Preparation of a one-curie ^{171}Tm target for the detector for advanced neutron capture experiments (DANCE)

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Approximately one curie of ^{171}Tm ($T_{1/2} = 1.92$a) has been produced and purified for the purpose of making a nuclear target for the first measurements of its neutron capture cross section. Target preparation consisted of three key steps: (1) material production; (2) separation and purification; and (3) electrodeposition onto a suitable backing material. Approximately 1.5 mg of the target material (at the time of separation) was produced by irradiating ca. 250 mg of its stable enriched ^{170}Er lanthanide neighbour with neutrons at the ILL reactor in France. This production method resulted in a "difficult-to-separate" 1:167 mixture of near-neighboring lanthanides, Tm and Er. Separation and purification was accomplished using high-performance liquid chromatography (HPLC), with a proprietary cation-exchange column (Dionex, CS-3) and alpha-hydroxyisobutyric acid (α-HIB) eluent. This technique yielded a final product of ~95% purity with respect to Tm. A portion (20 μg) of the Tm was electrodeposited onto thin Be foil and delivered to the Los Alamos Neutron Science Center (LANSCE) for preliminary analysis of its neutron capture cross section using the Detector for Advanced Neutron Capture Experiments (DANCE). This paper discusses the major hurdles associated with the separation and purification step, including scale-up issues related to the use of HPLC for material separation and purification of the target material from α-HIB and 4-(2-pyridylazo)resorcinol (PAR) colorant.

Introduction

Neutron capture cross sections, σ(n,γ), for long-lived radioactive nuclei lying close to the valley of β$^-$-stability are of importance to the quantitative understanding of the s-(or slow) process in nucleosynthesis, Science Based Stockpile Stewardship (SBSS), the Advanced Fuel Cycle Initiative, and nuclear attribution and forensics. Theoretical estimates of σ(n,γ) for β$^-$-unstable nuclei vary by a factor of 2 or more from experimental values and in many cases key cross sections are unknown, so experimental measurements are necessary. Recently, the Detector for Advanced Neutron Capture Experiments (DANCE),[1] located at the Los Alamos Neutron Science Center (LANSCE), has been commissioned to experimentally measure σ(n,γ) for radioactive targets. The DANCE is designed as a highly-segmented, high-efficiency, 4π BaF$_2$ detector array to calorimetrically detect γ-rays following neutron capture. The measurement of nearly 100% of the gamma-rays emitted from neutron capture allows resonant peaks to be assigned to individual isotopes based upon their Q-values. Combined with the strong neutron source of LANSCE, the DANCE is able to measure σ(n,γ) of radioactive isotopes with activity levels up to several Ci. However, targets of radioactive material must be prepared first before measurements can be made.

Currently, the preparation of three radioactive lanthanide targets are planned for measurement of their σ(n,γ) using DANCE. These targets include ^{147}Pm, ^{155}Eu, and ^{171}Tm and each represents "branching point" nucleus within the s-process of nucleosynthesis. To date, no accurate measurements of their neutron capture cross sections have been made. This paper discusses the hurdles associated with the production of the first of these targets, ^{171}Tm.

Experimental

Material production

To provide sufficient quantities (~mg) to make accelerator targets, several hundred milligrams of isotopically enriched ^{170}Er was irradiated at the Institut Laue-Langevin (ILL) high flux ($2 \cdot 10^{15}$ n·cm^{-2}·s^{-1}) reactor in Grenoble, France. This material was subsequently shipped to Los Alamos National Lab for further processing. The radioactive product, ^{171}Tm (β$^-$, 1.9 a), was produced in the following reaction: ^{170}Er (n,γ)^{171}Er (β$^-$, 7.5 hr)^{171}Tm. This production and decay scheme is illustrated in Fig. 1. Based upon the planned 50-day irradiation, it was expected that ~10 mg of ^{171}Tm would be produced from 250 mg of ^{170}Er.

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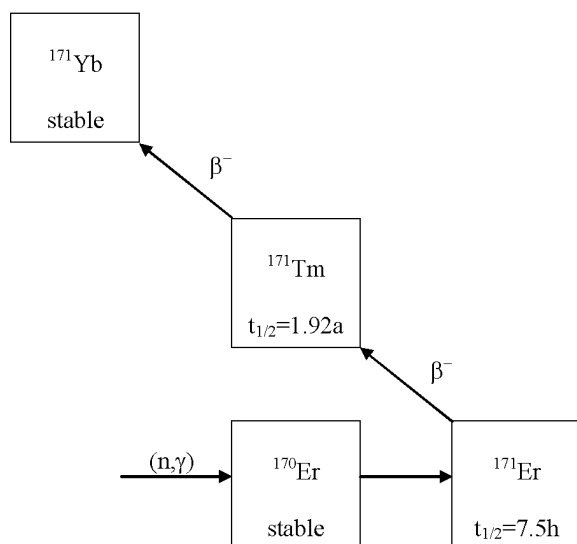E-mail: Jon.Schwantes@pnl.gov

