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# CALIBRATIONS OF A TRITIUM EXTRACTION FACILITY

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## Abstract

A tritium extraction facility has been built for the purpose of measuring the absolute tritium concentration in neutron-irradiated lithium metal samples. Two independent calibration procedures have been used to determine what fraction, if any, of tritium is lost during the extraction process. The first procedure compares independently measured  $^4\text{He}$  and  $^3\text{H}$  concentrations from the  $^6\text{Li}(n,\alpha)\text{T}$  reaction. The second procedure compares measured  $^6\text{Li}(n,\alpha)\text{T}/^{197}\text{Au}(n,\gamma)^{198}\text{Au}$  thermal neutron reaction rate ratios with those obtained from Monte Carlo calculations using well-known cross sections. Both calibration methods show that within experimental errors ( $\sim 1.5\%$ ) no tritium is lost during the extraction process.

## 1. Introduction

The tritium extraction facility at the Argonne National Laboratory (ANL) was developed for the purpose of measuring absolute tritium activities in neutron-irradiated metallic lithium samples. Combining such measurements with relative reactivity determinations and other absolute reaction rate data, energy-integrated capture-to-fission ratios were obtained for the fissile and fertile isotopes of uranium and plutonium in neutron spectra characteristic of Liquid Metal Fast Breeder Reactors (LMFBR).<sup>1-4</sup> In addition, these techniques permitted the direct measurement of the breeding ratio of a typical LMFBR core.<sup>5</sup> More recently the tritium extraction facility was used in the measurement of the  $^7\text{Li}(n,\alpha)\text{T}$  cross section in the 7-9 MeV range.<sup>6</sup> All these measurements depend on the absolute tritium activity in irradiated lithium metal samples. Because these same techniques can be used to measure tritium production rates in the Lithium Blanket Module of the Tokamak Fusion Test Reactor, our experience with this tritium extraction facility is described below.

## 2. Experimental Methods

### 2.1 Sample Preparation

Since lithium is an extremely reactive metal, all the metallic samples were prepared in a clean dry argon atmosphere. A special glove box with thick rigid walls was first evacuated to remove  $\text{N}_2$ ,  $\text{O}_2$  and water vapor, all of which react rapidly with lithium metal, and then filled to one atmosphere with ultra pure argon. Within this atmosphere lithium metal was melted in a stainless beaker and any oxide formation was skimmed off the surface. This clean molten metal was poured into a stainless steel extrusion die and allowed to cool. Either cylindrical or thin ribbon samples, from which foils were punched, were extruded using a hydraulic jack. After cutting to size and weighing on an analytical balance, samples were clad in aluminum (Type 1100) using a cold welding technique. Throughout these operations the lithium metal samples retained their bright shiny appearance.

Two methods have been used to leak-test the samples. The first method simply compares the sample weight obtained immediately after fabrication with that measured immediately before use. A gain in weight signals a cladding failure due to the formation of  $\text{LiOH}$ . The second method is more sensitive and involves submerging the sample in boiling water and looking for the formation of bubbles.

Using these techniques, lithium metal samples have been prepared with masses ranging from 18 mg to 3.5 g. Samples have been made with isotopic abundances from natural lithium to 98.72%  $^6\text{Li}$  to 99.95%  $^7\text{Li}$ .

### 2.2 Tritium Extraction Procedure

Tritium is extracted from irradiated lithium samples by an isotopic dilution method<sup>7</sup> using normal hydrogen as the carrier gas. The extraction apparatus is illustrated in Fig. 1. After placing the sample

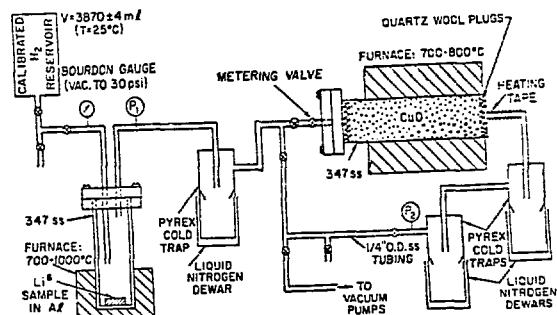


Fig. 1. Schematic Diagram of the Tritium Extraction Facility.

