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Seeded Tangential Filtration: Progress and Prognosis

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1. Introduction

As a qualification for filtration the adjectives "tangential", "ultra", and "cross' are synonyms meaning a filtration system for a liquid containing finely divided solids which is recirculated at pressure from a reservoir through porous tubes at a velocity of order $1 ms^{-1}$, and the solid-free permeate collected from the outer surface of the tubes. It is seen as a potential technique for concentrating radium and thorium in water onto small masses of the powerful absorbers developed at AEA Technology, Harwell, and elsewhere.

The absorber materials are tested by "batch contact", typically a few hundred ml of spiked water shaken with a few tens of ppm of absorber for some minutes and then centrifuged to separate the solids from the water. The measures of the absorber quality is the decontamination factor, D/F, the ratio of the water activity before and after contact with the absorber, and the contact time necessary to achieve a satisfactory D/F.

The previous report (Lilley and Omori, April 1992) concentrated on the removal of ^{234}Th from NaCl solutions, mostly via batch contact but also with tangential filtration. Thorium is readily removed but at pH7 the chemistry is more complicated than simple attachment to an absorber.

This report is about the extraction of the tracer ^{226}Ra by tangential filtration and is mainly concerned with establishing the gross features. There has been no fine tuning and in a number of respects the measurements are considered incomplete.

The plumbing arrangements are shown in the sketch attached.

2. Tangential Filtration of Radium

Two filter types have been tested:

(i) Carbosep M4

Graphite tube coated internally with zirconia, bore 6mm ϕ, pore size about 10nm. For the measurements a single tube 20cm long was used.

(ii) Amicon HP30 43

A cartridge of 55 polysulfone tubes 20cm long and 1.1mm bore, with a nominal MW cut-off of 30,000 which is believed to correspond to about 10nm pore size.

In general the following conditions have been maintained:

- reservoir 0.6l., maintained constant
- recirculation 1l/min (non-turbulent)
- pressure 1.5 bar
- absorber HTiO
- absorber concentration 100ppm of reservoir
- tracer ^{226}Ra (some KBq added to reservoir)
- tracer samples, 50ml, recorded via 186 keV γ-ray and Ge-counter
- water, Millipore 10 Mohm cm for all serious purposes, Elga deionized or tap water for flushing,
- pH7
- temperature, ~ 25°C
- acid wash, ~ 100ml, 3M HNO_3 for Carbosep, 0.1M HCl for Amicon
- permeate flow, 6ml/min for Carbosep, 40ml/min for Amicon (80ml/min for Amicon without HTiO).
- final concentrate, ~ 100ml in reservoir plus hold up volume (~ 100ml).

Without any HTiO absorber ^{226}Ra simply passed through the filter membrane without any inhibition. With HTiO no 186 keV γ-rays significantly above background were detected from any permeate, even after 10l. of water had been passed (which is a long time at 6ml/min). ^{226}Ra α-decays were detected in the permeate, but it was unclear whether or not this Ra originated with contamination. The decontamination factor was at least 100.

The ^{226}Ra content of the reservoir decreased with the volume of water passed through the filter in much the same way for both Carbosep and Amicon. It was observed qualitatively that the turbidity of the reservoir decreased with time/permeate volume but that could as well be caused by the break-up of the HTiO to a smaller grain size as by the deposition of HTiO within the filter system; no provision had been made for monitoring the HTiO content of the reservoir and that will have to be rectified in the future. The lowest ^{226}Ra content of the reservoir observed was 3.6% of the initial load, after 12l. and 35 hours via Carbosep; doubling the recirculation flow and the pressure put up the reservoir content to 12%, draining down and back-washing yielded another 2% only. The ^{226}Ra was recovered by acid washing and in general 90% or more of the ^{226}Ra could be accounted. The corresponding story for Amicon is 12% in the reservoir after 11 litres of permeate, nothing in the permeate, 2% recovered by back washing, and the rest by acid washing.

The problem of "absorptive losses' from dilute solution is well known and Amicon do not recommend going below 4000ppm concentration. The obvious (but inconvenient) solution to the recovery problem is to reduce the volume of the reservoir and maintain or increase the mass of HTiO, but that will have to be investigated as it also changes the contact time and possibly the throughput.

The properties of Carbosep M4 and Amicon HI P30 43, do not differ dramatically. Even the permeate rates are similar if allowance is made for the possible 8 bar operating pressure for Carbosep c.f. 1.7 bar maximum for Amicon. With 100ppm HTiO both take out Ra effectively and both lock up the Ra within the filter, very probably attached to HTiO absorbed on the filter surface.

3. Prognosis

It is proposed to pursue the tests with the existing Carbosep and Amicon filters, in particular the recovery of HTiO, and to measure the radio-activity of Carbosep which may rule out the use of Carbosep. There will then be two decisions to be made:

(i) organic v. inorganic filters, and

(ii) the manner in which radium can be recovered from a filter.

The next stage would be the planning of a pilot plant for at least one ton and possibly ten tons, and in parallel developing the techniques for the further concentration of radium.

The detailed design and manufacture of the pilot plant could be pursued while testing other candidate filtration membranes, particularly those of larger pore size and higher throughput, and subsequently subjecting the preferred membrane to comprehensive tests with respect to:

— pH

— pressure

— temperature

— recirculation flow

— absorber type

— absorber concentration

— NaCl content

— Ra, Th and Pb

Finally it will be necessary to commission the pilot plant and determine its characteristics.