Fig. 1. Production and decay scheme for the radioactive target material ^{171}Tm. The radioactive isotope was produced by neutron irradiation of ^{170}Er at the high-flux ILL reactor in Grenoble, France

Process overview

The contamination ratio, or the ratio of stable enriched ^{170}Er to ^{171}Tm, was expected to be approximately 25. This meant that prior to producing a usable target, radioactive ^{171}Tm must be separated from the excess amount of ^{170}Er. Our goal was to produce a target of ^{171}Tm that was >90% pure. A target of higher purity in this case was not warranted since it is possible with DANCE to discriminate between the capture gamma-rays based on the Q-values of the target isotopes: ^{171}Tm (Q-value = 6.2 MeV) and ^{170}Er (Q-value = 5.7 MeV).[2] By producing ^{171}Tm from a completely different isotopically enriched element, we could employ a chemical purification method to produce an isotopically pure target. Chemical purification of Tm was accomplished using HPLC and a proprietary cation exchange column. We used alpha hydroxyisobutyric acid (α-HIB) as the eluent of choice for its known performance in effectively separating neighboring lanthanides[3] and for its characteristic reverse (highest mass first) elution pattern for lanthanides. This latter characteristic minimized the loss of product while maximizing the extent of separation by eluting the ^{171}Tm prior to the contaminant ^{170}Er.

HPLC apparatus and rapid lanthanide separation method

Separations were carried out using a Dionex IC-2500 HPLC, equipped with a GP-50 gradient pump, CG-3 guard and CS-3 analytical columns. The CS-3 cation-exchange column (0.4 cm diameter × 25 cm length) is comprized of a polystyrene/divinyl benzene support and was placed in-line following the CG-3 guard column. The CG-3 column is identical in all respects to the CS-3 column except column length (5 cm long). Tm fractions were collected manually, assisted by a shielded Geiger-Müller counter, which monitoring the activity of the effluent from the HPLC.

It was vital to employ a method capable of extremely effective separations since our goal here was to separate one lanthanide element from a large excess of neighboring lanthanide. We previously developed a separation method for this purpose.[4] Briefly, separation of near-neighboring lanthanide pairs was optimized by altering both the pH and concentration of the α-HIB eluent. Separation resolution (SR) is used to characterize the extent of separation between two peaks ($x = 1, 2$) and is equal to:

$$SR = \frac{2(R_{T,2} - R_{T,1})}{(W_2 + W_1)} \quad (1)$$

where RT_X and W_X represent the retention time (RT) and peak width (W) for peak X, respectively. The published average SRs for HPLC methods utilizing α-HIB and a cation exchange column range from 1.5–2.4.[5–14] However, Schwantes et al.[4] found by optimizing both pH and α-HIB concentration for each individual pair of neighboring lanthanides, higher SRs (between 2.8 and 3.4) are obtainable in a fraction of the time of a traditional gradient elution scheme. For the Er/Tm lanthanide pair, an eluent at 0.1M α-HIB at a pH of 3.6 produced a SR of 2.9 in 14 minutes.[4] For our application, this method was scaled up for a system using a 1-ml injection loop.

Initial sample preparation

Ultra-clean laboratory practices were utilized in all steps of this work. All lab ware was allowed to soak for a minimum of 3 days in a Liquinox bath and rinsed 5 times with 8M trace-metal-grade HNO_3 and >18 MΩ deionized water before use. Reagent preparation of the α-HIB and the post reaction reagent, 4-(2-pyridylazo)resorcinol (PAR, Aldrich), followed the published method[15] with the exception of substituting KOH for NH_4OH during pH adjustment. PAR was used as a colorant for detecting lanthanides by UV-Vis spectrophotometry. When complexed with lanthanides, the PAR-lanthanide complex absorbs light in the visible spectrum at a wavelength of 530 nm. Measurements of eluent pH were conducted in triplicate using a model 617 Keithley electrometer and a Ross electrode from Orion and converting electrode potential readings to pH using standard solutions (Hydraion) at pH values of 2.00, 4.00, and 7.00. All pH measurements were accurate to ±0.05 pH units at two standard deviations.

Roughly 250 mg of mixed Er/Tm oxide was shipped back from the ILL reactor in France and delivered to a hot cell within building RC-1 at Los Alamos National Laboratory (LANL). This material was initially dissolved in ~30 ml of 0.5M HCl. This was a relatively strong acid strength for a HPLC load solution, but was chosen in order to maximize the lanthanide concentration in solution, prevent precipitation, and minimize the number of injections required to pass the entire sample through the HPLC. However, column overloading was also a concern during the first cycle through the HPLC. Preliminary experiments indicated this mass loading was below the combined capacity of both the guard and analytical columns.