(aluminum can and lithium) in the furnace, the system is evacuated and valved off. About 1 atm of ultra pure hydrogen is added to the oven from the calibrated reservoir. The oven is heated causing the lithium and aluminum can to melt,  $\text{LiH}$  to form, to melt, and then to dissociate as the oven temperature rises above  $850^\circ\text{C}$ . Using a metering valve to control flow rates, the hydrogen-tritium mixture is then allowed to slowly pass over a hot ( $\sim 750^\circ\text{C}$ ) bed of freshly activated  $\text{CuO}$ . Steam ( $\text{H}_2\text{O} + \text{HTO}$ ) from the  $\text{CuO}$  column is collected as ice in a flask maintained at liquid nitrogen temperature. Hydrogen still remaining in the reservoir serves as a carrier to sweep out the last traces of tritium from the system. When all the hydrogen has been converted to ice the run is terminated, the sample weighed, and the water transferred and flame-sealed in a glass ampoule for storage prior to the tritium activity determination. In a typical run about 5.8 g of tritiated water are collected. Before reuse, the stainless steel (Type 347) liner in the lithium oven is carefully immersed in water to dissolve the lithium residue, etched with 6 molar  $\text{HCl}$ , cleaned, and outgassed in a vacuum furnace for 6 hours at  $1000^\circ\text{C}$ .

Type 347 stainless steel was chosen for the liners and furnace walls because of its low hydrogen permeation rate and because it is more corrosion-resistant to molten lithium and LiH than most materials.

Hydrogen losses during the extraction process are determined from the initial P, V, T values for the hydrogen carrier gas and the final mass of the collected water. Pressures are measured with a fused quartz gauge calibrated against standard weights from the National Bureau of Standards (NBS) and periodically verified at atmospheric pressure with an absolute mercury barometer. Without a lithium sample present, the observed hydrogen loss is about 0.3% with the lithium oven on and about 0.1% with the oven off. For small lithium samples (~23 mg) about 0.7% of the hydrogen is lost whereas for 3.5 g samples the loss is about 1.5%. Most of the hydrogen is lost by condensation of LiH onto the cooler surfaces of the liner near the top of the oven, but some is also lost by adsorption and permeation into the furnace walls. If one assumes that the fractional tritium loss is about the same as that for hydrogen, a correction can be applied. Table I shows that this is a reasonable assumption, at least for the small samples (~23 mg) tested. Hydrogen loss is given by the calculated-to-measured water mass ratio  $M(C)/M(E)$ . Tritium loss is given by the activity ratio  $(A_p + A_r)/A_p$  where  $A_p$  is the measured  $^3\text{H}$  activity in the primary water sample and  $A_r$  is the activity of the water in which the Li-Al residue was dissolved.

Table I. Expected and Observed Tritium Losses During the Extraction Process.

Sample	$M(C)/M(E)$	$(A_p + A_r)/A_p$
M4	1.0073	1.0053
M8	1.0068	1.0041
M12	1.0075	1.0051
M16	1.0065	1.0077
M20	1.0074	1.0067
M22	1.0070	1.0060
M24	1.0077	1.0054
M30	1.0080	1.0080
Ave.	$1.0073 \pm 0.0005$	$1.0060 \pm 0.0013$

### 2.3 Tritium Activity Measurement

The activity of tritiated water samples is determined by liquid scintillation methods using a commercial counting facility.<sup>8</sup> With this instrument and Packard's scintillation cocktail INSTA-GEL, 43% counting efficiencies are typical with background count rates of 15-20 cpm. The system is calibrated using standards prepared from the NBS tritiated water standard SRM 4926-C whose stated activity<sup>9</sup> is  $3.405 \times 10^3$  dps/g ( $\pm 0.63\%$ ) on the reference date 9/3/78. For decay corrections a tritium half life of  $12.33 \pm 0.04$  yr<sup>10</sup> was assumed. Irradiated lithium metal samples with tritium activities in the range from 0.5 nCi to  $9 \times 10^4$  nCi have been analyzed by these methods.

