ABSTRACT
The most commonly method for determining $^{222}\text{Rn}$ in water is always calibrated with a $^{226}\text{Ra}$ standard solution, which have a long term and also have not been purified. $^{226}\text{Ra}$ standard solution usually contains several progenies such as $^{210}\text{Pb}$, $^{210}\text{Bi}$ and $^{210}\text{Po}$. But there are high efficiency instrument to solve this problem, which ultra-low liquid scintillation counter has been had pulse shape analysis (PSA) for separate alpha and beta assess the activity of $^{226}\text{Ra}$ standard solution using two different organic cocktails. The least interference of alpha and beta have been selected PSA level at 175.
INTRODUCTION

Since the early 1950s, liquid scintillation counting (LSC) has been a very popular technique for the detection and quantitative measurement of low energy beta radioactivity (L’Annunziata, 1998). LSC have a coincidence system, geometry essentially $4\pi$ and give the high counting efficiency (nearly 100%), those was a good advantages. Moreover, ultra-low level liquid scintillation counting (Wallac 1220 Quantulus) coupled to alpha-beta discrimination or called pulse shape analysis (PSA) allows rapid radioactivities measurement and simple separation of alpha and beta spectrums analysis, that are simultaneously measured through alpha-beta discrimination technique. PSA is used to discriminate long scintillation events from the shorter scintillation of true beta events. PSA values are adjustable within a finite range and the optimum value. Therefore, PSA is necessary condition to ultra-low liquid scintillation counter that alpha backgrounds are greatly reduced and representative of the best energy resolution at lower quench levels.

$^{226}$Ra have a long half-life (1602 years), which contains 9 daughter radionuclides ($^{222}$Rn, $^{218}$Po, $^{214}$Pb, $^{214}$Bi, $^{214}$Po, $^{210}$Pb, $^{210}$Bi, $^{210}$Po, $^{206}$Pb), undergoes a decay chain: 5 $\alpha$-decays, 4 $\beta$-decays, accompanied by emission of $\gamma$-rays and ends with a stable isotope of lead ($^{206}$Pb). The $^{226}$Ra standard solution was popular to use in calibrating the method for determining $^{222}$Rn in water (ASTM, 1998). If the standard has not recently been purified that have interference from the long-lived progenies ($^{210}$Pb, $^{210}$Bi and $^{210}$Po) cause these progenies depending on the time elapsed from standard preparation and purification to the calibration (Pates et al., 2006). Therefore, the actual activity of the standard is very important.

In this work, we propose a first step that can be considered optimum PSA for alpha/beta separation and divided interference in alpha or beta event. In addition to, examined the interference from $^{210}$Pb, $^{210}$Bi and $^{210}$Po in calibrating the method for $^{222}$Rn in water (Salonen, 2010). The $^{226}$Ra standard solution were prepared in two different organic cocktails. The measurements were performed with a ultra-low level liquid scintillation counting (Wallac 1220 Quantulus) using alpha/beta PSA to separately assess the interference.

EXPERIMENTAL PROCEDURE

1. Apparatus

All samples were counted by ultra-low level liquid scintillation counting, using a Wallac model 1220 Quantulus from Perkin Elmer (Turku, Finland), which an external spectral standard of $^{152}$Eu that allows to measure external spectral quench parameter (SQP[E]) to quantify quenching, and uses PSA for alpha/beta separation. Additionally, background is reduced by guard, which is an external radiation event detector, and coincidence event detection.

2. $^{241}$Am, $^{90}$Sr and $^{226}$Ra standard

The $^{241}$Am standard was prepared from a standard solution with a specific activity of 8.799 kBq/g overall uncertainty of ±3.2%. Two consecutive dilutions were made up in 0.1 M hydrochloric acid with 10 µg/ml of Eu in order to reduce its specific activity to 38 ± 0.5 Bq/g in the final dilution used in this study. Meanwhile, the $^{90}$Sr standard was prepared by three steps dilutions continuously. The specific activity of the $^{90}$Sr standard solution was 0.3711 ± 3.4 MBq/g. Eventually, $^{90}$Sr had specific activity to 37
± 0.4 Bq/g. At the same time, \(^{226}\)Ra standard was diluted two step in order that finally had specific activity 24 ± 0.4 Bq/g from \(^{226}\)Ra standard solution (specific activity 9.106 ± 3.5 kBq/g). Furthermore, long term keeping \(^{226}\)Ra standard solution (26 years) will be used for radioactivity correction examined.

3. Cocktails and vials

The scintillation cocktails used in this work are Ultima Gold AB and Optiphase Hisafe 3 (PerkinElmer). Those cocktails employ the same solvent (id-isopropynapthalene ; DIN). Ultima Gold AB have been specifically design for alpha/beta separation but Optiphase Hisafe 3 have a broad range of solutes also used for a variety of scintillation application. The samples were prepared in low-potassium borosilicate glass vials (PerkinElmer)

4. PSA optimization

The optimum PSA levels for alpha/beta separation were determined using \(^{241}\)Am standard (alpha emitting) and \(^{90}\)Sr standard (beta emitting). The samples prepared by those standard weighing 1 g with analytical balance into glass vials and was add 19 ml of scintillation cocktail was capped tightly. A comparable background was prepared with de-ionized water for each standard. On the another hand, the mix sample was prepare by 1 g of each standard in the same vials then add 18 ml of scintillation cocktail. The samples were shaken by hand and measurement by ultra-low level liquid scintillation counting used range of PSA setting from 100 to 220. The optimum PSA was obtained when the beta spillover in the alpha window on the right-hand side of the alpha peak was as low as possible and simultaneously the alpha spillover peak on a continuous beta spectrum was almost invisible. The PSA level was examined in 60 minutes and the count rate will be evaluated the optimum PSA by calculate Interference of alpha and beta by the equation (Meesat, 1998):

\[
T_\alpha = \frac{C_\alpha}{C_\alpha + C_\beta}
\]