Post-HPLC processing

Alpha-HIB and PAR had to be removed from the Tm fraction following its separation from Er via HPLC. To accomplish this, a series of column separations were designed and tested on Tm/PAR/α-HIB mixtures. Since PAR-lanthanide complexes are known to form at high-pH[16] and α-HIB-lanthanide complexes are known to form at moderate pH,[17] it was assumed the PAR and α-HIB could be easily separated from the lanthanide under acidic conditions using a simple cation-exchange column. Experiments were first carried out separately for PAR/lanthanide and α-HIB/lanthanide mixtures using a large-bore, 1.2-cm i.d., gravity fed column containing 5-ml of Biorad AG-50x8, 100–200 mesh, cation-exchange resin. Mixtures were acidified dropwise with 6M HCl to the desired starting acid concentration (between 0.03M and 2M) and loaded onto the column. Once loaded, progressively stronger acid, from 0.5–8M HCl, were tested in an attempt to separate Tm from the PAR and α-HIB. Load solutions and eluent were gamma-counted for material balance of the [171]Tm tracer to measure progress of the separation. Due to its characteristic coloration, the location of PAR within the column was easily observed, and typically remained within a small band at the top of the column. Each load solution and eluent of the α-HIB/lanthanide separation experiments were also taken to dryness to confirm the presence/absence of α-HIB residue visually.

Electrolytic cell, molecular plating method, and target mounting

Significant efforts were expended identifying a suitable target backing material, selecting and optimizing an appropriate deposition technique, and designing a capable electrolytic cell. Ti and Be were compared with respect to their nuclear cross section characteristics. Backing material with the lowest overall total cross section, σ(n,tot.), was preferred. The total cross section is defined here as the sum of both neutron capture σ(n,γ) and neutron scattering σ(n,n') cross sections and both are a function of neutron energy. The ratio of the Be/Ti total cross section as a function of energy are shown in Fig. 2. The higher ratio at lower neutron energy is largely due to neutron scatter by Be, while the dip in ratio at moderate neutron energy is due to a neutron capture resonance peak of Ti. From this figure it is evident that within the energy range of interest for the DANCE (i.e., 1 keV–30 keV), Be has a lower σ(n,tot.) than that of equal thickness Ti. As a result, Be was the backing material of choice for neutron capture experiments with DANCE.

Target preparation techniques typically include evaporation or condensation, electrospraying, precipitation, and electrodeposition.[18] Electrodeposition, which is the process of depositing one metal upon another using electrolysis,[19] was chosen for this work as it provides for relatively high yields, simple handling, easy recovery of the target material following deposition, and the ability to deposit material homogeneously over a range of mass thicknesses.[18]

A high-voltage molecular plating procedure from isopropanol solution, originally developed for actinides[20] was adapted for depositing lanthanides on Be.[21] Molecular plating was necessary since aqueous electroplating procedures are incompatible with Be metal. Briefly, a lanthanide (Ln) stock solution in isopropanol was prepared by first converting the Ln to a nitrate form by dissolution of the oxide in concentrated nitric acid followed by evaporation to dryness. Following several dissolution-evaporation cycles to ensure complete conversion, the dried nitrate salt was dissolved in isopropanol. Be backing foil, ~10-μm thick, was placed on the O-ring between the bottom of the cell and the Al base. Approximately 4.2 ml of isopropanol containing 100 μg·ml^{-1} Ln stock was added to the plating cell. The high voltage cable was connected to the plating cell and the glass stir rod was lowered into the solution and rotated at 10–60 Hz. A voltage of ~+100 V to +300 V sufficient to keep a current density around 0.8 mA·cm^{-2} was applied to the cell for 30 minutes. Targets were deposited in four cycles, gently drying the target under a heat lamp for 10 minutes and then baking for 30 minutes at 550 °C after each cycle.
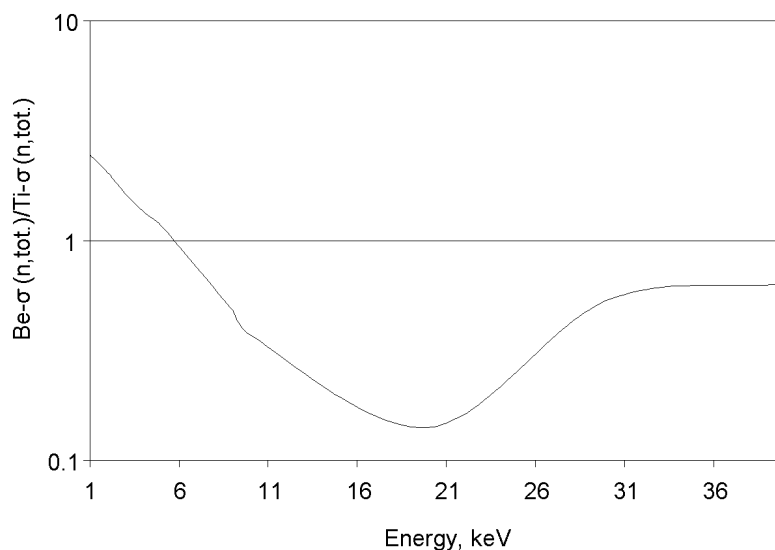
Fig. 2. Ratio of Be/Ti total neutron cross sections [σ(n,tot.)] as a function of energy

