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Title:

Determination of *In Vitro* Lung Solubility and Intake-to-Dose Conversion Factors for Tritiated $LaNi_{4.15}Al_{0.85}$ (LANA.85) and 13X Zeolite

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Determination of *In Vitro* Lung Solubility and Intake-to-Dose Conversion Factors for Tritiated $LaNi_{4.15}Al_{0.85}$ and 13X Zeolite

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Samples of tritiated $LaNi_{4.15}Al_{0.85}$ (LANA.85) and 13X zeolite were analyzed to obtain particle size distributions and tritium evolution rates in a simulated lung environment. This information was used to calculate intake-to-dose conversion factors (DCFs), which estimate the committed effective dose (CED) a worker would receive after inhaling either tritiated particulate. The DCFs for tritiated LANA.85 and 13X particulate with a default activity mean aerodynamic diameter (AMAD) of 5 μm were determined to be 1.01E-11 Sv/Bq and 1.11E-11 Sv/Bq, respectively. These results are comparable to that of HTO, 1.8E-11 Sv/Bq, indicating that urine bioassay results can conservatively estimate the dose delivered if the worker was exposed to any mixture of HTO, LANA.85, or 13X.

Keywords:

Tritium

LANA.85

13X Zeolite

Dose Conversion Factor

I. INTRODUCTION

Special Tritium Compounds (STCs) are defined by DOE-STD-1129-2015 *Tritium Handling and Safe Storage* as: "Any compound, except for H_2O, that contains tritium, either intentionally (e.g., by synthesis) or inadvertently (e.g., by contamination mechanisms)."[1] The Savannah River Tritium Enterprise (SRTE) relies on many different materials to process tritium, any of which can be considered to be a STC. These STCs can retain various amounts of tritium in particulate form and present special radiological protection challenges different than those of elemental tritium or tritiated water. These challenges can be divided into three areas: detection, physical and chemical behavior, and radiation emission, any of which can complicate deconvolution of bioassay results.

Tritium evolution from inhaled particulate occurs through reaction or dissolution of the host material or by diffusion of the tritium from the intact host material. In this discussion the release of tritium by any mechanism is referred to as "dissolution." STCs that readily release tritium are classified as "soluble" and result in lower DCFs than those that are "insoluble". Once released from a soluble material, the tritium is assimilated as either tritiated water or organically bound tritium (OBT). Tritiated water and OBT are flushed from the body via normal metabolic processes with biological half-lives of 10 and 40 days, respectively.[1] Materials classified as insoluble allow tritium beta decay energy to deliver an overall higher dose to the area surrounding the STC.

II. EXPERIMENTAL

Tritiated samples of LANA.85 and 13X zeolite were prepared by the SRTE and sent to the Lovelace Respiratory Research Institute (LRRI). A portion of each powder sample was suspended in ethanol. Two drops of each suspension was placed on a slide and images were taken with an optical microscope. These images were analyzed for particle size using Image Pro Plus software. SigmaPlot software was used to fit each distribution to a lognormal equation.

The in-vitro dissolution apparatus was designed to measure the tritium released to Serum Ultrafiltrate[2] (SUF, a simulated lung fluid) and the reactor headspace. Figure 1 shows a schematic of the dissolution apparatus which consists of a water bath, dissolution flask, propylene glycol bubblers, catalyst column to oxidize any elemental tritium, carbon filter, gas supplies, and temperature controller. Testing of each material was performed individually by sandwiching approximately 5 mg of material between two 47 mm membrane filters and securing the material and filters in a Teflon filter holder. The filter holder was placed into a 500-mL glass flask containing 100 mL of SUF and incubated in a 37°C water bath. Background system activity was measured with all solutions present before introduction of the tritiated sample and after testing was complete.

