

Seeded Ultra Filtration for Th chain assay in the D₂OP. TRENT *et al*

Nov. 13/92

Introduction

The assay of the Th chain in the D₂O is critical to the success of SNO. It is therefore considered essential to have several independent methods for this assay. We present the case for using seeded ultrafiltration as part of an independent assay method (see Fig. 1) which is an alternative to packed bed adsorption. These two methods are compared and some of the difficulties associated with packed bed adsorption are discussed.

Packed Bed Adsorption

The details given in the minutes of the meeting of 28th July specify the primary radioactivity measurements as coming from the extraction of ²²⁴Ra in columns 320-AC01/02/03. These columns are then to be dried in situ ready for passing gas through them to extract ²²⁰Rn for measurement in the TPC or by other means. After passing through these columns, the D₂O flows through an ultra filtration system, ensuring the removal of any particulate matter.

If we assume a lower limit of 500 ²²⁴Ra atoms for the monitoring sample size, and accept the need to maintain the radioactivity level of the D₂O at one disintegration per day per tonne, then it becomes necessary for us to process samples of 100 tonne size in a period of about one day in order that we do not lose too much activity by decay before measurement. This would require a flow rate of about 100 litres per minute. The size of packed bed necessary in order to obtain a reasonably high extraction efficiency is then dictated by the residence time needed. From the work of Ferraris using MnO₂ spiked acrylic (Seakem) as an adsorber it appears that radium is (at high concentration) extracted with about 80% efficiency from a pH 7 solution after a residence time of 2 seconds, i.e. an adsorber bed volume of between three and four litres is needed for 100l/min flow. (In passing, it must be noted that at neutral pH thorium extraction by MnO₂ is not very efficient, and two seconds residence time would give an extraction efficiency of about 6% at best).

Some problems would be presented by the need for quick drying of a relatively large amount of the MnO₂/acrylic beads at the end of the sampling run, before gas

flow extraction can be used. Additionally, large columns of adsorber can give rise to difficulty in the extraction of emanated ^{220}Rn because of the risk of re-adsorption of the thoron.

Seeded Ultra Filtration

In comparison with the use of packed bed adsorption, a seeded ultra filtration unit in place of AC01/02/03 would appear to have considerable advantages. Based on preliminary data obtained at Oxford, the amount of seeding material used as adsorber in a primary (main) ultra filtration (U/F) loop would only amount to some 50g, assuming the membrane area necessary for the desired flow rate (100l/min) to be 50m^2 and using the established 95% efficiency level of $1\text{g}/\text{m}^2$ with hydrous titanium oxide (HTiO) as adsorber.

The operational details of this primary U/F loop are discussed in Fig. 2. The output of this primary U/F loop (after steps 1-7 in Fig. 2) would be ^{224}Ra adsorbed onto a suspension of 200 mg of HTiO in a 20 litre solution of 0.1M NaCl in D_2O with a contamination of less than 2g of Hydrogen. This solution could be passed through a membrane filter (see Fig. 1) to extract the HTiO suspension and hence the ^{224}Ra (>99% efficiency). After drying, the membrane filter could be used as a ^{220}Rn emanating source, by passing gas in the same direction as during the filtration. The emanation efficiency of such a source has not been investigated yet. But, given the small mass (200 mg) of finely divided adsorber, one can anticipate an efficiency at least comparable with that of 3-4 kg of MnO_2 beads. The activity on this membrane filter could then be measured by the TPC, by an electrostatic deposition counter or by the cryopumping deposition method.

An alternative method for extracting and transforming into a countable form the ^{224}Ra from the 20 litre output solution of the primary U/F loop is being worked on at present in Oxford. In essence, this method consists of repetitive concentration of the extracted radium (see Fig. 1). After a secondary U/F stage and a centrifuge stage the ^{224}Ra is evaporated onto a planchette. It can then be counted by a conventional alpha counter. With reasonable care, it is possible to reduce the background in the alpha counting units at present in use at Oxford to less than 3 counts per day. With

efficiencies of greater than 90% for each of the U/F stages, and a 25% efficiency for the centrifugation, evaporation and counting (this figure is based on recent work by Robert Black) we obtain a figure of a little better than 20% for the overall assay efficiency, ignoring losses from decay. This includes the 40% geometrical efficiency of the alpha counter.

The time involved in the processing above, including the main U/F 17 hour flow time, can be kept down to less than one day, so that we could expect to record about 80 decays from the original sample of 500 ^{224}Ra atoms. The alpha energies from ^{224}Ra down to ^{208}Pb range from 5.7 MeV up to 8.8 MeV and can all be distinguished. The numbers in the ^{212}Po to ^{208}Pb branch should be approximately double those in the branch from ^{213}Bi to ^{208}Tl and this would provide an additional check.