Results and discussion

Peak width, retention time, and separation resolution as a function of scale

Retention times (RTs), peak width, and SR were all affected by scale up. Retention times shifted roughly linearly with mass loading, decreasing by 1 percent per 40 µg of loading. Peak widths also increased linearly with mass loading. Figure 3 shows the change in relative peak width with mass loading. The overall shape of the peak was not changed with scale but peak tailing was always a prominent feature. On average, the slope of the leading edge of elution peaks were 1.85 times larger than the trailing edge, so 65% of the increase in peak width as a result of mass loading occurred on the tailing edge of the peak. Peak tailing is a common problem during lanthanide separations using HPLC, but has been minimized in the past with heat at the cost of increasing retention times and thus the amount of waste produced.[22] We chose not to heat our column for those reasons. Based on the change in peak width and RT with mass loading, we were also able to estimate the expected change in SR between Tm and Er as a function of Er mass loading relative to 1:1 mass loadings (Fig. 4). Since we were able to obtain SRs of around 3 for 1:1 mass loadings in preliminary experiments[4] and expected our initial separation cycle for producing the [171]Tm target would correspond to a mass loading of 25:1 (C_2/C_1 in Fig. 4), we, therefore, expected to achieve a SR of roughly 1.5. It was expected that this initial SR would be sufficient for our purposes, thereby avoiding the need to heat the column, which would have extended retention times and increased waste volume.

Separating lanthanides from α-HIB and PAR following HPLC

Figure 5 presents the results from thermodynamic modeling of the speciation of an aqueous mixture of α-HIB and Eu (as an analog for lanthanide behavior) at different pH. Based on these predictions, α-HIB was not expected to protonate or form cationic complexes with lanthanides in acidic aqueous solutions, and therefore, was not expected to interact with the cation-exchange resin. PAR is also not expected to form complexes with lanthanides at low pH, but may protonate into a cationic species.[16] This meant lanthanides should have been easily separated from α-HIB by simply loading the mixture on a cation column at low pH. The lanthanide would subsequently be separated from the PAR by elution with higher concentration acid.
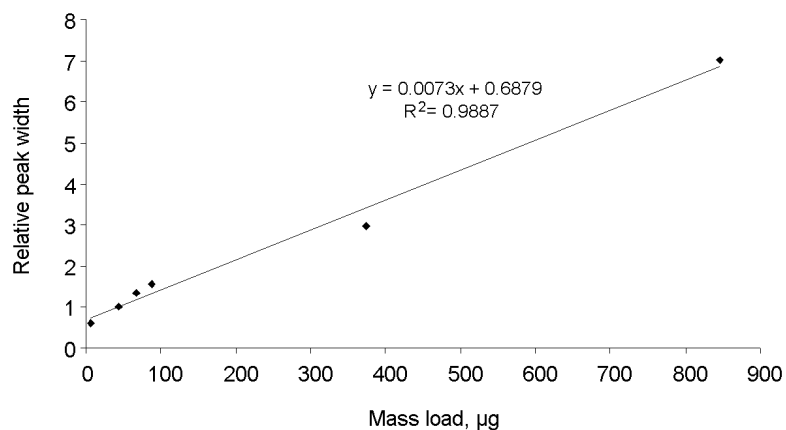
Fig. 3. Relative peak width (normalized to the width at 43 μg loading) of Er as a function of mass loading. Eluted with 0.1M α-HIB at pH 3.6
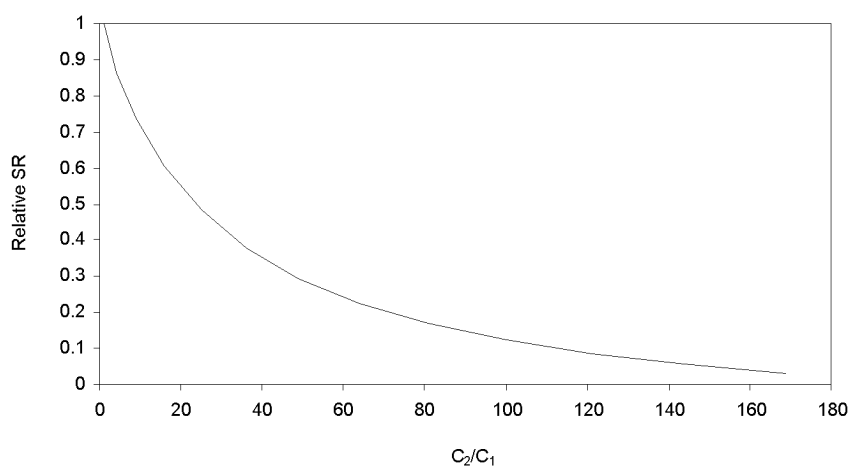