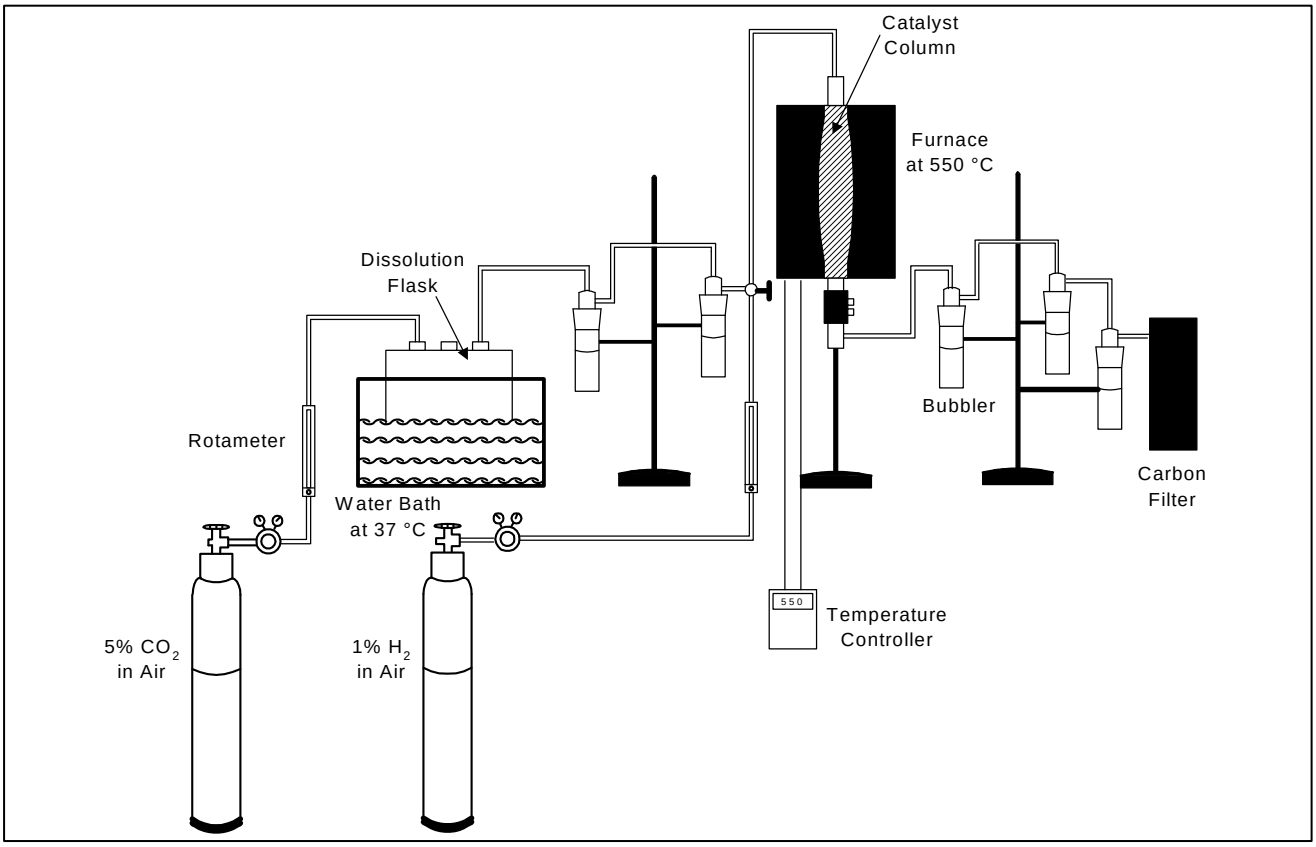

Fig. 1. Diagram of the Dissolution Testing System.