### 3. Calibration of the Tritium Extraction Facility Using Helium Mass Spectrometry Methods.

An equal number of tritons and alpha particles are produced in the  $^6\text{Li}(n,\alpha)\text{T}$  reaction. This calibration experiment consisted of independent measurements of tritium and helium produced in identical lithium metal samples exposed to the same neutron fluences. Tritium extraction and counting was done at ANL using the methods already discussed. The helium accumulation

in the samples was measured at Rockwell International (RI), Canoga Park, California using specialized mass spectrometer techniques. Lithium metal ( $^6\text{Li}$ ,  $^7\text{Li}$  and natural Li) wires 1.8 mm diameter and 18 mm long (~20 mg) were extruded, weighed, encapsulated in aluminum and leak tested by the methods discussed earlier. Forty-eight of these capsules were irradiated in the TRIGA reactor at the Hanford Engineering Development Laboratory (HEDL) on September 23, 1981 in two separate irradiations. The neutron fluence in the second irradiation was approximately 25 times higher than that of the first. Twelve samples were mounted symmetrically in each of two layers in an assembly which rotated (1 rpm) during each of the irradiations. Thus, each capsule type in each layer saw the same fluence. Details concerning the Rockwell-ANL intercomparison experiment are given in Ref. 11.

### 3.1 Helium Mass Spectrometer System

The high-sensitivity mass spectrometer system at Rockwell International is specifically designed for low-level helium concentration measurements. The system consists of:

1. a 2-inch-radius permanent magnet mass spectrometer, which operates in isolation from its vacuum pumps to give high sensitivity,
2. furnaces which are used to vaporize and thereby release helium from the sample and which are connected to the mass spectrometer through a series of getters, and
3. several "spike" systems which dispense precisely known small quantities of either  $^3\text{He}$  or  $^4\text{He}$ . The mass spectrometer system is shown schematically in Fig. 2.

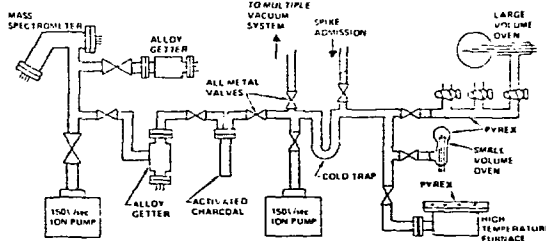


Fig. 2. Schematic Diagram of the Helium Mass Spectrometer System.

To determine helium content, the sample is placed in the furnace and, after evacuation, a precisely known amount of  $^3\text{He}$  spike is added. The sample is then vaporized in resistance-heated tungsten coils or graphite crucibles. After mixing, the gas passes over getters to remove impurities and into the mass spectrometer volume where the  $^4\text{He}/^3\text{He}$  ratio is measured. This ratio together with knowledge of the  $^3\text{He}$  spike and the sample mass determines the helium concentration. The same system is used to measure  $^3\text{He}$  in a sample by means of a spike consisting of  $^4\text{He}$ . Since  $^3\text{He}$  is the decay product of  $^3\text{H}$ , this method permits the measurement of tritium in neutron-irradiated lithium.

The absolute accuracy of the mass spectrometer system is mostly determined by the accuracy of the various helium spiking systems and the linearity of the mass spectrometer electronics. By measuring  $^3\text{He}/^4\text{He}$  ratios for known mixtures  $^3\text{He}$  and  $^4\text{He}$ , by reproducibility considerations, and by combining uncertainties

in the P, V, T measurements for the spiking systems, it has been concluded that the mass spectrometer helium measurements have a 1σ absolute uncertainty of <1%. As an additional check, the helium content of dry air, which is constant over the earth's surface and is accurately known,<sup>12</sup> was measured. The mass spectrometer determination agreed with the accepted value to within 0.3%.