Fig. 4. Calculated change in SR between Tm and Er as a function of Er mass loading expressed as the ratio of the Er (C_2) to Tm (C_1) concentration within the load solution. SR shown relative to that for a 1 : 1 mass loading of Tm and Er of 7 μg each
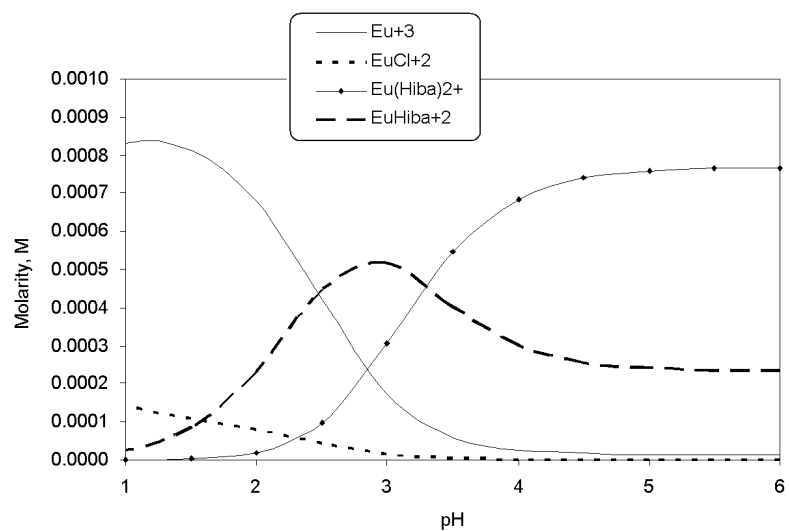

Fig. 5. Thermodynamic modeling of $1 \cdot 10^{-3}$M Eu and 0.1M α-HIB as a function of pH in HCl. Modeling was conducted using PHREEQC[23] geochemical modeling software, based upon reviewed published thermodynamic constants[24,25]

Experiments were first conducted to separate Tm/PAR mixtures. The initial mixture was acidified to a pH between 1 and 1.5 and loaded on a column containing 5-ml AG-50x8, 100–200 mesh, cation exchange resin. Eluting with 4 column volumes (CVs) each of progressively more concentrated acid from 0.5–6M HCl, we attempted to strip Tm from the column without also stripping the PAR. The movement of Tm through the column was tracked by gamma-counting analysis of each of the eluents and is summarized in Fig. 6. PAR began to slowly migrate from the top of the column at 4.5M HCl, with significant break-through occurring with 5M HCl. Figure 6 indicates 4M HCl qualitatively strips Tm from the column. However, only 85% of the total Tm loaded on the column was recovered. A follow-on isocratic elution experiment confirmed that 7 CVs of 4.5M HCl quantitatively recovered the Tm from the column and effectively separated it from PAR (Fig. 7).

The same basic strategy applied to lanthanide/PAR mixtures, namely the application of a series of stepwise elutions with progressively more acidic solution, was initially used for lanthanide/α-HIB mixtures in hopes of developing a capable separation technique. An HCl load solution at pH 1 containing a mixture of lanthanide and α-HIB was loaded onto an identical column as that used for the lanthanide/PAR separation experiments. At this pH, the lanthanide ions were expected to be retained on the column while allowing the α-HIB to pass through the column. Interestingly, this expected behavior was not observed. Instead, the α-HIB remained in the higher-strength eluent (>3M HCl) fractions that also contained a significant portion of the Tm. In addition, Tm began to elute from the column at a much lower acid strength in the presence of α-HIB versus PAR.

Our second attempt at separating α-HIB/lanthanide mixtures began with loading the mixture on the column at much higher acid strength, closely following the method developed by MILLER et al.[22] Our approach, however, utilized AG 50x8 cation-exchange resin rather than AG MP-50 resin, and loaded the lanthanide/α-HIB mixture on the column at an acid strength of 2.7M rather than 2M. Under these conditions the majority of the α-HIB ran through the column during loading while the lanthanide was held up on the column. Breakthrough of the Tm began to occur at ~4M HCl, however, final refinements of this procedure called for 8M HCl to elute Tm from the column.

