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Deposition Rates of Uranium and Thorium by Neutron Activation Analysis with witness plates

A Preliminary report of measurements
for the period July 27 - August 27, 1995

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Introduction

Estimates of acceptable deposition rates of mine dust on the acrylic vessel have been reported in various documents[1-3]. The amount usually quoted for the deposition rate of mine dust is 220 $\mu g/m^2$/month (0.4 $\mu g/cm^2$/18 months). The nominal values for Th and U in mine dust are 5 ppm and 2 ppm respectively.

Mine dust is estimated by measuring Fe in the fall-out deposited on 2.54cm diameter witness plates by the XRF technique. A standard ratio of Th ppm to Fe% in the mine dust is used as a conversion factor to report equivalent Th deposition rates. A factor of 3 is also assumed between Th and U. Even though XRF measurements are simple and fast, and hence very cost effective for routine monitoring of Th and U for SNO purposes, a confirmation by direct determination of Th and U is needed occassionally.

Thorium and uranium deposition rates may be determined directly by employing neutron activation analysis (NAA) of suitable witness plates. In this method the witness plate samples are first irradiated with neutrons in a nuclear reactor together with aluminum foil standards for a suitable length of time and the γ rays from ^{233}Pa (312 keV) and ^{239}Np (106 keV) are counted on high resolution HPGe detectors to detect thorium and uranium respectively. The counts detected at the γ-ray peaks of interest with the samples and the standards are used to calculate the thorium and uranium values directly.

Results are reported below for the witness plates deployed on location in the SNO underground laboratory during a one month period in July - August 1995.

Experimental details

Preliminary n-irradiation of samples of the polyethylene material used for covering the PSUP in the SNO cavity and some food wrap film indicated that the food wrap film has no thorium or uranium at the detection limit of 0.04 ng of thorium or uranium. The food wrap film is also found to give readily a more favourable mass ratio for the fall-out collecting on the film to the mass of the film used as a witness plate. Therefore, witness plates of dimensions 20 cm x 24 cm (usable area) of this controlled food wrap film stretched across cardboard frames were deployed at three locations in the SNO underground laboratory in Sudbury, and one location in the laboratory at University of Guelph. The film is stretched across the frame so as to expose only one surface to the fall out at any location and the collection area was horizontal.

After a one month exposure period, the usable area of each witness plate was cut out with a metal knife from the frame on location, and folded to a 3 cm x 3 cm size on location and wrapped in an outer film for irradiation purposes. Vinyl gloves were used for handling the samples during all operations. A 3 cm x 3 cm Al foil standard was added to each sample at Guelph before submitting the samples for irradiation.

The samples were irradiated for 30 minutes at a neutron flux of 1.5E13 n/cm^2/s in the McMaster Nuclear reactor. Counting started after a delay of 5 days to determine uranium from the ^{239}Np γ-ray at 106 keV. A second count was taken after a delay of 12 days to determine thorium from the ^{233}Pa γ-ray at 312 keV. Samples were prepared for counting after irradiation by separating the standard aluminum foil from each sample and removing the outer wrap from the sample itself. The witness plate samples and the aluminum foil standards were separately encapsulated again in an outer film to contain the radioactivity from the irradiated samples.

The germanium detector fully resolved the energies at 103.2 keV and 106.2 keV from ^{153}Sm and ^{239}Np activities respectively. There were no interfering γ-ray energies at 312 keV.

Results and discussion

The counting sensitivity was an average of 105 counts / h /ng U at 106 keV, and 7 counts / h / ng Th at 312 keV.

Table 1 summarizes the results at the various locations.It can be seen fromn the table that the uranium and thorium deposition rates are comparable in the control room and Guelph locations.

Table 1. Deposition rates of uranium and thorium during July-August 1995. Errors quoted below are 2σ errors on counting statistics alone.

Location	uranium ng/month/m^2	thorium ng/month/m^2
top of PSUP	42 +-15	
Deck	4.2+-12	
CTRL Room	19+-10	94+-25
Guelph	46+-35	45+-10

These deposition rates may be converted to dust loads for comparison with the desirable dust specification of 0.22mg/m^2/month (0.4 μg/cm^2/18 months). The control room Th/U ratio is consistent with the ratio in mine dust within experimental errors, and indicates a dust deposition rate of about 15 mg/m^2/month.

No large particulate matter such as paint chips of various colours and other debris was observed on the witness plates. As such the measured Th and U values by NAA are attributed to fine mine dust in the air falling on the witness plate in the control room during the one month period under consideration.

The U value on the PSUP indicates a factor of two higher deposition rate and the Deck seems to have a factor of five lower deposition rate than the control room. Thorium values are not available at this time. While an inspection of the top surface of the PSUP identified fall out of various types visible to the naked eye (paint chips of various colours, shining particulate matter when a light was shone on the surface, white dandruflike material with black cloth swipes, and brown dirt with white cloth swipes), the NAA values of Th and U indicate that this deposit is primarily related to ambient mine dust falling from the air which is of concern to us.

The measured deposition rates are approximately one hundred times higher than the desirable number. The following discussion addresses some of the concerns that arise from this higher deposition rate than the desirable.

If we assume that the fine dust is the ambient dust level achievable with the current level of construction activity with the air handling system in the laboratory the immediate concern is the amount of dust depositing on the acrylic vessel (AV) panels being assembled at the moment in the cavity. The total Th in the bulk of 30 tonnes of acrylic is 30 μg at a nominal value of 1 pg/g Th. If we assume that the upper half of the AV is the collection surface for the mine dust in the ambient air in the cavity then we get a Th deposition rate of 14 μg/month using the control room depostion rate, and an even higher rate judging from the U deposition rate on top of the PSUP. On the other hand, the deposition rate will only be about ten times higher than the acceptable rate if the Deck value for uranium is considered.

A concern is that contamination by Th and U of this type is difficult to take off from the surface of the AV panels. This concern is based on measurements of Th in bulk acrylic[4] made by NAA which showed that the bulk acrylic is probably very clean to better than 1 pg/g Th, but incidental surface contamination may be of the order of 45 pg/g, and the surface contamination can only be eliminated by machining the surfaces in a clean room environment and not by washing or etching with acids or ultrasonic cleaning methods.

These results are still very preliminary because this is the first attempt to use NAA for determining dust deposition rates in the SNO cavity. Although Th and U deposition rates are not yet measured near the acrylic panels now being assembled in the cavity, if the depositon rate were 100 ng/m^2/month, as it was on top of the PSUP, during an assembly period of five months the inside lower half of the AV will collect about 100 μg of Th. Therefore, 99% of this must be cleaned off to get to the level of 1 μg which is the amount of Th in the D_2O at 10^{-15} g/g.

Another concern is the dust falling on the joints between the panels during the bonding period. 3 μg of Th is estimated to accumulate in the bond joints exposed for a one month period at the above rate. This is 1/10th of the AV Th at 1 ppt. This Th will get locked into the bond joints without additional effort to control the dust deposition rate.

The results reported by Earle[5] for bond joints made at the 4600 level test set up may be of interest in considering what needs to be done. The interpretation of these results should be reexamined to identify if the results relate to the bulk of the

acrylic material near the bond joint or to a very small amount (like 10 g of acrylic) from the test bond.

Further work is planned to measure the Th and U deposition rates during the assembly of the acrylic vessel in the cavity.

References

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3. R. Stokstad, SNO-STR-92-023.
4. P.Jagam, J.X.Wang, and J.J. Simpson, J. Radioanl. Nucl. Chem., 171 (1993) 277.
5. E.D.Earle, SNO-STR-