

# Development of an Activated Thulium Source for the *in vivo* Measurement of Hg in Humans Using XRF

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## Résumé

Cette étude cible le développement d'une nouvelle source de rayonnement ( $^{170}\text{Tm}$ ) pour mesurer *in vivo* le niveau de Hg dans les reins par fluorescence X en vue d'évaluer l'exposition en milieu de travail intégrée dans le temps. Puisque les rayons  $E_\gamma$  du 84,25 keV émis dépassent légèrement la discontinuité d'absorption K du mercure à 83,10 keV, on s'attend à ce que la limite de détection du Hg s'améliore, comparative-ment à l'utilisation antérieure de 88,04 keV en rayons  $\gamma$  émis par  $^{109}\text{Cd}$ . L'activation d'une feuille de thulium a été effectuée au réacteur nucléaire McMaster par réaction  $^{169}\text{Tm}(n, \gamma) ^{170}\text{Tm}$ . Une réaction  $^{170}\text{Tm}$  émet aussi des rayons X K Er, Yb et Tm devant être filtrés par 2,69 mm de Cu afin de diminuer la dose du patient et le taux de comptage dans le détecteur HPGe. Les particules  $\beta$  énergétiques aussi émises par  $^{170}\text{Tm}$  ont été arrêtées par un porte-source en graphite spécialement conçu, permettant ainsi une émission minimale de rayonnement de freinage, ce qui peut augmenter le continuum d'arrière-plan dans le spectre. Le blocage de déchets radioactifs de cette source empêche aussi les fuites de rayonnements et permet la manipulation sécuritaire de  $^{170}\text{Tm}$ . Certaines simulations ont été effectués selon le code Monte Carlo (J. O'Meara) afin d'optimiser la collimation et de réduire la quantité minimale détectable de Hg. Le porte-source était contenu dans un collimateur W fait sur mesure, qui a été utilisé pour effectuer des expériences XRF sur des molécules fantômes de Hg sous format de rétrodiffusion. Au cours de l'activation des feuilles de thulium pures à 99,9 %, on a constaté que celles-ci contenaient du tantale à près de 0,4 %, un élément qui subit la capture neutronique pour former  $^{182}\text{Ta}$  et émet ensuite des rayons  $\gamma$  énergétiques, affectant sérieusement la détection de Hg. Nous sommes maintenant à la recherche d'une feuille de thulium de plus grande pureté pour remédier à ce problème.

## Abstract

This study is focused on the development of a new source of radiation, namely  $^{170}\text{Tm}$ , for *in vivo* measurement of Hg in kidneys by X-ray fluorescence to assess time-integrated exposure to that toxic metal especially in occupational settings. With  $^{170}\text{Tm}$ , the  $E_\gamma$  of 84.25 keV emitted is just above the K absorption edge of mercury at 83.10 keV, and hence it is expected there will be an improvement in the detection limit of Hg compared to the previously used 88.04 keV  $\gamma$  rays emitted by  $^{109}\text{Cd}$ . Activation of a thulium foil was carried out at the McMaster Nuclear Reactor using the  $^{169}\text{Tm}(n, \gamma) ^{170}\text{Tm}$  reaction.  $^{170}\text{Tm}$  also emits Er, Yb, and Tm K X-rays and these had to be filtered by 2.69 mm of Cu in order to decrease patient dose as well as the count rate in the HPGe detector. The energetic  $\beta$  particles also emitted by  $^{170}\text{Tm}$  were stopped by a specially designed graphite source holder so that there was minimal emission of bremsstrahlung radiation that could increase the background continuum in the spectrum. This source encapsulation also prevented radiation leakage and allowed  $^{170}\text{Tm}$  to be handled safely from a radiation protection perspective. Some simulations were run using the Monte Carlo code written by J. O'Meara (1998) so as to optimize the collimation and decrease the MDL of Hg. A W collimator was then custom-made to house the source holder and perform XRF

experiments on Hg phantoms in a backscatter geometry. Nevertheless during activation of the 99.9% pure thulium foil, the thulium foil was found to contain almost 0.4% tantalum, which undergoes neutron capture to form  $^{182}\text{Ta}$  that emits energetic  $\gamma$ -rays, thus seriously affecting the detection of Hg. Presently a thulium foil of higher purity is being sought in order to overcome this problem.

