Savannah River National Laboratory™

OPERATED BY SAVANNAH RIVER NUCLEAR SOLUTIONS

We put science to work.™

Determination of In-Vitro Lung Solubility and Intake-To-Dose Conversion Factors for Tritiated $LaNi_{4.25}Al_{0.75}$ Hydride

Greg Staack

34th Tritium Focus Group
September 23-25, 2014

SRNL-STI-2014-00424

Savannah River NUCLEAR SOLUTIONS™
FLUOR • NEWPORT NEWS NUCLEAR • HONEYWELL

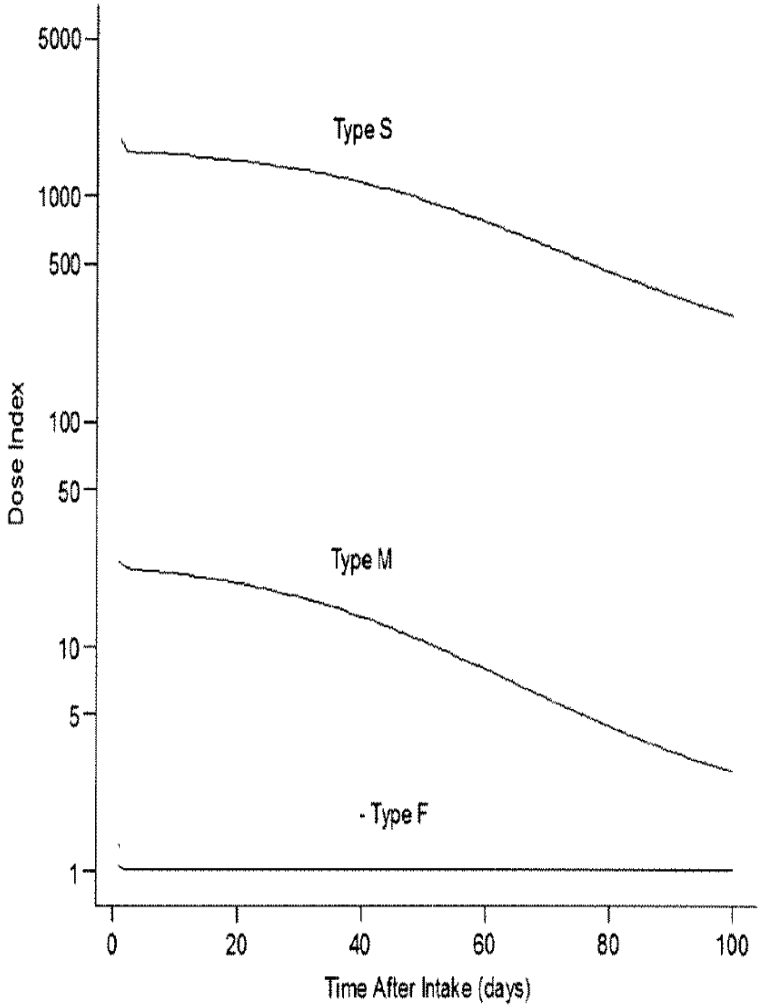
What are *Special* Tritium Compounds (STCs) and why are they *Special*?

ANY compound (except for H_2O) that contains tritium, either intentionally or inadvertently – Tritium Handling and Safe Storage, DOE-HDBK-1129-2008

- Organically bound tritium
- Metal tritides
- Adopt physical and chemical characteristics of host material
  - Particle size
  - Biological behavior
- Present special challenges
  - Detection
  - Characterization

How are STCs Classified?

