到10%以上的测量误差，这主要是由液闪谱仪光电倍增管的面响应特性引起的。在获得的最佳计数体积基础上，对闪烁液的用量进行了研究。为了获得好的测量结果，闪烁液与样品的比例应高于2.4。另外，还通过实验研究了光照对测量结果的影响。在实验室条件下，实验室灯光及自然光所引起的本底辐射在样品静置2 h后将不会对测量结果构成影响。

关键词： 氚测量；液闪；体积效应；计数条件

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