## Introduction

Mercury is a toxic heavy metal with no known benefit for human physiology. Occupational exposure is primarily due to elemental metallic mercury, which is a dense silvery liquid at room temperature and is highly volatile. Of the inhaled vapour, 80% diffuses to the blood and since it is lipid soluble, it is easily transferred to different organs such as the brain, liver, and cortex of the kidney. In the cells, the mercury vapour is converted to  $\text{Hg}^{++}$ , which is the toxic form of mercury and inhibits the biological activity of protein (Asano et al., 2000). After inhalation of mercury vapour, mercury is retained in the kidney (Kazantzis, 1970) and can even lead to renal failure (Zalups, 2000). The central nervous system is also affected and neurological as well as behavioural symptoms can occur at high mercury levels (WHO). Exposure to mercury vapour is usually determined by urine

## Interview with Bibi Najah Fajurally

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### What is the most valuable lesson you learned from attending the CNA conference and other, similar conferences?

The whole conference was an incredible experience for me and for everyone who attended it. I got a taste of all the different jobs in the nuclear field and met several people in the nuclear industry, which broadened my network. I can definitely say that the CNA and other similar conferences are a must for students who are graduating since many prospective employers can be found at them. These conferences also enable students to attend many interesting lectures given by knowledgeable and experienced persons and so remain updated on the latest news in the nuclear field.

Thanks for your time, Najah!

and blood samples but due to the rapid rate at which urine is eliminated from the body and blood is replaced, these methods of mercury analysis do not reliably indicate the total body burden. X-ray fluorescence is an alternative technique for biological monitoring since it is non-invasive and rapid and reflects chronic mercury exposure (Block & Shapiro, 1981). It also exposes the patient to a very small radiation dose and is a sensitive method of detecting low levels of mercury accumulated in the kidneys. A new source-excited XRF which consists of an activated thulium foil has been suggested by Grinyer (2008). The aim is to lower the minimum detection limit (MDL) of mercury and have an optimized system which can be routinely used to follow up with people occupationally exposed. With  $^{170}\text{Tm}$ , the 84.25 keV  $\gamma$ -ray emitted is just above the K absorption edge of mercury and so provides a favourable condition for increasing the yield of Hg K X-rays. Since  $^{169}\text{Tm}$  has a high thermal neutron cross-section of 105 barns, a reasonable length of time is required to produce a desired activity. Moreover,  $^{170}\text{Tm}$  has a fairly long half-life of 128.6 d and so it is a good source both for economic and calibration reasons. When its activity has been reduced to below the optimum level required for XRF, the same source can be reactivated again.  $^{170}\text{Tm}$  is better compared to  $^{109}\text{Cd}$  as the exciting source, as can be seen from the results of the Monte Carlo simulation using O'Meara code in figure 1. Moreover, the photoelectric absorption cross-section for  $^{170}\text{Tm}$  is  $7.80 \text{ cm}^2/\text{g}$ , and is thus larger than that of  $^{109}\text{Cd}$  at  $6.96 \text{ cm}^2/\text{g}$  (XCOM, n.d.). For  $^{109}\text{Cd}$  and  $^{170}\text{Tm}$  in a backscatter geometry, the Compton peaks are produced at 65.5 keV and 63.4 keV respectively. Hence there is a reduced Compton background underneath the Hg K X-rays in the latter case and the Compton peak is at an energy level well away from the Hg K X-rays.

## Experimental procedure

### Activation of the thulium foil

A thulium foil (99.9 % purity from Espi Metal) of 6 mm diameter and 0.10 mm thickness was placed in a high density polyethylene vial and sent through the RABBIT system of the McMaster Nuclear Reactor (MNR) to be irradiated. Using (2), the irradiation time,  $t$ , was calculated to be 9 min to obtain a  $^{170}\text{Tm}$  of 1.85 MBq.

The neutron activation equation is given by (Turner, 2007):