Similar arguments on efficiency can be adduced for the alternative ^{220}Rn emanation methods, by assuming a greater than 90% efficiency for the primary U/F loop. The principal advantage of the seeded ultra-filtration extraction method lies in the efficiency of small amounts of HTiO in adsorbing radium. Further measurements on this process, with and without salt present in solution are in progress at Oxford.

Conclusion

Given the critical nature of the neutral current background measurement it is considered essential to employ several independent and complementary methods to assay the Th chain in the D_2O . Also given the time pressures involved in the production of a final design for the D_2O purification plant, it is clearly not possible to pursue exhaustive investigation into the ideal form of this plant. On the basis of the work so far carried out on ultra filtration, we would strongly recommend the use of such a plant in the D_2O cleanup and assay.

Initially, it would appear that the requirements of the experiment could be met by the use of the Amicon P100 membrane. This is a polysulphone membrane and the largest module has 2.4m^2 area, so that 22 modules would be needed. The size of the whole unit, including some control gear would be about 2m high by 2m^2 . The cost is about £35K. Also two pumps are required, each delivering 100l/min, one at 1 bar, the other operating at 1 bar to deliver at 2 bar. The total power consumption is estimated

at 3 kW.

Before any final decision can be made, it would be desirable to check the thorium and uranium content of the membranes. Oxford has obtained a large sample from Amicon and measurements are in progress. It would also be desirable to check the level of D to H exchange that may take place in the membrane, although it is noted that the total mass of the polysulphone membrane is about 20 kg.