Where \(T_\alpha\) is interference of alpha in the beta event, when beta emitter form \(^{90}\)Sr standard sample, \(C_\alpha\) is count rate of alpha (cpm) and \(C_\beta\) is count rate of beta (cpm).

\[
T_\beta = \frac{C_\beta}{C_\alpha + C_\beta}
\]

Where \(T_\beta\) is interference of beta in the alpha event, when alpha emitter form \(^{241}\)Am standard sample, \(C_\alpha\) is count rate of alpha (cpm) and \(C_\beta\) is count rate of beta (cpm).

5. Assurance of the activity concentration of \(^{226}\)Ra standard solution

The activity concentration of \(^{226}\)Ra standard solution was confirmed by an alpha/beta separation collaborated with solve equations. the sample was prepare by 1 g of standard add scintillation cocktail to 20 ml then measurement with LSC.

RESULTS AND DISCUSSION

1. The optimum PSA of the ultra-low level liquid scintillation counter

The PSA level of ultra-low level liquid scintillation counter (Wallac 1220 Quantulus) has been using \(^{241}\)Am and \(^{90}\)Sr standard sample. The alpha window was determined by visual examination of the LSC spectrum. Figure 1, illustrate Ultima Gold AB being less quenched than Optiphase Hisafe 3 because of the alpha region of interest (ROI) that means the alpha window in the alpha spectrum. Ultima Gold AB have alpha spectrum range from 580 to 730 but Optiphase Hisafe 3 occurred from 550
Quenching causes the spectrum to be shifted to lower energy. The interference of alpha in the beta event had been decreasing when PSA level is increase.

Conversely, if PSA level will increase then the interference of beta is lower as show in Figure 2. On the other sides the mix sample had slightly different result, which alpha/beta separation occurred in the low level of PSA that cause of the sample have simultaneously alpha/beta emitting. Therefore, in order to achieve for alpha/beta separation have to collaborate with ROI. Consequently, the optimum of PSA considered on minimum interference of alpha into beta and beta into alpha that influence of both cocktail (Ultima Gold AB and Optiphase Hisafe 3) have PSA level at 175 is the most appropriate.

2. $^{226}$Ra standard solution

Figure 3, indicate that the accumulation of $^{210}$Pb, $^{210}$Bi and $^{210}$Po in the $^{226}$Ra standard solution will cause a calibration error when $^{222}$Rn in water is measured. The error depends on the time elapsed from the $^{226}$Ra standard purification to the calibration and on the efficiencies with these progenies. The activity of $^{210}$Pb, $^{210}$Bi and $^{210}$Po have accumulated in $^{226}$Ra sample are equal and are calculated by the equation

$$A_{Po} = A_{Bi} = A_{Po} = A_{Ra}(1-e^{-\lambda t})$$
Where $A_{Ra}$ is activity of $^{226}$Ra in sample (dpm), $t$ is time $^{226}$Ra standard purification to the calibration (y), $\lambda$ is decay constant of $^{210}$Pb (0.693 / 22.3 y$^{-1}$).

For two different in-growth times of $^{226}$Ra (1 and 26 years). The truly activity of $^{226}$Ra obtained from Table 1. The long term $^{226}$Ra standard able to resolved the correct value as well as for the fresh $^{226}$Ra standard the count rate show the normally. Hence, the equation also help estimated real activity of long term $^{226}$Ra standard solution for calibration method of determinate $^{222}$Rn in water.

Figure 3. The alpha/beta separation of $^{10}$Pb, $^{210}$Bi and $^{210}$Po (in equilibrium) at the optimum PSA. The sample was $^{226}$Ra standard solution with Ultima Gold AB.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$1-e^{-\lambda t}$ (Bq/20ml)</th>
<th>Percentage Efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ultima Gold AB</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{226}$Ra 1 year</td>
<td>24.481</td>
<td>92.479</td>
</tr>
<tr>
<td>$^{226}$Ra 26 years</td>
<td>25.267</td>
<td>92.479</td>
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</tbody>
</table>

Optiphase Hisafe 3

<table>
<thead>
<tr>
<th>Sample</th>
<th>$1-e^{-\lambda t}$ (Bq/20ml)</th>
<th>Percentage Efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{226}$Ra 1 year</td>
<td>24.434</td>
<td>91.648</td>
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<tr>
<td>$^{226}$Ra 26 years</td>
<td>25.154</td>
<td>91.648</td>
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</tbody>
</table>
CONCLUSIONS

This study demonstrated that PSA was very important to increased ability of alpha/beta separation and the optimum PSA was selected by conditional of interference that selected at 175 for both of cocktails by using ultra-low level liquid scintillation counting (Wallac 1220 Quantulus). Optiphase Hisafe 3 had slightly quenching than Ultima Gold AB in all PSA level. The long term $^{226}$Ra standard solution containing $^{10}$Pb, $^{210}$Bi and $^{210}$Po that raise alpha and beta count rate. Then used the optimum PSA with the equation to solve the real activity that very useful for calibrated method.

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REFERENCES