A comparison of results from the first and second attempts at separating Tm from α-HIB likely underlines the importance of kinetics of complex formation between the lanthanide and α-HIB. In the first attempt, where acid strength was steadily increased in a stepwise fashion, the majority of the α-HIB and Tm were observed in the 3M acid strength eluent. In the second attempt, however, the majority of the α-HIB eluted with the 2.7M acid strength load solution, while Tm was held up on the column under much stronger acid conditions (breakthrough occurring at ~4M HCl). We believe these differences suggest the lanthanide/α-HIB complex was still intact (at pH ~1) during the first experiment when the solution was loaded onto the column, remaining at least kinetically stable through some portion of the experiment. This lanthanide/α-HIB complex would be expected to have a lower affinity for the cation resin than the free Tm ions, which could explain why Tm eluted at a lower acid strength (~3M) in the first lanthanide/α-HIB separation experiment than in the lanthanide/PAR experiment (~4M). Presumably, this Tm/α-HIB complex was destroyed prior to loading during equilibration and acidification of the load solution in the second Tm/α-HIB separation experiment.
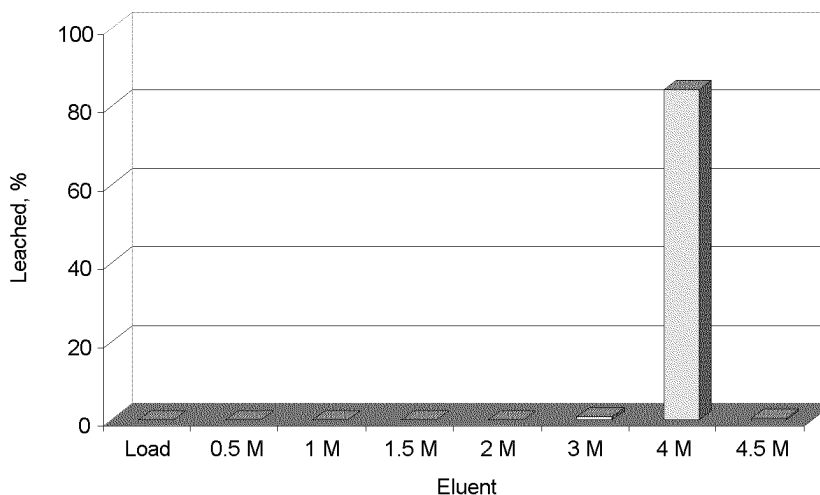

Fig. 6. Percent Tm eluted from AG 50x8 cation-exchange resin in the presence of PAR
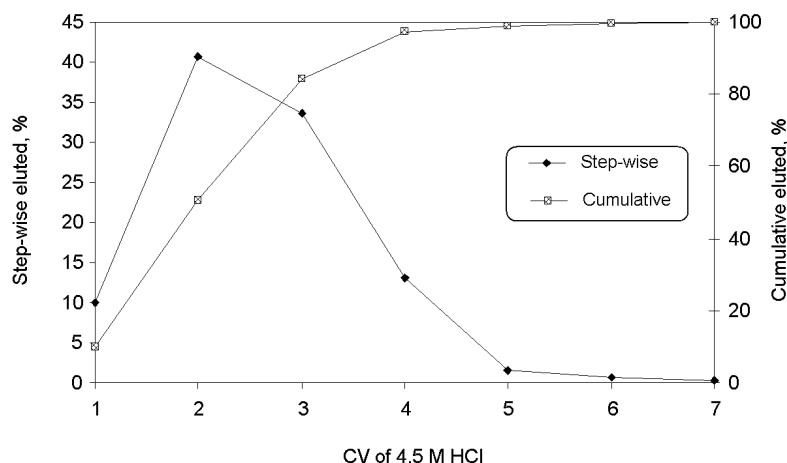
Fig. 7. Elution of Tm from AG 50x8 cation-exchange resin with 4.5M HCl

Effectiveness of HPLC for material separation and purification

It was necessary to cycle Tm/Er mixtures through the HPLC a total of three times to obtain a final Tm purity of >90%. The progress of the HPLC separation/purification for each of the cycles is illustrated in Fig. 8. The purity of the Tm fraction after the third and final cycle was roughly 95%. It should also be noted that the completion of each cycle represented approximately 25 separate injections into the HPLC. While this represents an appreciable amount of effort due to the limitations of sample size that could be introduced into our HPLC, the advantages with respect to separation performance, waste minimization, and automation far outweigh the disadvantages over that of conventional chromatography. Additionally, the 0.4-cm diameter CS-3 column is also available in a 1-cm diameter version that is compatible with the GP-50 gradient pump we used here. Had we utilized the 1-cm column, the injection size and material loading could have been increased by roughly a factor of 6.