Flask and glycol solutions were changed at 1, 6, 24, 48, and 72 hours, then twice weekly for three months. One hour before each solution change, air with 5% CO_2 was passed through the flask head space. Tritiated water was trapped in the first series of glycol bubblers. Any elemental tritium was converted to oxide in the heated catalyst column and trapped in the second series of bubblers. Before introducing new solutions, the kettle and glycol traps were wiped out with paper towels. The dissolution rate in terms of percent activity per day was calculated by dividing the tritium released by the time and total activity. After three months of testing, the remaining solid samples were digested to quantify the amount of tritium remaining in the solid. LANA.85 was digested with forty mL of aqua regia; 13X was digested with forty mL of 25% hydrofluoric acid.

The total activity of each sample was calculated by summing the activity collected in the dissolution flask, the bubblers, and the digest solution. The percentage of tritium released from the sample was plotted as a function of time and fit using nonlinear least-squares estimates[3,4] procedure implemented in the Statistical Software R version 2.14.0. (Ref. 5)

III. RESULTS AND DISCUSSION

Optical analysis of hundreds of particles was used to fit the particle size distribution for each material to the bimodal lognormal equation below:

$$f(d) = \frac{a}{\sqrt{2\pi}\, d \ln \sigma_{g1}} \exp\left[- \frac{(\ln d - \ln d_{o1})^2}{2 (\ln \sigma_{g1})^2} \right] + \frac{1-a}{\sqrt{2\pi}\, d \ln \sigma_{g2}} \exp\left[- \frac{(\ln d - \ln d_{o2})^2}{2 (\ln \sigma_{g2})^2} \right] \qquad (1)$$

where a is the fraction of smaller particles d_{o1} is the count median diameter of the smaller particles (where 50% of particle counts exceed the value), and σ_{g1} is the geometric standard deviation of the smaller particle distribution. Likewise, d_{o2} is the count median diameter of the larger particles and σ_{g2} is the geometric standard deviation of the larger particle distribution. Because 13X measurements fell within a single distribution curve ($a=1$), the second term drops out. Using these distributions and the physical properties of each material, activity mean aerodynamic diameters (AMADs) and geometric standard deviations (GSDs) were calculated. Results are given in Table 1.

TABLE I. Particle Analysis Results.

	LANA.85	14 X Zeolite
a	0.76	1
$AMAD_1$ (μm)	1.85	3.71
GSD_1	2.7	2.10
$AMAD_2$ (μm)	23.92	-
GSD_2	1.6	-
Total Tritium Content (Bq/g)	6.58E8	4.44E8

Figure 2 shows in-vitro tritium retention data for LANA.85 and 13X zeolite collected during the 90 day test. ICRP 66 (1994) Type F and M materials are shown for comparison. Data for each test material was fitted to Equation 2 to model tritium retention in the solid *A(t)* at any time. In Equation 2, *b* is the fraction of tritium that quickly dissolves and c_1 and c_2 are time constants for the quickly and slowly dissolving portions, respectively. Fit results for each material are given in Table 2 and are displayed graphically in Figure 2.

$$A(t) = b\exp(-c_1 t) + (1-b)\exp(-c_2 t) \tag{2}$$

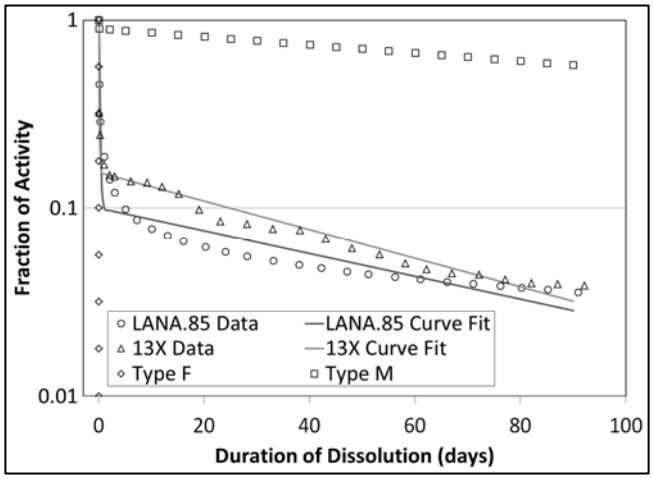

Fig. 2. Tritium Retention in Samples Tested.