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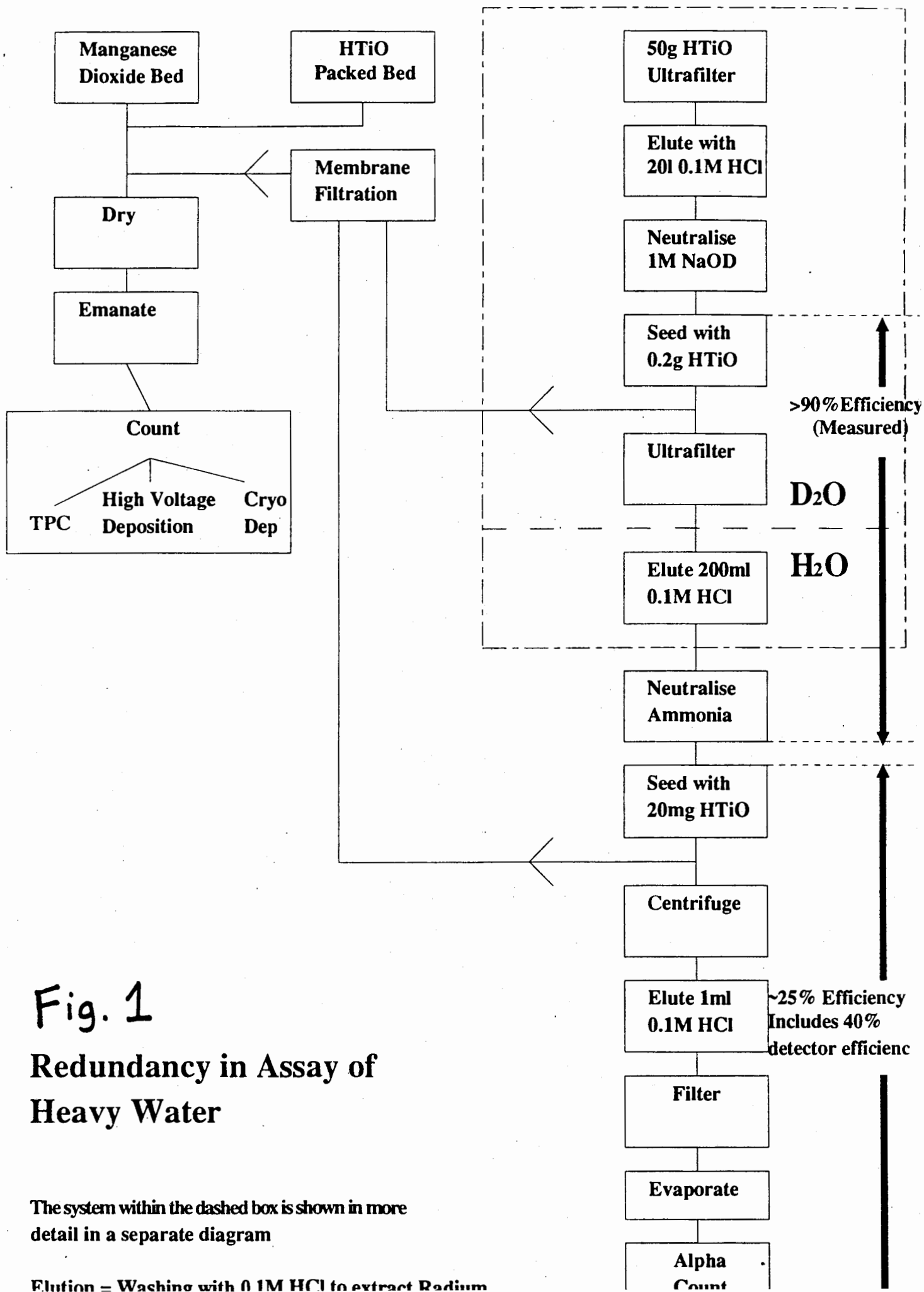
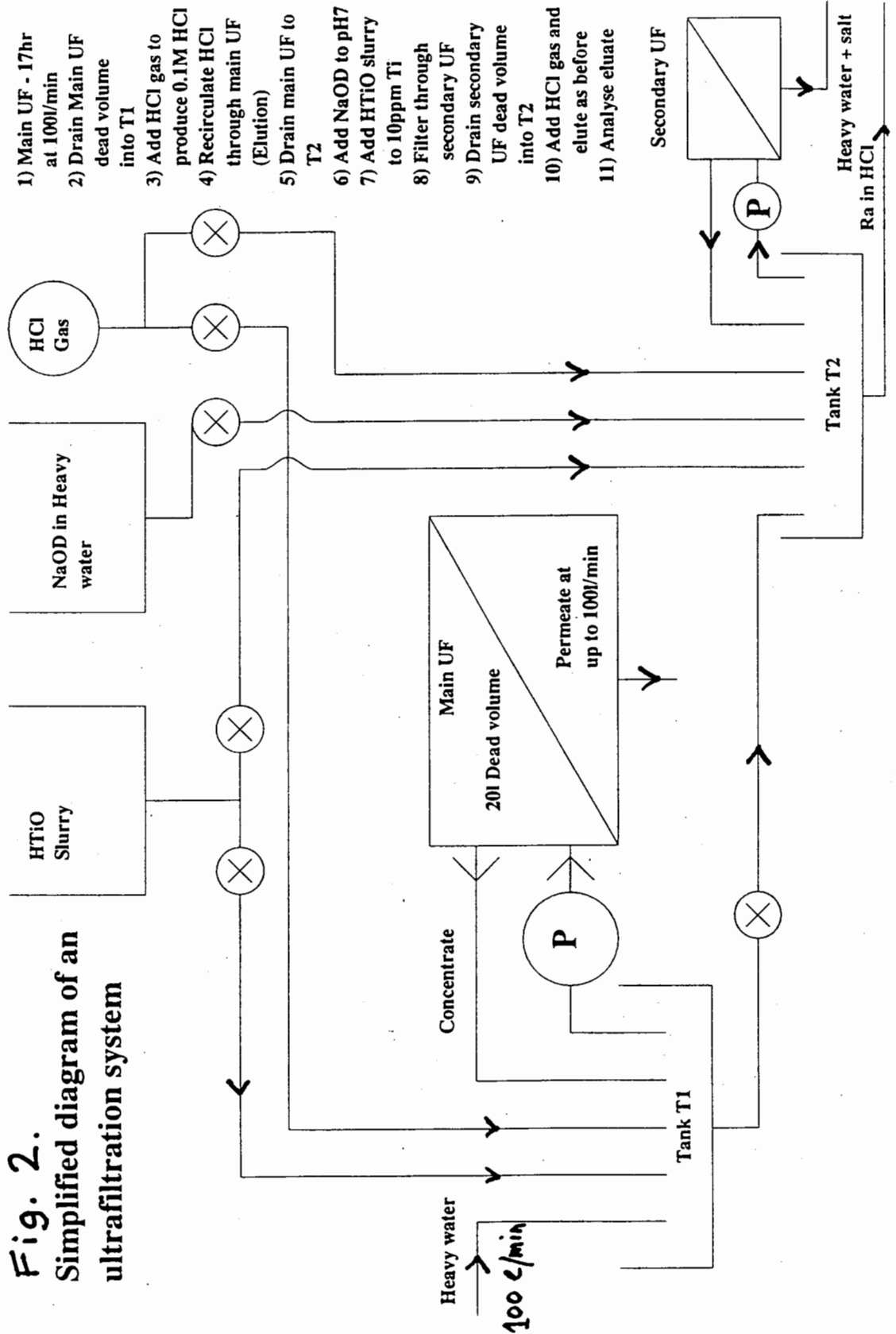


Fig. 2.
Simplified diagram of an
ultrafiltration system



- 1) Main UF - 17hr at 100l/min
- 2) Drain Main UF dead volume into T1
- 3) Add HCl gas to produce 0.1M HCl
- 4) Recirculate HCl through main UF (Elution)
- 5) Drain main UF to T2
- 6) Add NaOD to pH7
- 7) Add HTiO slurry to 10ppm Ti
- 8) Filter through secondary UF
- 9) Drain secondary UF dead volume into T2
- 10) Add HCl gas and elute as before
- 11) Analyse eluate

Secondary UF

Heavy water + salt

Ra in HCl