- Soluble or Insoluble depending on how quickly the tritium is transferred to the bloodstream
- International Commission on Radiological Protection (ICRP) categorizes inhaled radionuclides on the rate of absorption from the respiratory tract to the bloodstream.
  - *Fast (Soluble) – days*
  - *Moderate (Insoluble) – weeks*
  - *Slow (Insoluble) – years*
- Uncharacterized materials considered to behave as HfT
- May lead to overly conservative dose estimates



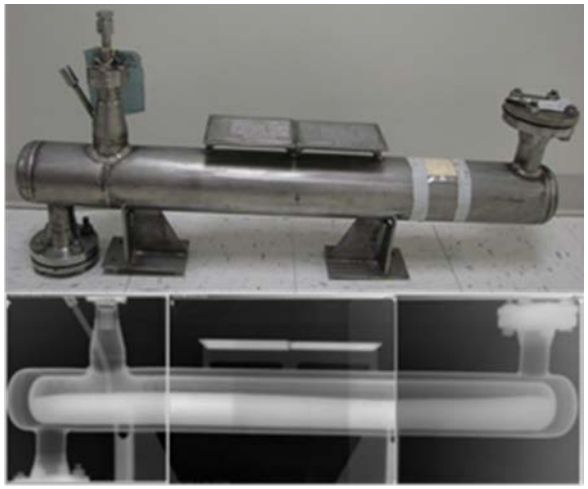
What is a Dose Conversion Factor (DCF)?

- Estimate of the dose an individual would receive in a given situation
- The International Commission on Radiological Protection (ICRP) Publication 66 Human Respiratory Tract Model provides an approach to calculate the DCF for inhaled materials.
  - Particle size
  - Dissolution rate (dissolution of metal matrix OR diffusion of T from solid)

STC/Isotope	Ratio of DCF for selected STC to DCF for selected radioactive isotope or compound
HfT/HTO	24
OBT/HTO	2
HfT/^{137}Cs	1/20
HfT/^{235}U	1/77,000
HfT/^{239}Pu	1/270,000

DCF Determination for Tritiated LANA.75

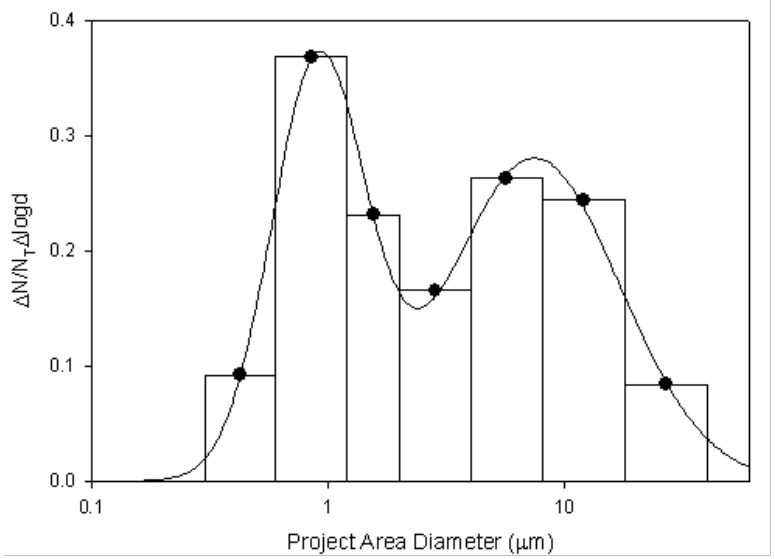
- $LaNi_{4.25}Al_{0.75}$ or LANA.75 has been used by the Savannah River Site Tritium Facilities for years to safely store hydrogen isotopes
- Hydrides pose special risks due to tritium storage capacity and decrepitation
- Testing was performed on tritiated LANA.75 to determine whether current Radiological Control practices are sufficient