Contamination issues in the final product

While we effectively separated Tm from Er, a significant amount of stable [169]Tm was present in our final product. Based on an assay of the initial starting material, the contamination ratio of Er:Tm was ~167. Correcting for decay since the time of irradiation (Er:Tm at the time of irradiation was ~63), this ratio was significantly greater than what was expected (~25) from initial predictions for a 50-day ILL irradiation of 250 mg of [170]Er. Additionally, assuming 100% of the Yb signal in the HPLC assay was from [171]Yb as the decay product of [171]Tm, the measured peak for Tm was approximately 50% larger than expected based upon the date of irradiation. This suggested a significant amount of a stable Tm might be present as contaminant within the sample. The presence of stable [169]Tm contaminant was confirmed by $\sigma(n,\gamma)$ resonance analysis using DANCE on a small electrodeposited target (~20 μg). Figure 9 shows the sum of the measured γ-energy (Esum) for the small Tm target as a function of neutron energy. For comparison, the spectrum from a 2-Ci Tm target measured by an early version of DANCE that utilized C_6D_6 detectors is also provided (Fig. 10). The C_6D_6 detectors have since been replaced with BaF_2 crystals to avoid issues related to peak summing at higher neutron energies (notice the rising baseline with energy in Fig. 10). However, since energy resolution was not an issue with C6D6 detectors, comparison of resonance peaks generated from these two different types of crystals is warranted. Qualitatively, the majority of the resonance peaks in Fig. 10 can be assigned to [171]Tm, while the strongest peaks in Fig. 10 are attributed to the stable [169]Tm. These peaks were assigned to specific isotopes by comparison of these spectra with published[2] resonance peaks, further discriminating peaks based upon Q-values (not shown). Furthermore, a semi-quantitative estimate of contamination from the stable Tm isotope was made by normalizing the area under a known resonance peak for each isotope with respect to their published $\sigma(n,\gamma)$ cross sections.[2] Based on these calculations, [169]Tm made up roughly 80%±30% of the total Tm present on the small target. The high error bars were largely due to errors associated with published $\sigma(n,\gamma)$ cross section values, but agree with earlier estimates based upon HPLC assays. In any case, DANCE analysis provided positive confirmation that a large amount of [169]Tm was present in the sample.
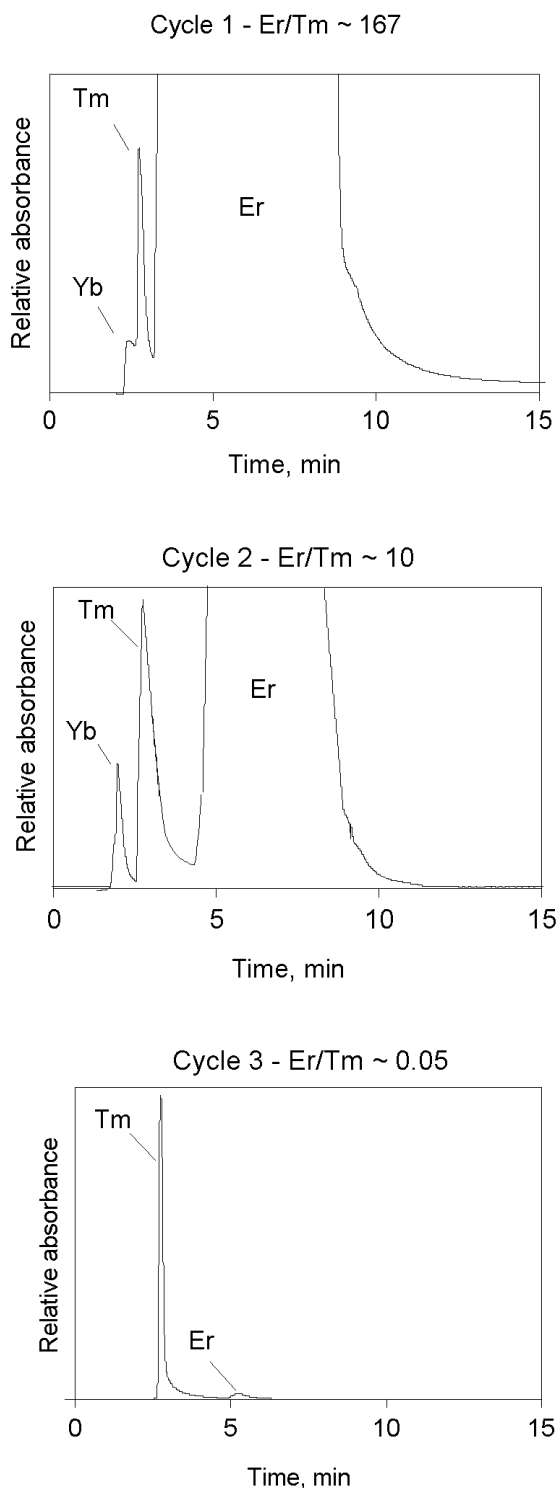
539

Cycle 1 - Er/Tm ~ 167

Cycle 2 - Er/Tm ~ 10

Cycle 3 - Er/Tm ~ 0.05

Fig. 8. Progress of Tm separation from excess Er using HPLC

The source of [169]Tm in our sample was most likely either originally present at less than one part in one thousand within the original enriched [170]Er parent material prior to irradiation or was produced during irradiation via neutron capture by a small amount (few percent) of contaminant [168]Er. Of these two scenarios, the former is most probable based upon the following argument.

The DANCE spectrum shown for the previous Tm target in Fig. 10 was produced from the identical parent material used to produce the target represented in Fig. 9. The main difference between these two samples is simply the level of radioactivity of [171]Tm present in the target. By our initial estimates, we expected roughly an order of magnitude more [171]Tm would be produced than what was actually received. Had we received this amount of [171]Tm, the level of contamination from stable [169]Tm would have been about 10%, which roughly corresponds to the level of contamination found in the older target based upon DANCE analysis of resonance peaks from Fig. 10. Since only a fraction of the expected [171]Tm was produced, the relative abundance of [169]Tm in the newest sample was therefore increased and found to be a major source of contamination.