### 3.2 Helium Measurements of Tritiated Water

<sup>3</sup>He is the daughter product of the β-decay of tritium and so accumulates with time in sealed tritiated water samples. To prepare a tritiated water sample for analysis it is degassed to remove previously generated <sup>3</sup>He and any absorbed atmospheric gases, sealed under vacuum in an aluminosilicate glass vial which is impervious to helium diffusion, and stored under refrigeration (~5° C) for subsequent analysis. After a sufficient quantity of <sup>3</sup>He has accumulated in the sample, it is connected to the mass spectrometer system and degassed by ultrasonic vibration methods. After mixing with a known amount of <sup>4</sup>He "spike" a portion of the mixture is admitted to the mass spectrometer and the <sup>3</sup>He/<sup>4</sup>He ratio determined. Combining this measurement with knowledge of the <sup>4</sup>He spike, the sample mass, the tritium decay time and the tritium half life gives the tritium concentration in the water sample. A small correction (~0.1%) is applied to compensate for tritium enrichment due to evaporation loss during the initial degassing.

Because the ANL tritium counting system is calibrated relative to the NBS tritiated water standards, the mass spectrometer system was used to analyze the same NBS standards. Both NBS standards, SRM-4927B and SRM-4926C, were analyzed. The first standard was recalibrated by NBS by internal gas counting in 1978.<sup>9</sup> The second standard was prepared from the first by gravimetric dilution and so the two standards form an interlocked set. Results of the mass spectrometer measurements relative to the NBS certified value for the tritium activity are summarized in Table II. For this comparison a tritium half life of 12.33 yr<sup>10</sup> was used.

Table II. Measurements of <sup>3</sup>He in NBS Tritiated Water Standards.

NBS Standard	No. of <sup>3</sup> He Measurements	Ave. Measured/Certified Ratio
4926C (Lot 1)	5	1.0004 ± 0.0042
4926C (Lot 2)	8	1.0112 ± 0.0025
4927B (Lot 2)	7	1.0074 ± 0.0028
	Mean:	1.0072 ± 0.0052 (1 σ)

These Rockwell measurements (Ref. 11) indicate that the activity of the NBS tritiated water standards is ~0.7% higher than the NBS certified value. Since these measurements were made only a few years after the NBS re-calibration,<sup>9</sup> the results are very insensitive to uncertainties in the tritium half life. NBS quotes an error estimate of 0.63% in their calibration.

Following the tritium extractions at ANL from eight of the irradiated lithium-filled capsules, a portion of the collected tritiated water was sent to Rockwell for <sup>3</sup>He analysis while the remaining portions were counted by liquid scintillation methods. The results of the RI <sup>3</sup>He and the ANL <sup>3</sup>H analyses are compared in Table III.

Table III. RI and ANL Measurements of ANL-Extracted Tritiated Water.

Sample	Tritium Activity (10 <sup>3</sup> dps/g)		
	RI	ANL	RI/ANL
NLi-5	6.591	6.532	1.009
NLi-7	6.784	6.699	1.013
<sup>6</sup> Li-2	25.26	25.08	1.007
NLi-23	152.5	151.4	1.007
NLi-25	151.6	149.2	1.016
<sup>6</sup> Li-7	561.6	556.9	1.003

Ave. 1.010 ± 0.0035

Since the ANL results are based on the calibration of the NBS tritiated water standard which RI found to be low by ~0.7% (Table II), it is concluded that the Rockwell and ANL measurements on these tritiated water samples are entirely consistent within the experimental uncertainties.

### 3.3 Helium Measurements of Vaporized Lithium Capsules

By the methods discussed earlier, the Rockwell mass spectrometer system was used to measure the <sup>4</sup>He content in the irradiated lithium capsules. Small corrections were applied for <sup>4</sup>He released from unirradiated control capsules. Two of the samples were analyzed for both <sup>3</sup>He and <sup>4</sup>He in order to show that equal amounts of <sup>4</sup>He and <sup>3</sup>H were generated during the irradiation and that no tritium was lost from the capsules. This was done by measuring the <sup>4</sup>He/<sup>3</sup>He ratio of an unspiked portion of the sample gas and then analyzing the remaining gas mixed with a known <sup>3</sup>He spike. The results of these measurements on vaporized lithium samples are summarized in Table IV and are compared with the ANL tritium results obtained from identically irradiated samples. A tritium half life of 12.33 years was used for this comparison.