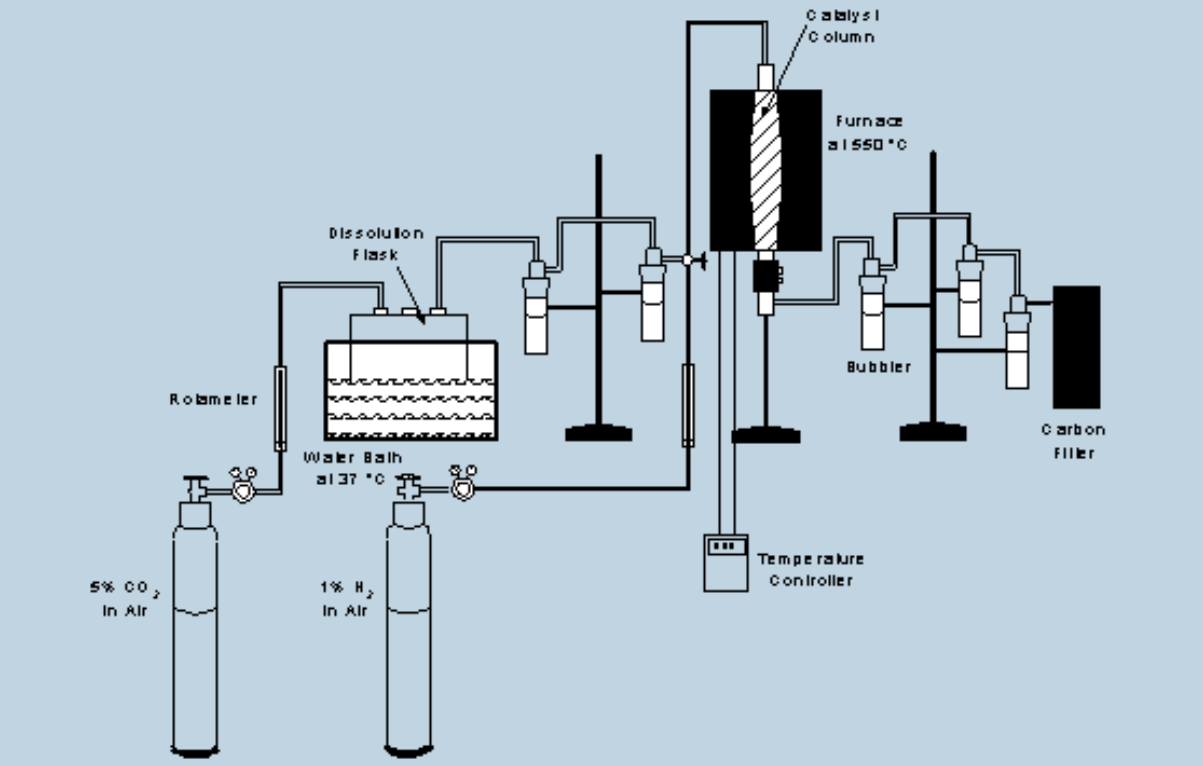
Particle size distribution

- Portion of the sample is suspended in 0.5 ml ethanol
- Images taken with an optical microscope
- >300 particles analyzed using Image Pro Plus software to determine particle size distribution
- Particle size distribution was used to calculate the Activity Mean Aerodynamic Diameter (AMAD), Geometric Standard Deviation (GSD), and Fractional Split
- Distribution was fit using the bimodal lognormal equation:

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



Dissolution Testing



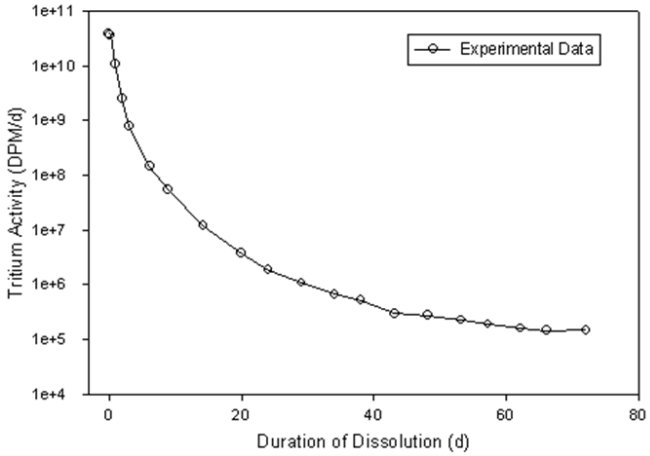
- Hydride sandwiched between two 0.2µm membrane filters
- Submerged in 100ml of Serum Ultra Filtrate (SUF) and incubated at 37°C
- Headspace purged and SUF changed periodically over 2.5 months to measure tritium release rate from the sample
  - Bubblers accounted for elemental and oxide tritium in the head space
- Remaining sample dissolved in aqua regia to determine total activity
- All solutions measured via liquid scintillation

