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Proposed Procedure for Salt Purification

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1. Introduction

Some 2.5 tons of salt is required with a ^{232}Th (or equivalent for daughters) concentration of $< 2 \times 10^{-13}$ by mass, corresponding to $< 0.5 \times 10^{-15}$ when dissolved in 1000 tons of D_2O. Recent reports to the Water Group (20th June and 28th August 1992) have indicated that the purest salt available has a thorium concentration of about 2×10^{-9}.

The favoured, and possibly the only, procedure for purification is the absorption of thorium and radium from a 5% salt solution by finely divided materials such as MnO_2 and HTiO. As always the critical requirement is an assay technique of sufficient sensitivity which for a 5% solution would be 10^{-14} thorium (or equivalent). The uranium chain is not being ignored but is an order of magnitude less demanding, and if the thorium chain is assayed via the extraction and α-counting of 3.66 day ^{224}Ra then the 1600 year ^{226}Ra of the uranium chain will also be measured. Generally the requirements for purification and assay of 5% salt in solution are similar to those for D_2O or D_2O plus 0.25% salt, and it is hoped that the same techniques can be applied in all cases.

Assay at the level of 10^{-14} or 10^{-15} is formidably difficult and we are unlikely to have full confidence in the measurements unless different techniques are applied and shown to agree. It would be particularly attractive to determine ^{232}Th by mass spectrometry, specifically ICPMS-ETV, and compare with chemically extracted ^{224}Ra or its daughter 55 sec. ^{220}Rn (thoron). Any disequilibrium can be normalized out by comparing chemically extracted ^{232}Th and ^{228}Th by α-counting, since ^{228}Th must be in equilibrium with 3.66 day ^{224}Ra. There is a serious risk at pH7 that thorium will deposit temporarily on the surfaces of pipes and vessels releasing radium into solution, and it may be desirable to acidify the salt solution to pH4 or 5 during purification.

2. Summary of Development Work in Oxford

The efficacy of various absorbers including MnO_2 and HTiO has been studied by batch contact with satisfactorily large decontamination factors and no evidence of saturation, both for pure H_2O and 5% brine. Concentrations as low as 1 ppm HTiO have been shown to be effective.

Attention has now moved to tests of radium extraction using small tangential filtration rigs with a throughput of 10 to 100 ml/min. fed from a reservoir with 0.01 to 1% HTiO. Decontamination factors > 100 have regularly been observed both with organic and inorganic membrane with a cut-off of $\sim$ 40,000 Daltons. It has emerged that the most economic prospect in terms of space, cost and power is to deposit the HTiO on the membrane surface with a density of $\sim 1 g/m^2$ and to recover quantitatively the absorbed radium by subsequent washing with 0.1M HCl which (to 99% it is believed) leaves the HTiO in place. The acid can be neutralized and the radium further concentrated by a second stage of absorption and filtration, ready for drying for thoron emanation or elution and preparation of sources for α-counting. All of the stages of this procedure have been tested separately on a small scale.

At present filters of various materials, pore sizes, and deposits of HTiO are being investigated to determine the maximum throughput that can be obtained with a satisfactory decontamination factor. This will settle the details of a pilot plant with a minimum throughput of 2 l./min., which it is planned to build over the next few months. Such a pilot plant should be capable of establishing the operational characteristics of the radium concentration technique down to an equivalent thorium level of 10^{-13} and to deliver 50 kg of purified salt in 5% solution, providing everything scales simply with

volume of water and concentration of radium which on the whole is unlikely. It is anticipated that the work on the pilot plant will take 6 months to a year and represents something like the limit of our resources in terms of funds and effort.

It is supposed that the development work above would be carried out with H_2O but, if the development work is satisfactory, it will eventually be necessary to carry out tests with D_2O for which about 100 l. will be required. There will be some losses and degradation, respectively estimated at 0.1 l. and 0.1%, but these should not be significant on the scale of 1000 tons.

3. Proposal

It is proposed that the 2.5 tons of salt should be purified and assayed as a 5% solution in 50 tons of D_2O using a 10 l./min. tangential filtration system with the membrane coated with HTiO, the design being based on the one fifth scale H_2O pilot plant developed in Oxford.

The alternative would be to purify a 5% H_2O solution in a 10 l./min. system, dry the salt at some cost and risk of contamination, and re-dissolved in D_2O. We are nervous about maintaining 10^{-13} thorium purity during the drying process, which of necessity would have to be contracted out to industry, with no possibility of verifying the cleanliness without re dissolving the salt.

The salt as delivered comes as a concentrate solution/slurry and will have to be dried, but at that stage it is contaminated with thorium at $\sim 10^{-9}$ and it is unlikely that much harm would be done. Oxford would expect to attend to the arrangements for this drying procedure and check redissolution.

It is noted that the 10 l./min. D_2O/NaCl system proposed would look like a one tenth pilot plant for the purification/assay of 0.25% D_2O brine at 100 l./min. which will be eventually required.

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