Table IV. Rockwell and ANL Measurements of the Products of the <sup>6</sup>Li(n,α)T Reaction.

Sample	10 <sup>15</sup> Atoms per Gram of Lithium			Ave. Ratio RI/ANL
	<sup>4</sup> He (RI)	T from <sup>3</sup> He (RI)	T* (ANL)	
NLi-4	1.110			
NLi-5			1.086	
NLi-6	1.107			
NLi-7			1.070	
NLi-8	1.097			
NLi-9			1.096	
Ave.	1.1047 ± 0.0068		1.084 ± 0.013	1.019 ± 0.014
<sup>6</sup> Li-1	4.45			
<sup>6</sup> Li-2			4.404	1.010
NLi-22	25.21	25.23		
NLi-23			25.54	
NLi-24	24.60	24.41		
NLi-25			24.23	
NLi-26	24.46			
NLi-27			23.87	
Ave.	24.76 ± 0.40		24.21 ± 0.34	1.022 ± 0.022
<sup>6</sup> Li-6	99.45			
<sup>6</sup> Li-7			100.6	0.9886
			Ave. 1.010 ± 0.015	

\*Based on RI's measurement of the NBS tritiated water standard (see Table II).

Table IV shows that tritium concentrations based on  $^3\text{He}$  measurements are in very good agreement with the  $^4\text{He}$  concentrations. This proves that no measurable amounts of tritium escaped from the capsule during and following the irradiation and that  $^4\text{He}$  generation from impurities (such as boron) was negligible. The last column in the table shows that tritium concentrations in the extracted water samples agree, within experimental uncertainties, with the  $^4\text{He}$  concentrations determined by mass spectrometry. This suggests that there are no significant systematic tritium losses associated with the ANL tritium extraction methods. The data also indicates that there is no observable variation in the tritium extraction efficiency over the concentration range encountered in these measurements.

To compare the ANL tritium measurements with the Rockwell helium determinations requires converting tritium activities to triton atom concentrations. This conversion is directly proportional to the tritium half life. Based on counting results of the NBS standards over an 18 year period, NBS<sup>9</sup> suggests a half life of 12.43 years. If the ANL data were normalized to this value, the RI/ANL ratio in the last column of Table IV would be reduced by 1.0%. Preliminary  $^3\text{He}$  data obtained by Rockwell indicate a tritium half life of  $\sim 12.38$  years.

The results presented in this paper are based on a second ANL/RI intercomparison experiment. Results from the first intercomparison are given in Ref's. 11 and 13 and showed that the Rockwell measurements were about 6% larger than those obtained at ANL. The Rockwell data suggested that an unrecognized tritium loss occurred during the extraction process at ANL. Measurements of  $^3\text{He}$  and  $^4\text{He}$  concentrations in the same lithium capsules indicated equal  $^4\text{He}$  and  $^3\text{H}$  production rates thereby showing no failure in the cladding or cold weld seals. On the other hand, it is difficult to postulate some other mechanism which would result in a 6% tritium loss during the extraction without changing the loss of hydrogen carrier gas (typically about 0.7%). For this reason the measurements were repeated. The same procedures were used at ANL and Rockwell for both intercomparison experiments although greater attention was paid to sealing and leak-testing the lithium capsules for the second intercomparison.

#### 4. Calibration of the Tritium Extraction Facility Using the $^6\text{Li}(n,\alpha)^3\text{T}/^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ Thermal Neutron Activity Ratio.

A second method, independent of the first, was undertaken to see if a systematic loss of tritium associated with the extraction procedures could be identified and, if necessary, accounted for. This calibration method consists of simultaneously irradiating in equal thermal neutron fluxes lithium and gold samples. The measured  $^6\text{Li}(n,\alpha)^3\text{T}/^{197}\text{Au}(n,\gamma)^{198}\text{Au}$  activity ratio is then compared with that calculated from the well-known thermal neutron cross sections.