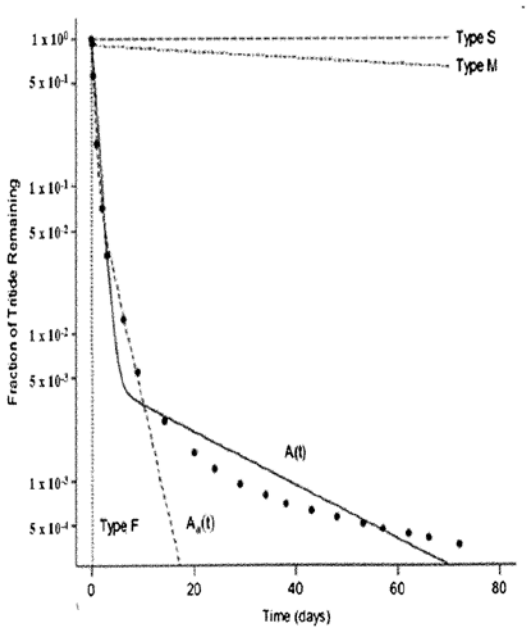
Dissolution Results

- Tritium recovered from the system (SUF + bubblers) was plotted versus time
- Filters and remaining sample were dissolved in aqua regia to determine the residual tritium in the solid
- System background was measured after removal of the tritide
- The fraction of activity remaining in the solid as a function of time was:

$$A(t) = 0.995\exp(-1.177t) + 0.005\exp(-0.042t)$$

- Tritiated LANA.75 dissolution behavior is between “Fast” and “Moderate”