$$A = \frac{\theta m \phi N_A \sigma (1 - e^{-\lambda t})}{M} \quad (1)$$

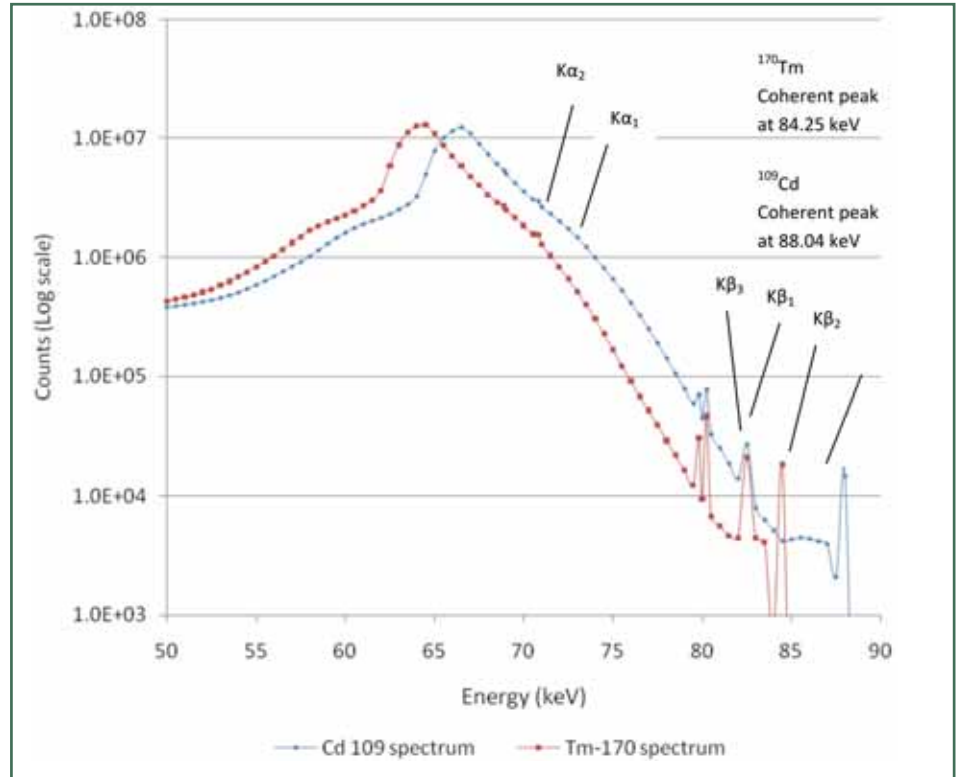


Figure 1: Difference between  $^{109}\text{Cd}$  and  $^{170}\text{Tm}$  spectra for the detection of Hg K X-rays

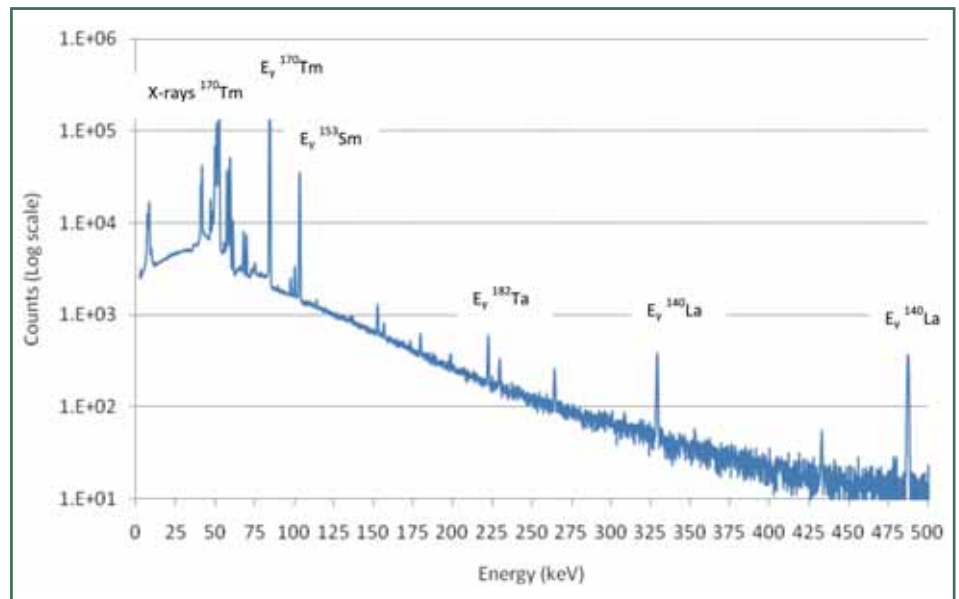


Figure 2: Thulium spectrum, with the major peaks identified

Where:

- $A$  is the activity of the Tm-170 obtained immediately after irradiation (in Bq)
- $\theta$  is the isotopic abundance of Tm-169 (100%)
- $m$  is the mass of the Tm-169 sample
- $\Phi$  is the neutron flux at the MNR ( $5 \times 10^{12} \text{ neutrons cm}^{-2} \text{ s}^{-1}$ )
- $N_A$  is Avogadro's number ( $6.02 \times 10^{23} \text{ atoms/mole}$ )
- $\sigma$  is the cross-section of the neutron capture reaction ( $105 \text{ E}^{-24} \text{ cm}^2$  for  $^{169}\text{Tm}$ )

- M is the atomic mass of Tm-169 (168.93 g)
- $\lambda = \ln 2 / T_{1/2}$  with  $T_{1/2} = 128.6$  days for Tm-170

Rearranging (1)

$$t = -\frac{1}{\lambda} \ln \left( 1 - \frac{AM}{\theta m \phi N_A \sigma} \right) \quad (2)$$

Afterwards, the sample was left to decay as there could have been formation of short-lived radioisotopes. The activated sample was then measured using a planar HPGe detector. An EG&G ORTEC 92X spectrum Master spectroscopy system was connected to the detector for data acquisition and the Maestro software was used to analyze the data. Figure 1 shows the spectrum acquired after thulium irradiation.

### Absolute determination of activity

It was important to determine the induced activity since the time of irradiation has been calculated by assuming a constant thermal neutron flux. Using (3), the activity, A, of the thulium was determined (Debertin, 1988).

$$A(Bq) = \frac{N_{net} C}{f_{\gamma} \epsilon_{abs} t_1} \quad (3)$$

Where:

- $\epsilon_{abs}$  is the absolute peak efficiency of the detector
- $N_{net}$  is the net counts under the photo-peak of 84.25 keV
- $t_1$  is the live time
- $f_{\gamma}$  is the emission probability of the  $E_{\gamma}$  emitted by Tm-170 (i.e., 2.5%)
- C is a correction factor due to self-absorption within the source and attenuation in the HPGe detector crystal and Be window. This was calculated to be 1.33.

The activity of Tm-170 was thus found to be  $(2.25 \pm 0.22)$  MBq.

### Quantification of $^{153}\text{Sm}$ and $^{182}\text{Ta}$ impurities in the thulium foil

From the spectrum shown in figure 2, it was found that the foil had impurities, identified to be  $^{153}\text{Sm}$ ,  $^{182}\text{Ta}$ , and  $^{140}\text{La}$ . The latter ( $T_{1/2} = 1.678$  d) is present as 180 ppm, from the certificate of analysis of the thulium foil.  $^{153}\text{Sm}$  and  $^{182}\text{Ta}$  were quantified by using the comparator method. Samples of samarium and tantalum atomic absorption standard solution and a thulium sample were irradiated simultaneously in the MNR. The standards

and thulium were then measured in the same source-detector distance configuration and for the same length of time.

The concentration of the sample,  $C_{\sigma\alpha\omega}$  is (Verma, 2007):

$$C_{sam} = C_{std} \frac{W_{std} A_{sam}}{W_{sam} A_{std}} \quad (4)$$

Where the subscripts 'std' and 'sam' stand for standards and sample respectively. W and A represent their weights and activities. The impurity levels of  $^{153}\text{Sm}$  and  $^{182}\text{Ta}$  in the thulium foil were calculated to be  $(88 \pm 8)$  ppm and  $(3832 \pm 230)$  ppm respectively.

### Characterization of the activated thulium foil

Figure 3 shows the unfiltered  $^{170}\text{Tm}$  spectrum after  $^{153}\text{Sm}$  and  $^{140}\text{La}$  decay, while figure 4 represents only the region containing the X-rays and  $E_{\gamma}$  of  $^{170}\text{Tm}$ . The labeled X-ray peaks are then explained in Table 1: K and L X-rays emitted by  $^{170}\text{Tm}$  and  $^{182}\text{Ta}$  (ESTAR, n.d.)

