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Recovery of a Tritiated LANA Sample For Dose Conversion Factor Determination

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1.0 Introduction

Special Tritium Compounds, or STCs are defined as any compound, (except for hydrogen in elemental or oxide form) that contains tritium. STCs can be solids, particulates, or liquids/vapors and are classified as Type F (fast), M (moderate), or S (slow) according to how quickly the tritium is transferred from the respiratory tract to the bloodstream. STCs pose a number of safety concerns: they are more difficult to detect because some portion of the beta decay will be blocked by the host matrix, they behave like the host material instead of tritium, possibly rendering bioassay and current dose models inaccurate, and it is difficult to differentiate between STCs and normal smearable tritium contamination. DOE-HDBK-1184-2004, Radiological Control Programs for Special Tritium Compounds states that:

"Soluble types of particulates rapidly release their bound tritium. For these types of tritiated particles, radiation protection measures should be based on those for HT or HTO. In addition, the detection problems that arise from the binding of tritium within the host particle are not an issue (see section 3.1). Examples of such particles are metal tritides of palladium and uranium."

It is important to emphasize that the label "soluble" is indicative of how quickly the tritium is released from the particulate and is not necessarily related to whether the particulate will dissolve in serum ultrafiltrate (SUF), a simulated lung fluid. Materials classified as Type M or S are considered "insoluble". DOE-HDBK-1184-2004 goes on to list DCFs for titanium, zirconium, and hafnium, all "insoluble" getter type hydrides.

All materials that have not been evaluated are treated as hafnium, which has the most conservative dose conversion factor (DCF) of materials tested to date. The purpose of this work is to develop a technical basis for both estimating the dose of a worker exposed to respirable tritiated $LaNi_{4.25}Al_{0.75}$ (LANA) and implementing hazard appropriate controls. Savannah River National Laboratory (SRNL) has agreed to provide Lovelace Respiratory Research Institute (LRRI) with a tritiated LANA sample. LRRI will determine the particle size distribution (PSD) as well as perform dissolution rate studies on the sample in SUF. The rate of tritium release from the sample will be measured over a three month period. Tritium release rate information will be used to calculate a DCF for respirable tritiated LANA.

Initial discussions on delivering a tritiated LANA sample to LRRI immediately recognized the fact that there are a number of variables that will likely impact how quickly tritium is released from the sample. These variables include: surface area of the sample, hydrogen isotopic ratios within the sample, and the equilibrium pressure of the sample (related to the amount of tritium aging). It was known that tritium aging results in the formation of a "heel", a decrease in the reversible capacity of the hydride. This is demonstrated by the shift between the "virgin" and "11-1/2 yr" isotherms in Figure 1. Particle size, which may be related to surface area, has been identified and compensated for in the dose conversion factor model.

The decision to use a deuterium exchanged legacy LANA sample for this study was based on two factors: an aged sample would retain more tritium for a longer period due to aging effects and would likely be more representative of material to which a worker might be exposed.