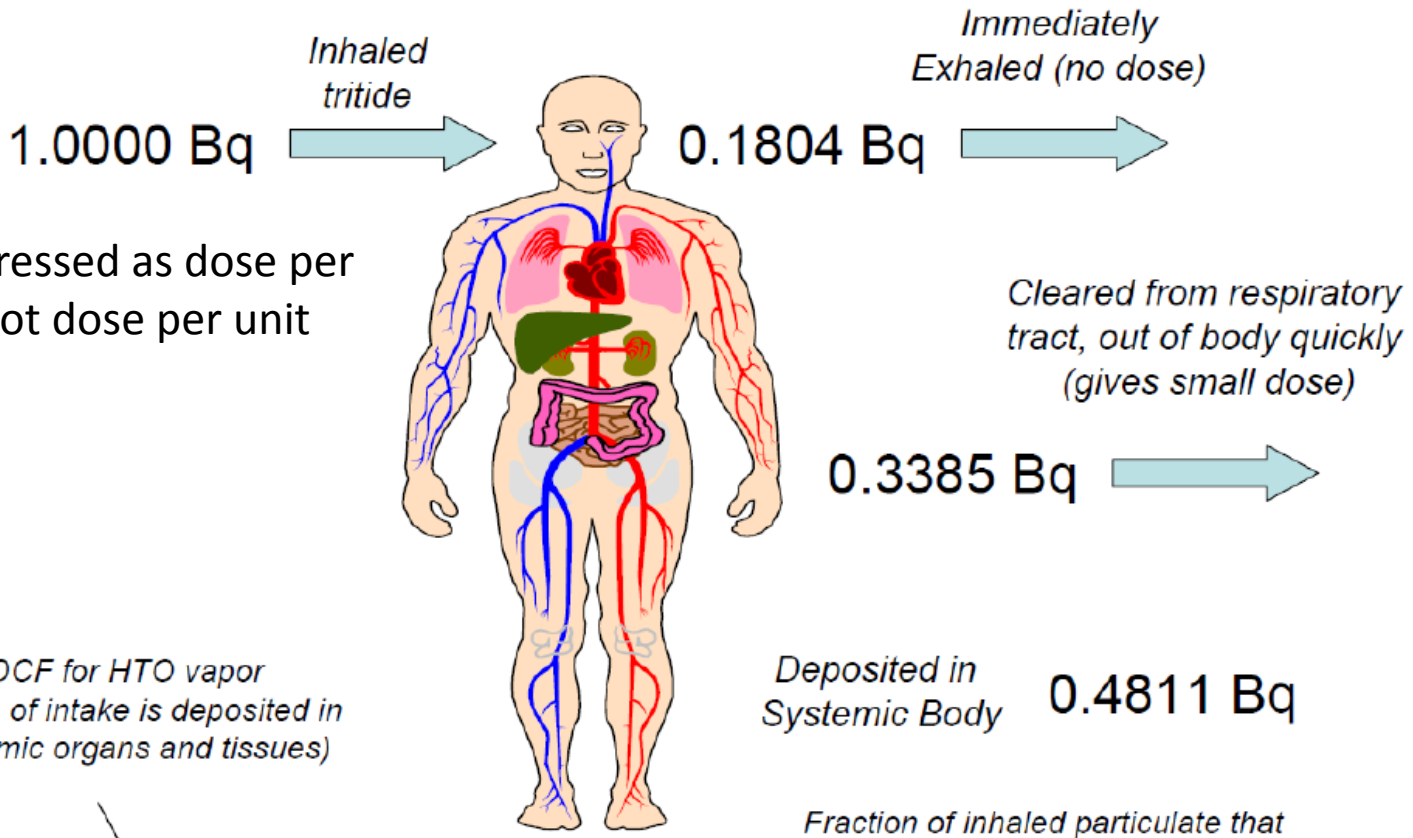
DCF Calculation

- Particle size analysis and dissolution data were used in conjunction with the ICRP model to calculate a defensible dose conversion factor for tritiated LANA.75
- Results were extrapolated to a default 5µ AMAD particle

Source	DCF (Sv/Bq)	Ratio
LANA.75 (5µ)	9.40×10^{-12}	0.51
Type F STC	9.03×10^{-12}	0.49
Type M STC	2.81×10^{-11}	1.54
Type S STC	1.30×10^{-10}	7.10
HTO	1.83×10^{-11}	1.00

Additional Information

DCFs are expressed as dose per unit intake, not dose per unit deposited.



Conclusions

- Tritiated LANA.75 quickly releases the majority of tritium in simulated lung fluid - 99.5% of the tritium “dissolved” with a half time of 0.6 day
- The calculated Dose to Conversion Factor for 5µ tritiated LANA.75 is 9.4×10^{-12} Sv/Bq
- Uptakes of tritiated LANA.75 would be detected by urine bioassay analysis at approximately the same limits as HTO
- Doses due to tritiated LANA.75 would not be significantly underestimated if the assumption is made that the uptake is due to HTO = No special monitoring equipment is necessary!

Acknowledgements

The authors of this report would like to thank the National Nuclear Security Administration (NNSA) Nuclear Safety Research and Development Working Group for its support of this project. The authors would also like to thank the many people in the SRS Tritium Facilities and SRNL who worked to safely prepare and deliver the tritided LANA.75 sample to LRRI.

Publication Information

E. B. FARFÁN, T. R. LABONE, G. C. STAACK, Y.-S. CHENG, Y. ZHOU, AND T. P. VARALLO, “Determination of In Vitro Lung Solubility and Intake-to-dose Conversion Factor for Tritiated Lanthanum Nickel Aluminum Alloy”, *Health Physics*, 103, 249 (2012).