There was production of Er, Yb, and Tm K X-rays as the foil was thick enough to cause self-excitation of the source. Since  $^{170}\text{Tm}$  is also a beta emitter, it produces a continuous bremsstrahlung spectrum from very low energies up to  $E_{\mu\alpha\beta}$  of 968 keV, with the intensity being greater for the lower beta energies and decreasing up to  $E_{\mu\alpha\beta}$ . The beta radiation also

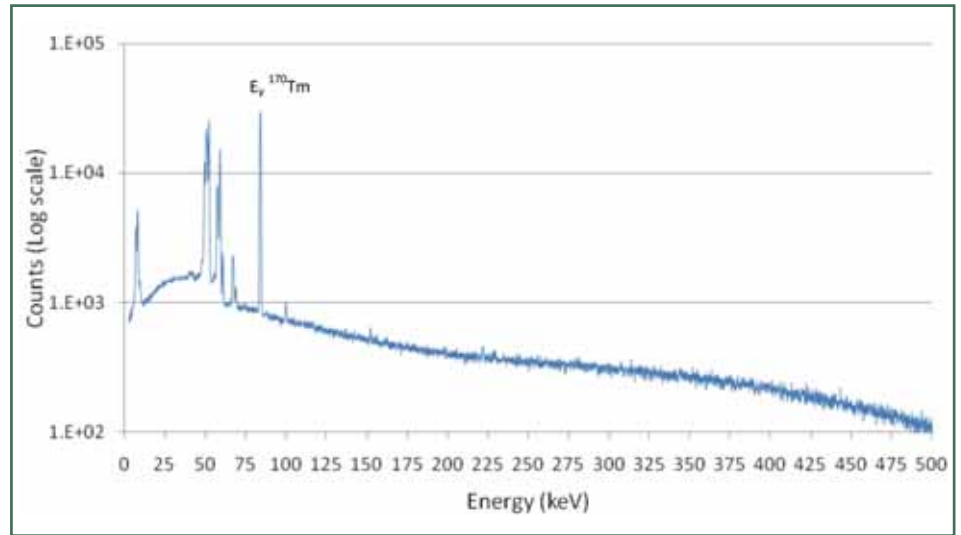


Figure 3: Unfiltered thulium spectrum

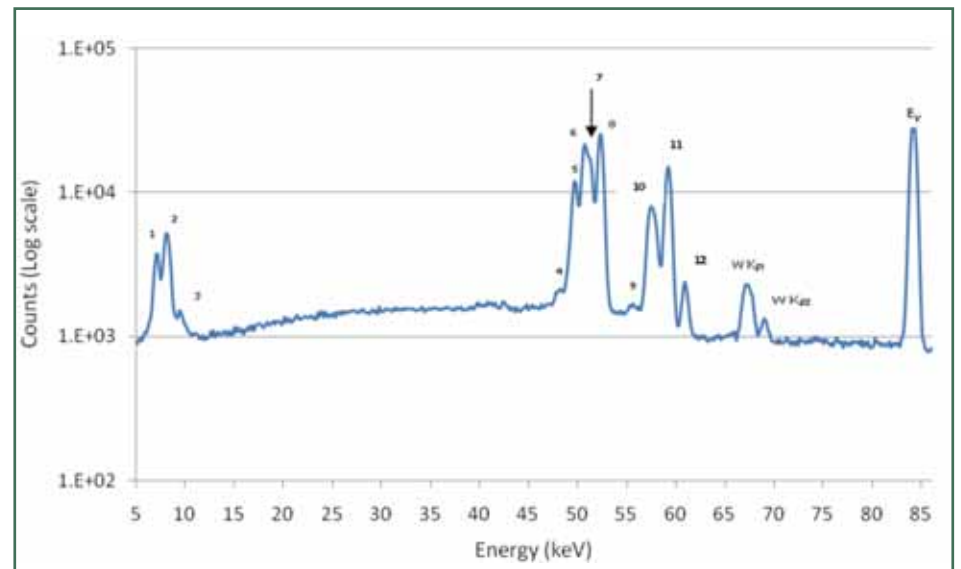


Figure 4: Spectral distribution of the K and L X-rays as well as the  $E_{\gamma}$  of  $^{170}\text{Tm}$

