Liquid scintillation determination of uranium ($^{234}U/^{238}U$) from water by coctail extraction

Key words: C-14, radioactive bicarbonate, phytoplankton

Introduction

It has been shown that the complexing agent bis(2-ethylhexyl)phosphoric acid (HDEHP) can be added to commercial lipophilic diisopropylnaphthalene (DIN) based liquid scintillation cocktails, making them able to extract uranium and other actinides from water (ref. 1). Ideally, the water pH should be adjusted between 1-2. Suitable cocktails are, e.g., OptiScint HiSafe and Betaplate Scint from Wallac, added with a few per cent of HDEHP. A few milliliters can efficiently extract uranium from several hundreds of milliliters of water.

The cocktail extraction combined with alpha/beta separation capability of Triathler results in a simple and sensitive uranium detection method. Advantageously, DIN solvent possesses good alpha/beta separation properties

Procedure

The natural water samples should first be thoroughly bubbled with a radon-free gas or air to remove all 222Rn. The extractive cocktail is then added on the water sample in an appropriate container and the system is vigorously shaken for about two minutes. After this, the mixture is let stand for about one hour during which the water and cocktail phases separate, the latter coming buoyant on the water. (Centrifuging, if possible, will obviously greatly speed the phase separation). Finally, the cocktail (or an aliquot of it), now containing the uranium, is collected and counted.

The procedure was applied to two test systems with different water/cocktail ratios:

T1): 100 mL of water + 5 mL of extractive cocktail (20-fold water excess)

T2): 200 mL of water + 5 mL of extractive cocktail (40-fold water excess)

- Water: tap water, both T1 and T2 with the same amount of U-standard solution (234U / 238U = 1); pH adjusted to 1.3 with nitric acid (HNO3).
- Extractive cocktail: Betaplate Scint added with 6 % (v/v) of HDEHP.

After phase separation, cocktail aliquots of 1 mL in Eppendorf tubes and 3 mL in minivials were collected for measurements.

Results

Fig. 1 shows the two-dimensional graph (x-axis: pulse amplitude, y-axis: pulse length) obtained with Triathler from the extracted uranium sample. It is seen that the alpha spot is well separated from the beta band. With this graph one can select a region occupied only by alphas and exclude betas.

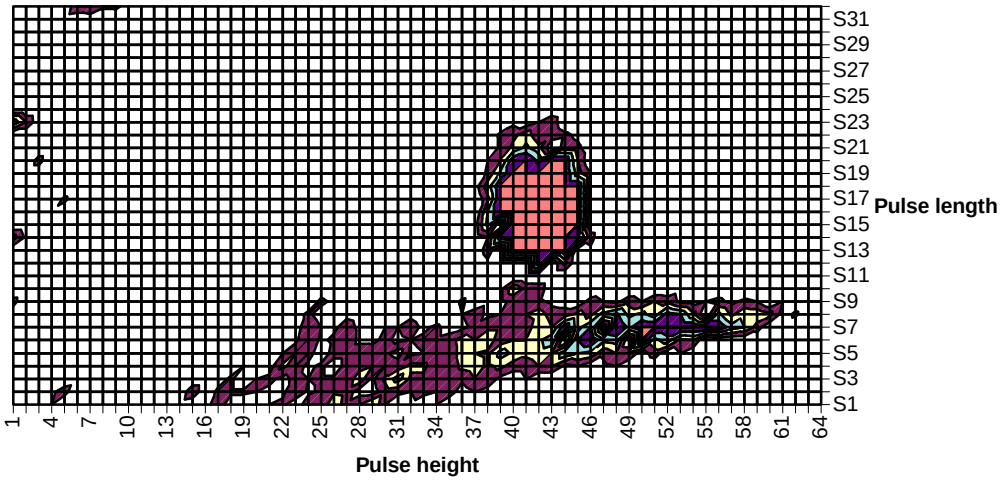

Fig. 1. The two-dimensional graph of the extracted 234U/238U sample X-axis represents pulse amplitude (height) and y-axis its length.

The separated alpha and beta spectra from the 1 mL Eppendorf tube are in Fig. 2. It is seen that the 238U and 234U peaks are partially resolved, allowing assessment of their ratio. Somewhat poorer resolution was obtained with 3 mL samples in minivials.

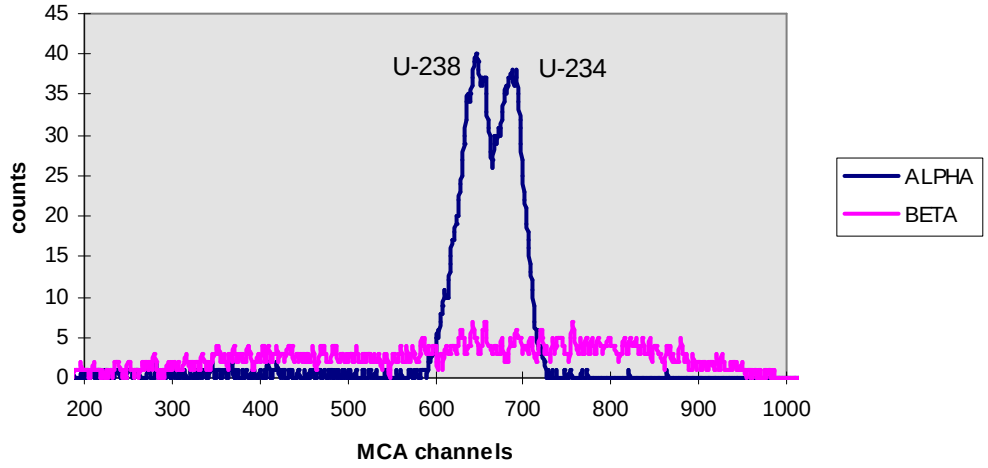

Fig. 2. The alpha and beta spectra of 234U/238U sample extracted from water into the cocktail (Betaplate Scint with 6 % of HDEHP) and counted as 1 mL volume in an Eppendorff tube.

Extraction Efficiency and Sensitivity

It was observed that both T1 and T2 gave practically identical results with the observed alpha CPM >90 % of the expected DPM, indicating quantitative extraction. This suggests that even greater water volumes (or smaller cocktail volumes) could be used while still achieving efficient extraction.

The alpha background (extractive cocktail without U) under the uranium peak pair was near 0.1 CPM for 1 mL samples in Eppendorf tubes and near 0.2 CPM for 3 mL samples in minivials. A crude approximation of lower limit of detection (LLD) in Bq/L can be obtained from the formula below:

$$LLD = 3\sqrt{b/t} * 1/V * 1/60 * C/c$$

where,
b = background in CPM
t = counting time in minutes
V = water volume in liters
C = cocktail volume used in extraction
c = cocktail volume collected for counting

For example, the values b = 0.2 CPM, t = 30 min, V = 0.2 L, C = 5 mL and c = 3 mL, give LLD less than 0.035 Bq/L. Still lower LLD can be reached with longer counting time, greater water volume (if possible) and minimized C/c ratio.

Conclusions

Cocktail extraction and counting with Triathler offer several benefits:

Cocktail extraction and counting with Triathler offer several benefits:

- On-site or near-site measurements become possible allowing fast decision making and flexible research plans.
- Cocktail volumes can be reduced to milliliter level for each sample, cutting costs and amount of waste.
- Large water volumes mean sensitive assays.

References

1. Venso E.A. et al., in Liquid Scintillation Spectrometry 1992, ed. Noakes J.E., Schönhofer F. and Polach H.A. RADIOCARBON 1993, pp. 425-430