##### 4.1 Measured $^6\text{Li}(n,\alpha)^3\text{T}/^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ Thermal Neutron Activity Ratios.

Two calibrations of this type were performed. In the first case 0.75 inch diameter foils of both used. To minimize flux perturbation and neutron self-shielding effects, metallic lithium foils ( $\sim 33$  mg) depleted in  $^6\text{Li}$  (1.55 wt%  $^6\text{Li}$ ) were prepared by the methods described in Section 2.1. Lithium and gold foils were simultaneously activated in the graphite thermal column of the Argonne Thermal Source Reactor (ATSR). Both foil types were mounted symmetrically in equal angular intervals on a thin aluminum disk 4.25 inches from the center. During the irradiations the disk, located within a void in the thermal column, was

inches from the center. During the irradiations the disk, located within a void in the thermal column, was uniformly rotated with its plane parallel to the reactor leakage face and about 111 cm from it. The methods described in sections 2.2 and 2.3 were used to determine the tritium activities. Absolute gold foil activities were measured by the  $4\pi\beta\gamma$  coincidence method and calibrated Ge(Li) detectors. The gold foil activities were multiplied by 0.9948 to correct for epithermal activation as determined from cadmium ratio measurements. These foil measurements are described in detail in Ref. 14. Table V summarizes the measured thermal neutron activity ratios and includes data not available when Ref. 14 was written.

Table V. Experimental  $^6\text{Li}(n,\alpha)^3\text{T}/^{197}\text{Au}(n,\gamma)^{198}\text{Au}$  Thermal Neutron Activity Ratios, AR(E).

ATSR Irrad.	Li Samples	Ave. AR(E)	Error (1 $\sigma$ ), %		
			Std. Dev.	Systematic	Total
4-10-80	6 Foils	$2.713 \times 10^{-3}$	0.72	0.93	1.2
4-17-80	4 Foils	$2.780 \times 10^{-3}$	0.72	0.93	1.2
8-08-80	8 Foils	$2.767 \times 10^{-3}$	0.77	0.89	1.2
Weighted Mean:		$2.752 \times 10^{-3}$	1.1	0.92	1.4
12-09-82	2 thick	$3.964 \times 10^{-3}$	1.6	0.99	1.9

For the second thermal neutron calibration thick 0.70 inch diameter cylinders of natural lithium metal were used. These samples had masses of the order of 3.5 grams. Gold foils 1 mil thick were placed directly onto the aluminum clad cylindrical lithium samples. As before, these samples were irradiated on a rotating disk in the graphite thermal column of the ASTR reactor. The measured activity ratio for these thick samples is shown at the bottom of Table V. The largest factors contributing to the systematic errors shown in Table V are uncertainties in the calibration of the tritiated water standard (0.63%) and the detector efficiency (0.67%) for measuring the absolute gold foil activities. Finally, the total error is obtained by adding the two component errors in quadrature.

##### 4.2 Calculated $^6\text{Li}(n,\alpha)^3\text{T}/^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ Thermal Neutron Activity Ratios.

For the Li/Au foil measurements standard foil perturbation methods were used to calculate the thermal neutron activity ratio analytically (Ref. 14, pp. 19-26). In addition to these analytical calculations, fixed source continuous energy Monte Carlo methods were used to determine the activity ratios for both thin foil and thick cylindrical samples. For these calculations the air-filled cavity in the ASTR thermal column, the aluminum disk, the gold foils and each of the aluminum clad lithium samples (foils and cylinders) were explicitly modeled. The cavity was represented as a rectangular box 16.53" x 12.0" x 4.0" with a vacuum (leakage) boundary condition on the top and reflective boundary conditions on all the other sides. The side parallel to the reactor leakage face and closest to it was chosen as the source plane for the Monte Carlo calculations.