Conclusions

Roughly 1-Ci of [171]Tm has been produced from neutron capture on ~250-mg stable enriched [170]Er for preparation of a nuclear target for DANCE. The target material has been separated from the parent material to an elemental purity of 95% with respect to Tm from an original mixture of 1:167 Tm:Er by scaling an existing rapid, high-purity HPLC method for lanthanides.[4] We found RTs, peak widths and SR were all significantly affected by scale-up. In addition, a unified method was devised for removing α-HIB and PAR from Tm fractions following HPLC separation from Er. DANCE measurements, nuclear counting and HPLC assays revealed the Tm fraction was contaminated with stable [169]Tm. The source of the [169]Tm was likely due to both a lower-than-expected irradiation time of the stable enriched [170]Er and the presence of one of several contaminants within this material including (1) a few percent of [168]Er producing [169]Tm during irradiation and/or (2) [169]Tm originally present at less than one part in one thousand. DANCE was instrumental in positively identifying the contaminant as [169]Tm.
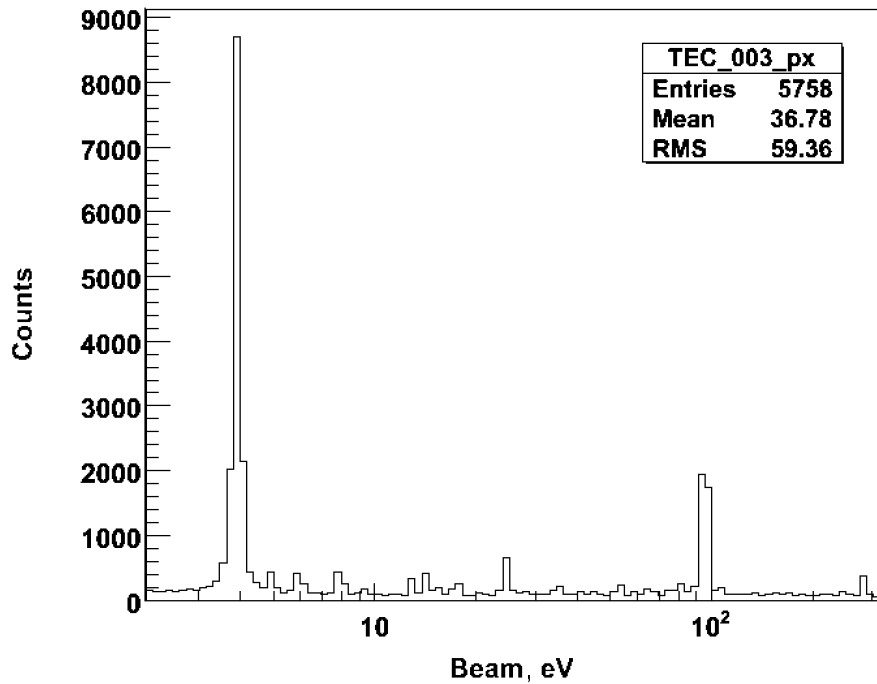
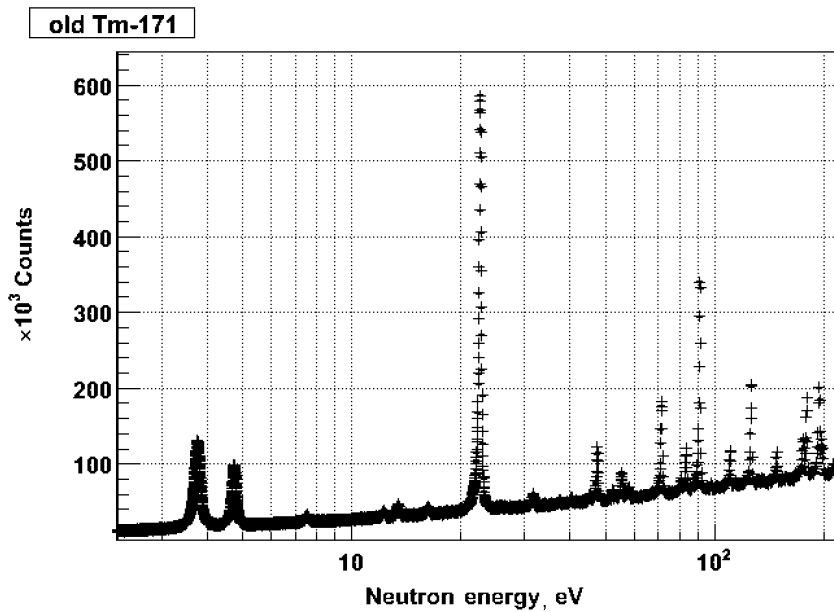*Fig. 9.* 2005 Esum versus neutron energy gated by multiplicity 3; DANCE spectrum for 20-μg [171]Tm target

Fig. 10. 1998 Esum versus neutron energy DANCE spectrum for 2-Ci [171]Tm target

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