TABLE II. Tritium Retention Equation Fit Results.

	LANA.85	13X Zeolite
b	0.900	0.845
c_1 (μm)	6.816	39.2
c_2 (μm)	0.014	0.0176

LANA.85 and 13X zeolite show rapid dissolution fractions of 0.900 and 0.845, indicating "soluble" materials. According to ICRP 66 (Refs. 6, 7), type F materials have a rapidly dissolved fraction of 1. For LANA.85, dissolution half-times for the fast and slow portions of the retention curve are 0.1 and 49.5 days, respectively. For 13X, the values are 0.018 and 39.4 days. Dissolution behavior of both materials is clearly between that of Type F and M materials.

IV. INTAKE TO DOSE CONVERSION FACTOR DETERMINATION

Measured AMAD values were used in conjunction with observed tritium retention functions *A(t)* to calculate the DCF for each STC using Integrated Modules for Bioassay Analysis (IMBA) Professional Plus V4.1.18 (Refs. 8, 9). DCFs calculated with IMBA are essentially the same as those calculated in Tritium Handling and Safe Storage.[1] These results can be tailored to estimate DCFs for other particulate sizes of the same material. DCFs calculated with IMBA do not account for self-absorption of the weak beta radiation by the host material, meaning that calculated committed effective doses for a given uptake are artificially high. Test results are presented in Table 3 as a function of aerosol AMADs with default materials for comparison.

The DCF for the bimodal LANA.85 ("LANA.85 comp" in Table 3) particulate was determined by calculating the DCF for the 1.85 μm component and the 23.92 μm component separately and then adding them together by the observed proportions. It should be noted that particles with AMADs greater than 10 μm are generally considered to be non-respirable. Inclusion of the 23.92 μm component provides additional conservatism because it is unlikely that a significant portion of these particles will reach the deep lung.

Table III. Intake-to-Dose Conversion Factors (Sv/Bq) for Various Tritiated Materials.

Tritiated Material	Fraction of Tritium Released	DCF (Sv/Bq)	Ratio to HTO
Type S 5.0μ	0.01	1.30E-10	7.10
Type M 5.0μ	0.1	2.81E-11	1.54
Type F 5.0μ	1	9.03E-12	0.49
LANA.85 5.0μ	0.9	1.01E-11	0.55
LANA.85 1.85μ	1	9.91E-12	0.54
LANA.85 23.92μ	1	5.97E-12	0.33
LANA.85 comp	1	9.00E-12	0.49
13X Zeolite 5.0μ	0.9	1.01E-11	0.55
13X Zeolite 3.71μ	1	1.11E-11	0.61
HTO	-----	1.83E-11	1.00

For routine applications in occupational radiation protection a DCF of 1.01E-11 Sv/Bq is recommended for both tritiated LANA.85 and 13X zeolite. These DCFs are approximately 55% that of HTO because only part of the inhaled particulate is transferred to the systemic organs and tissues. Conversely, all inhaled HTO is assumed to be assimilated because it is a vapor.

The DCF for a radioactive material gives the committed effective dose per unit intake. In practice, intakes of tritiated radioactive materials are usually not measured directly but are instead inferred from urine bioassay measurements. For this reason it is useful to examine the dose implied by a unit quantity of tritium in the urine rather than a unit intake. Further, it is often useful to express this dose per unit activity in the urine relative to the dose per unit activity in the urine following an intake of HTO. This *dose index* (DI) gives an idea of the magnitude of the error associated with the incorrect assumption that all tritium in the urine came from intakes of HTO when in reality the intake was tritiated particulate.