**Table 1:** *K and L X-rays emitted by <sup>170</sup>Tm and <sup>182</sup>Ta (ESTAR, n.d.)*

Peak no	Origin of peaks	Energy (keV)	Intensity per 100 k-shell vacancies (%)
1	<sup>170</sup> Yb L <sub>α1</sub>	7.42	8.30
2	<sup>170</sup> Yb L <sub>β1</sub> / W L <sub>α1</sub> (from Ta- <sup>182</sup> )	8.40 / 8.40	5.4 / 9.7
3	<sup>170</sup> Yb L <sub>γ1</sub> / W L <sub>β1</sub>	9.78 / 9.67	0.99 / 6.5
4	<sup>170</sup> Er K <sub>α2</sub>	48.22	27.0
5	<sup>170</sup> Tm K <sub>α2</sub> / Er K <sub>α1</sub>	49.77 / 49.13	27.2 / 47.5
6	<sup>170</sup> Tm K <sub>α1</sub>	50.74	47.4
7	<sup>170</sup> Yb K <sub>α2</sub>	51.35	27.2
8	<sup>170</sup> Yb K <sub>α1</sub>	52.39	47.2
9	<sup>170</sup> Er K <sub>β1</sub> / <sup>170</sup> Er K <sub>β3</sub>	55.67 / 55.48	9.77/5.06
10	<sup>170</sup> Tm K <sub>β1</sub> / Er K <sub>β2</sub> / W K <sub>α2</sub> (from Ta- <sup>182</sup> )	57.51 / 57.14 / 57.98	9.86 / 3.28 / 27.4
11	<sup>170</sup> Yb K <sub>β3</sub> / <sup>170</sup> Yb K <sub>β1</sub> / W K <sub>α1</sub> (from Ta- <sup>182</sup> )	59.16 / 59.38 / 59.32	5.18 / 9.99 / 47.0
12	<sup>170</sup> Yb K <sub>β2</sub>	60.96	3.38

**Table 2:** *Calculated thicknesses of materials to stop β<sub>max</sub> of <sup>170</sup>Tm*

β filter	Y	CSDA range (g/cm <sup>2</sup> )	Thickness from CSDA range (mm)
Graphite	3.22E-03	0.4797	2.82
Polyethylene	2.55E-03	0.4014	4.32

caused excitation of K X-rays of the tungsten collimator material as shown in figure 4.

The characteristics X-rays have to be filtered off to decrease patient dose and the number of incident photons interacting with the detector as these can lead to saturation. The energetic beta particles also need to be stopped so that there is minimal generation of bremsstrahlung radiation which can increase the background continuum in the spectrum. In turn, an optimum composite filter had to be chosen to remove the beta radiation and X-rays preferentially from the spectrum.

### Filtration of characteristics X-rays

Copper filters were used as they were readily available in the lab. The aim was to filter the X-rays without overly reducing the intensity of the 84.25 keV E<sub>γ</sub> so that the sensitivity of the system would not decrease. A good compromise was to calculate the Cu thickness so that the ratio of the transmitted intensity of E<sub>γ</sub> to that of the most intense K X-rays, i.e., <sup>170</sup>Yb K<sub>α1</sub> at E = 52.49 keV, from figure 4, was 50.

From the attenuation law for X and gamma rays:

$$T = \frac{I}{I_0} = e^{-(\mu/\rho)\rho x} \quad (5)$$

Where *T* is the transmission of the photons after going through the filter

- $\mu/\rho$  is the total mass attenuation with coherent scattering
- $\rho$  and *x* are the density (8.96 g/cm<sup>3</sup> for Cu) and thickness of the attenuator respectively.
- Let *T*<sub>1</sub> and *T*<sub>2</sub> be the transmitted intensity of the E<sub>γ</sub> to 52.49 keV X-ray respectively.

- Since  $\left(\frac{\mu}{\rho}\right)_{Cu}$  are 2.30 cm<sup>2</sup>/g and 0.674 cm<sup>2</sup>/g at 52.49 keV and 84.25 keV respectively,
- $T_1 = \exp-(2.30*8.96*x)$
- $T_2 = \exp-(0.674*8.96*x)$

Solving for *x* in the above equations, when the ratio of *T*<sub>1</sub> / *T*<sub>2</sub> is 50, gives a Cu thickness of 2.69 mm.

### Filtration of beta radiation

The β-ray filter should preferably be made of a low Z material because otherwise there would be production of significant bremsstrahlung radiation. For calculating the thickness of this shield, a conservative estimate was used whereby no self-absorption of the beta particles in the foil was considered. Hence the maximum β energy of 0.968 MeV emitted by <sup>170</sup>Tm was used. Table 2 gives the possible absorbers which were used, their radiation yield, Y, as well as their thicknesses, calculated using the CSDA range (ESTAR, n.d.).

### Design of source holder

In order to prevent leakage of radiation and excitation of W K X-rays with bremsstrahlung generated by β, it is of utmost importance to shield the source before it is placed in the tungsten collimator used in the XRF experiments. The source holder was made of either graphite or high-density polyethylene (HDPE) as shown in figure 5 so that all the energetic beta radiation from thulium could be stopped, with minimal emission of bremsstrahlung. The thickness of the wall and lid was 4.50 mm, chosen as a conservative estimate and to allow an extra margin for uncertainty. Once the foil was activated, it was allowed to decay under shield. Afterwards, it was quickly transferred to the basic container using long tweezers and the lid affixed. The procedure took place behind Perspex and lead shield to minimize β and γ dose to the personnel. Hence the basic container and the lid made up the entire source-holder assembly.