For these fixed source calculations it is necessary to specify the energy distribution, the angular distribution, and the position coordinates of each neutron emitted from the source plane. For the position coordinates random numbers uniformly distributed on the interval (-1,1) were used. Fermi's angular distribution<sup>15</sup> was used for thermal neutrons emitted by the source plane. It was found, however, that the same

activity ratio was calculated if one assumed an isotropic distribution of direction cosines. The energy distribution of the source neutrons was taken to be that of a pure Maxwellian,

$$P(E)dE = \frac{E}{kT} e^{-E/kT} \frac{dE}{kT}$$

where E is the energy, k the Boltzmann constant and T is the absolute temperature. Cadmium ratio measurements<sup>14</sup> were used to correct the gold foil activities for the small epithermal activation of the gold resonance at 4.91 eV. Because of neutron absorption by impurities in the graphite thermal column, the temperature of the Maxwellian distribution is somewhat greater than the ambient temperature. Based on a spectrographic analysis of the graphite impurities, the neutron temperature was calculated to be 322 K.

Calculations for the activity ratios were performed using the continuous energy Monte Carlo code VIM.<sup>16</sup> Cross sections were taken from the ENDF/B Version IV data base. Sufficient neutron histories were followed so as to provide better than 1% statistics in the calculated activity ratio.

Table VI summarizes the calculations for the  ${}^6\text{Li}(n, \alpha)\text{T}/{}^{197}\text{Au}(n, \gamma){}^{198}\text{Au}$  thermal neutron activity ratios and shows in the last column the calculated-to-measured quantity, AR(C)/AR(E). Note that for the lithium foil experiment, the analytical and Monte Carlo calculations for the activity ratio AR(C) are in very good agreement. Systematic errors include uncertainties in the thermal neutron activation cross sections for  ${}^6\text{Li}$  and gold, errors in the half lives, and errors associated with the measured  ${}^6\text{Li}$  content of the depleted lithium foils. The last column shows good agreement between the calculated and measured activity ratios

Table VI. Calculated Thermal Neutron  ${}^6\text{Li}(n, \alpha)\text{T}/{}^{197}\text{Au}(n, \gamma){}^{198}\text{Au}$  Activity Ratios, AR(C).

Li Sample Type	Calculation Method	AR(C)	Error (1 $\sigma$ ), %			AR(C)/AR(E)
			Random	Systematic	Total	
Thin Foil	Analytical	$2.7933 \times 10^{-3}$		1.1	1.1	$1.015 \pm 0.018$
Thin Foil	Monte Carlo	$2.7905 \times 10^{-3}$	1.1	1.0	1.5	$1.014 \pm 0.021$
Thick Cyl.	Monte Carlo	$3.9886 \times 10^{-3}$	0.84	0.62	1.0	$1.006 \pm 0.022$

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indicating no significant loss of tritium in the extraction process. This result supports the findings of the helium mass spectrometer measurements (see Table IV).

#### 5. Conclusions

These calibration studies were undertaken to see if a systematic loss of tritium associated with the extraction procedures could be identified and, if necessary, accounted for. The following conclusions have been reached:

- Rockwell helium mass spectrometer measurements on the NBS tritiated water standards indicate that the NBS-certified activities may be low by about 0.7%.
- Based on the Rockwell calibration of the NBS tritiated water standard, absolute measurements of  ${}^4\text{He}$  (RI) and tritium (ANL) concentrations from the  ${}^6\text{Li}(n, \alpha)\text{T}$  reaction agree to within 1%.
- Calculated and measured  ${}^6\text{Li}(n, \alpha)\text{T}/{}^{197}\text{Au}(n, \gamma){}^{198}\text{Au}$  thermal neutron activity ratios agree to within 1.5% when the results are normalized to the Rockwell calibration of the NBS tritiated water standard.
- Both calibration methods indicate that within the experimental uncertainties no systematic losses of tritium occur during the process used to extract  ${}^3\text{H}$  from neutron irradiated lithium metal samples.
- The Rockwell and ANL data support an increase in the tritium half life with respect to the 12.33 years used in these analyses. Preliminary helium data obtained by Rockwell indicate a half life of  $\sim 12.38$  years. This half life change would reduce the RI/ANL ratio (Table IV) by about 0.5% and likewise reduce AR(C)/AR(E) (Table VI) by a similar amount.