For example, assume that an acute inhalation intake of 5 μm AMAD Type F STC produces a urine bioassay result $M(t)$ at t days after the intake. Given the appropriate intake retention fraction (IRF) $m_{STC}(t)$ and DCF_{STC}, the committed effective dose (CED) is:

$$CED = DCF_{STC}\left(\frac{M(t)}{m_{STC}(t)}\right). \tag{3}$$

The DI or ratio of the dose due to an uptake of a particular tritiated STC to the dose that would be calculated assuming the tritium in the urine resulted from an intake of tritiated water is then:

$$DI(t) = DCF_{STC}\left(\frac{M(t)}{m_{STC}(t)}\right) \Big/ DCF_{HTO}\left(\frac{M(t)}{m_{HTO}(t)}\right) = \frac{DCF_{STC}[m_{HTO}(t)]}{DCF_{HTO}[m_{STC}(t)]}. \tag{4}$$

A dose index greater than 1 indicates that the dose from an intake of tritiated material will be underestimated if one assumes that the bioassay sample was a result of an intake of HTO. Dose indices for LANA.85 and 13X zeolite are shown in Figure 3 with standard Type F, M, and S tritiated materials

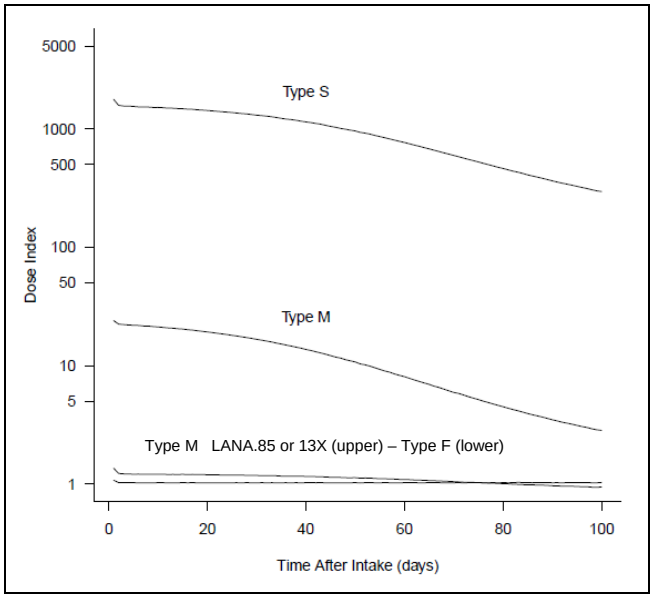

Fig. 3. Dose Indices Compared to Standard Type F, M, and S Tritiated Materials.

for reference. The plots show that the dose resulting from intakes of LANA.85, 13X, or Type F materials can be reasonably estimated if one assumes that the inhaled material was tritiated water. This provides justification for using the DCF of 1.01E-11Sv/Bq for both LANA.85 and 13X. In contrast, the plots show that if the positive bioassay was due to an intake of Type M or Type S STCs, the calculated CED would be significantly underestimated if the assumption was made that the intake was HTO. The practical significance of this is that an occupational internal dosimetrist can assume that all tritium in a urine

bioassay came from intakes of HTO if the only potential sources were HTO, LANA.85, or 13X. This greatly simplifies design of monitoring programs and the interpretation of bioassay data.

IV. Conclusions

The results of this study show rapid dissolution of both LANA.85 and 13X with most of the total tritium released to the simulated lung fluid within the first few days and over 95% dissolved during the 90 day test. After dissolution testing was complete, no activity above background was found to remain on the sample holder. Among the tested tritiated materials listed in Ref. 1, both LANA.85 and 13X have faster dissolution rates than titanium, zirconium and hafnium[10,11], but a slower dissolution rate than LANA.75 (Ref. 12). Dissolution test results and a default AMAD of 5 μm were used to calculate a recommended DCF of 1.01E-11 Sv/Bq for both tritiated LANA.85 and 13X. Of greater practical significance is that the data demonstrate that tritium in a urine bioassay can be reasonably interpreted as if it had resulted from an intake of HTO if the worker was potentially exposed to any mixture of tritiated water, LANA.85, or 13X.

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