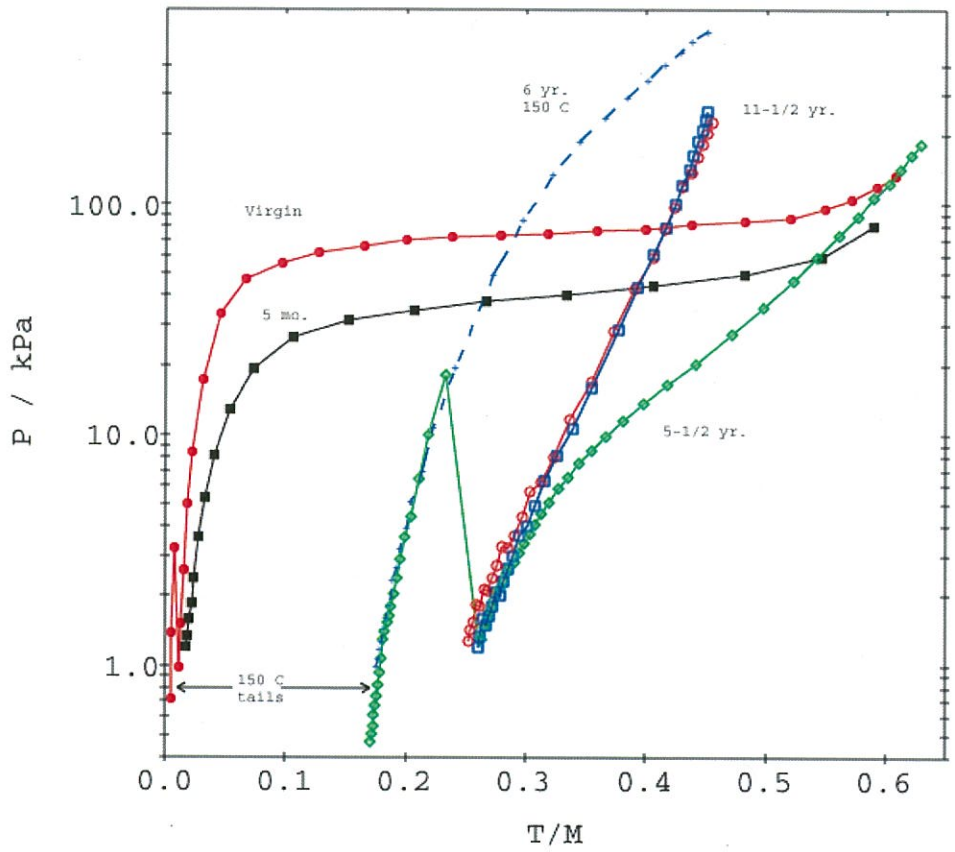

Figure 1. Tritiated LANA Isotherms

2.0 Sample Preparation

A series of isotopic dilutions were performed to reduce the total sample tritium inventory in a nominal 4.9 g sample to approximately 123 Ci in preparation for sample repackaging. Following inventory reduction, an attempt was made to "passivate" the LANA. Passivation is the formation of a thin oxide layer on the surface of the reduced metal powder, rendering it insensitive to air. Passivation is typically accomplished by adding air to the sample in a controlled manner and monitoring sample temperature. The addition of air with no accompanying temperature rise typically indicates that passivation is complete. Attempts to passivate the sample consistently resulted in a 0.1-0.3 °C temperature rise. A quick check with a TCVG indicated that the sample was evolving a hydrogen overpressure of approximately 0.1 torr. This overpressure was likely reacting with the oxygen in the presence of the metal surface to form water. Passivation activities continued until no temperature increase was observed. A ratio of initial to final pressure (before and after expanding the gas into the evacuated test cell) with an inert gas compared to the ratio with air indicated that a reaction was still occurring, but the amount of hydrogen available to react was too small to generate a measurable temperature increase. At that point the test cell was determined to be insensitive enough to air to safely open.

The sample was transferred to a glovebag inside of a glovebox and four different masses of LANA were safely recovered. The masses varied from about 35mg to about 100mg. The variety was deliberately chosen in an attempt to compensate for the unknown amount of tritium lost

during passivation. Primary containment for each recovered mass is a VCR8 male union. One of the four masses was sent to Analytical Development in SRNL for final tritium assay measurement prior to shipping another mass to LRRI. LRRI is only licensed for 4 Ci of tritium, so it was critical that the tritium content was verified. Results from Analytical Development indicated a final tritium concentration of 1.08 Ci/g LANA [1] which indicates that >95% of the tritium present after isotopic dilution was released during passivation activities.

3.0 Complications of Working with Offgassing Samples

Attempts to measure the tritium release rate on an unstable sample will likely not give results as reproducible as those obtained on a stable sample. While there is no such thing as a truly stable metal hydride, tritium release rates in SUF can vary widely depending on the solubility, reactivity, isotopic ratios, amount of tritium damage, surface area, and storage conditions of the sample. With more stable samples, either solubility or reactivity of the particulate in SUF will be the dominant tritium release mechanism and will likely be a function of the surface area of the sample. With less stable or offgassing samples, test results can be impacted by any of the variables listed above.

If offgassing is the dominant mechanism for releasing tritium, a particle with a lower equilibrium pressure will retain more tritium over a longer time period than a particle with a higher equilibrium pressure. Equilibrium pressures can be either functions of the material itself or the amount of tritium service the material has seen. Isotope composition can also impact tritium release rate experiments. A sample loaded to 0.1 Q/M with 10% tritium may generate a different release data set than a sample loaded to 0.01 Q/M with 100% tritium, even though the total tritium content is the same. Finally, treatment of the sample prior to testing can affect test results. An offgassing sample may release a substantial portion of its tritium inventory to the environment over a relatively short time frame. The amount lost will be functions of the equilibrium pressure of the material, gas flow around the sample, and time spent in that environment.

4.0 Discussion

The LANA sample was isotopically exchanged to a tritium inventory of approximately 123 Ci of tritium in about 4.9 g of sample. It was understood that passivation activities would further reduce tritium inventory by an unknown amount. Liquid scintillation of a dissolved portion of LANA harvested at the same time as the sample to be sent to LRRI gave a tritium inventory of 1.08 Ci/g LANA. LRRI had requested a sample containing between 10 mCi and 3.0 Ci of tritium per 50 mg of LANA. SRNL is sending 98.8 mg containing 107 mCi of tritium, equal to 54 mCi of tritium per 50 mg of LANA.

It should be reiterated that there are uncertainties as to the whether the tritium release rate measured on this sample is representative of all LANA.75 materials. It may be that this work will yield a DCF unique to this sample, or it may be that LANA releases tritium quickly enough to produce relatively consistent results regardless of the above variables. Additional work is needed to clarify which position is more accurate. In either case, it is believed that tritium aged LANA will give conservative values for tritium release rates, though as an offgassing sample, those rates are expected to be much closer to those of Pd or U than the default values of Hf.

[1]

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SQL*LIMS Task Results Report

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Submission ID	200059142	**Task ID**	401046349
Sample ID	300279314	**Task Status**	COMPLETE
User Sample ID	LANA MATERIAL	**Task Condition**	APPROVED
Submitter	GREGORY.STAACK@SRS.GOV/	**Date Done**	19-NOV-10

LIMS Method	Description	Result (one sigma % unc)	Units	Rv
TRITIUM COUNTING (B145)	TRITIUM	1.08E+06 (10.10%)	uCi/g	1