14.Questions Needing Answers

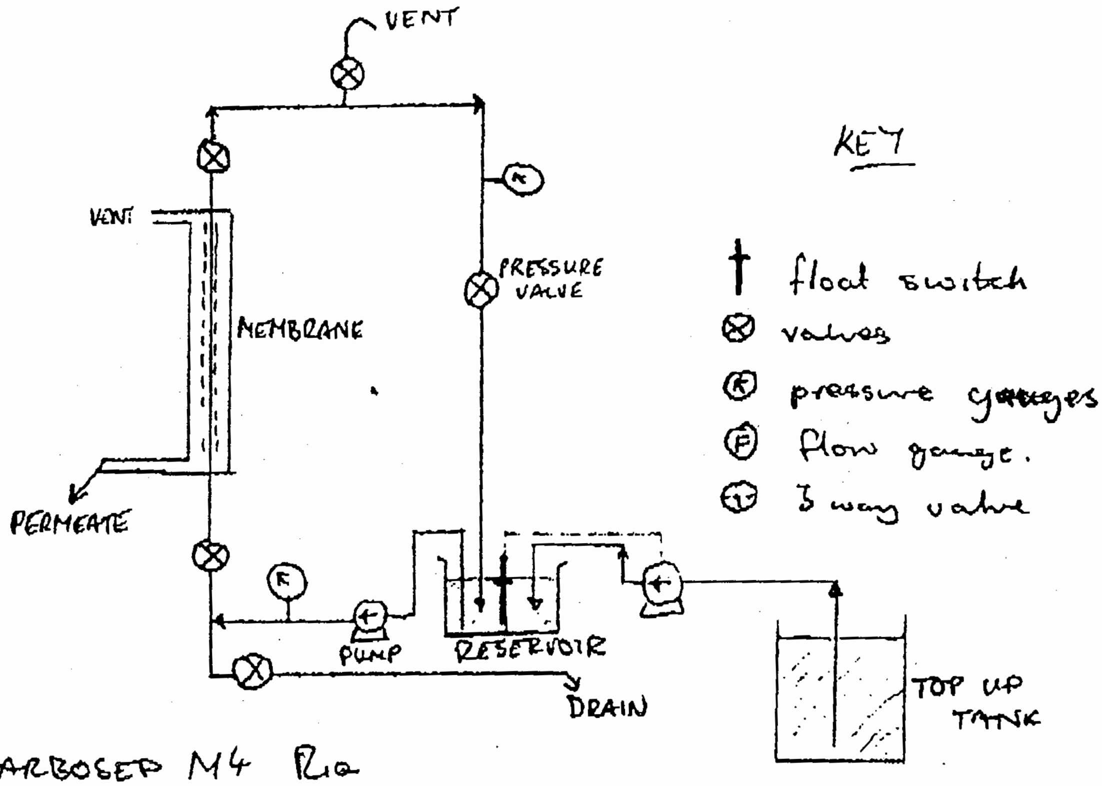
(i) Oxford has undertaken to develop a technique for purifying NaCl to 10^{13}, and if a 5% aqueous solution is used that becomes 5×10^{16}. Is it proposed to carry out the purification of NaCl in H_2O or D_2O? If the former then there will be a major concern about maintaining the purity during the drying operation.

(ii) It is likely that Th will be extracted by reducing the pH to 4 or 5 (see Lilley and Omori, April 1992) and assaying by α-counting and ICPMS–ETV, the latter being essentially independent of the chemical mysteries. In principle could the reduced pH technique be applied to the whole 1000 tons of D_2O? If not what action would be available to us if we suspect that thorium has deposited on the walls of the A.V. and the pipe-work?

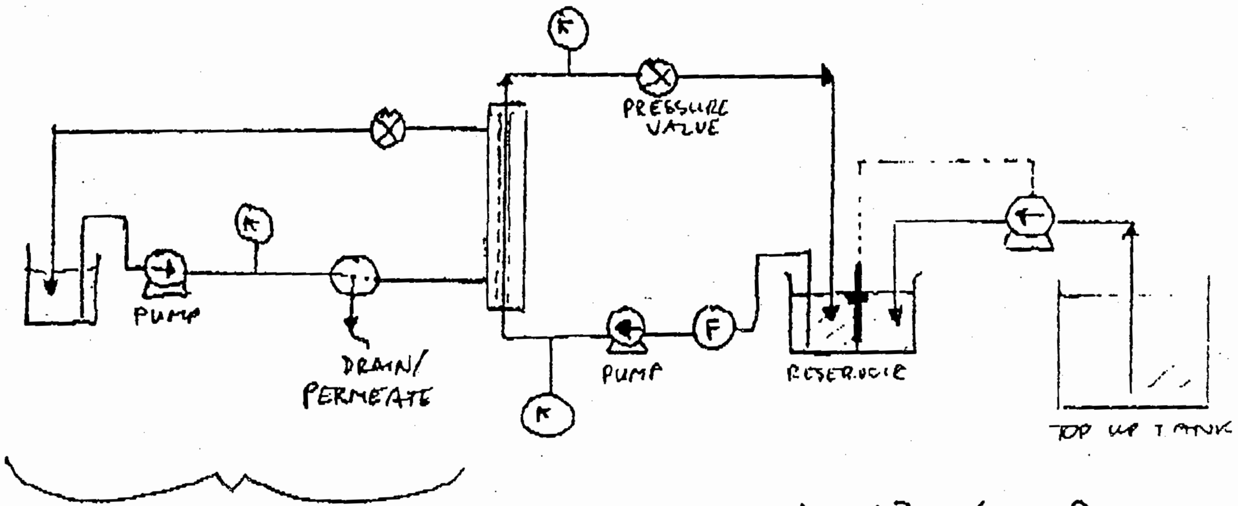
(iii) What is the time scale available for the assay development work, and what space and power are available for the assay equipment in the mine?

(iv) What is an acceptable level of redundancy for the assay measurements?

TANGENTIAL FILTRATION RIGS USED



CARBOSEP M4 RIG



AMICON H1P30-43 RIG