### Collimator design

The purpose of the W alloy collimator is to restrict the size of the radiation beam for exciting small regions within the body or water tanks such as the kidney or the phantom and so reduce the background. It is also used to prevent direct exposure of photons in the detector. Figure 6 illustrates the custom-made W collimator and its source holder, which were designed to contain the graphite or the HDPE housing the activated thulium foil. Using the Monte Carlo code written by O'Meara et al (1998), the optimized collimator length to detect Hg in a 1 cm depth kidney was found to be 3.5 mm. The collimator diameter was kept fixed at 6.0 mm. A copper disc of 20 mm diameter and 2.69 mm thickness was then glued at the exit of the collimator to filter off the X-rays emitted by <sup>170</sup>Tm.

### Testing XRF of Hg K X-rays using <sup>170</sup>Tm of 37 MBq

The <sup>170</sup>Tm source-excited Hg XRF system is given in figure 7. The spectrum shown in figure 8 was then acquired for a live time of 2000 s.

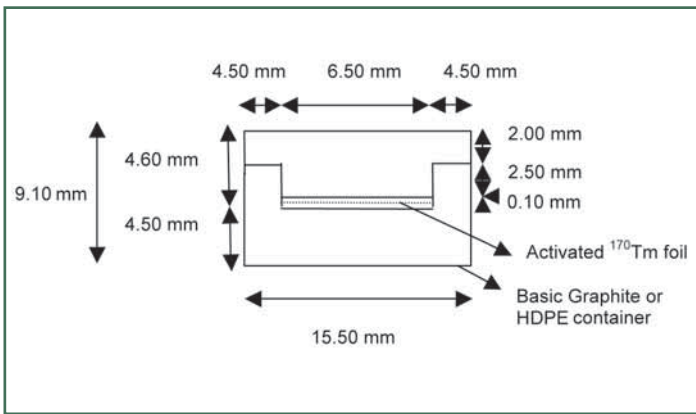


Figure 5: Thulium source holder

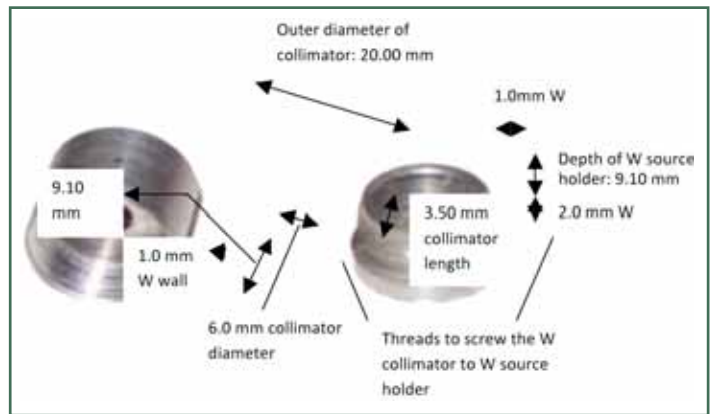


Figure 6: W collimator and its source holder

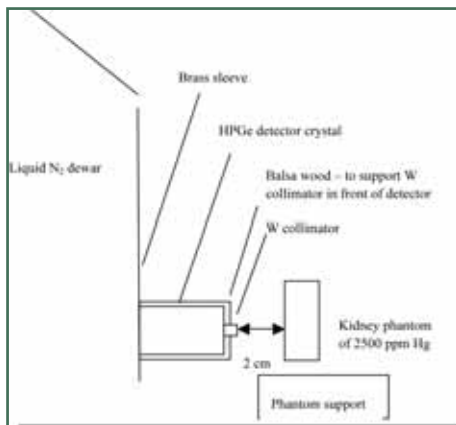


Figure 7: Experimental set up to verify XRF of Hg

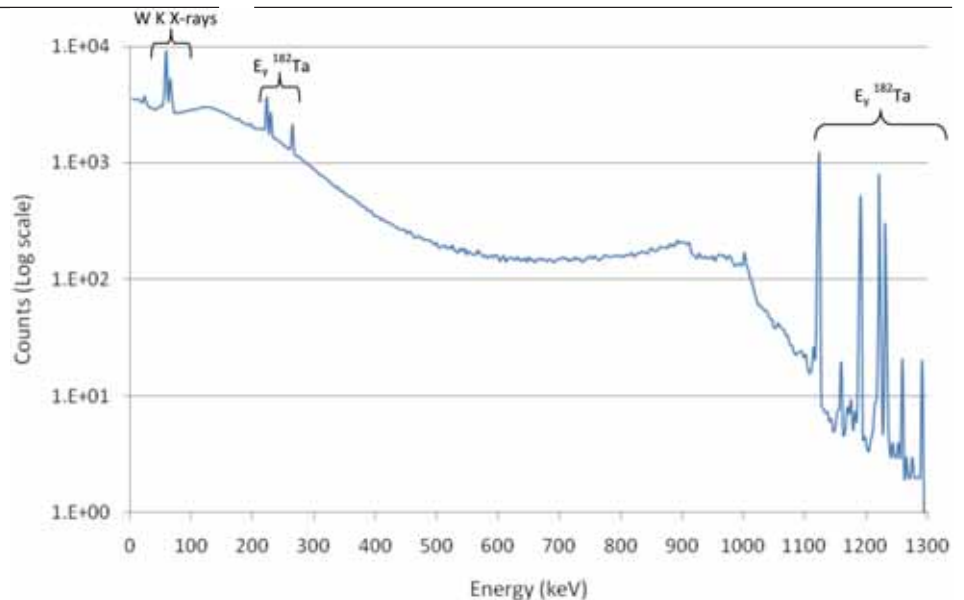


Figure 8: Spectrum showing high E<sub>γ</sub> from <sup>182</sup>Ta and W K X-rays

## Results and Discussion

There are several problems which can arise due to the presence of the impurities in the activated thulium foil, as given in section 2.2. Since <sup>140</sup>La and <sup>153</sup>Sm have short half-lives they can be allowed to decay under shield for at least 1 week.

On the other hand, <sup>182</sup>Ta has a half-life of 114.5 d, which is of the same order of magnitude as that of <sup>170</sup>Tm. Therefore, it is not worth waiting for it to decay as the <sup>170</sup>Tm will be decaying at the same rate. Moreover, <sup>182</sup>Ta emits intense gamma rays which will contribute unnecessary patient dose. The E<sub>γ</sub> of 100.11 keV (I<sub>γ</sub> = 14.10%) from <sup>182</sup>Ta can produce Compton scatter in a 180° geometry at an energy of:

$$E_{\gamma'} = \frac{E_{\gamma}}{1 + \frac{E_{\gamma}}{0.511} (1 - \cos\theta)} = 71.93 \text{ keV}$$

Hence the Compton scatter of the 100.1 keV line could be an important interference for detecting Hg K<sub>α1</sub> at 70.82 keV.

As shown in figure 8, there was generation of high intensity W K X-rays and <sup>182</sup>Ta gamma rays as well as a large bremsstrahlung

continuum which resulted in a considerable reduction of the Compton peak and meant the Hg K X-rays could not be identified. One potential reason for the high background continuum is that the intense γ-rays of <sup>182</sup>Ta (at 1121.3 keV – 34.9%, 1189.1 keV – 16.3%, 1221.4 keV – 27.0%, 1231.04 keV – 11.4%) are poorly attenuated by the 2 mm W collimator. Moreover, these energetic photons have caused the excitation of the W atoms, thus resulting in the production of high intensity W K X-rays.

## Conclusion and Future Research

Tantalum is usually present in thulium metals as Ta containers are used during the separation process of rare earth metals. Hence thulium of higher purity, such as 99.99%, and which has a lower Ta impurity has to be sought to overcome the above problems. Also the collimator should be made of a material whose characteristic X-rays are well separated from Hg K X-rays. Tungsten is not the ideal

candidate as it emits W K<sub>β2</sub> of 69.07 keV (I = 3.58 keV) and which is very close to the k<sub>α2</sub> of Hg. A good option is tantalum as it emits K X-rays not close to Hg K X-rays. As it has a density of 16.65 g/cm<sup>3</sup>, it will also be able to attenuate the photons incident on the detector. Eventually, for the purpose of the XRF experiment, a higher source activity of about 6 GBq would be required. This activity was chosen so that it would be almost the same as the 1 GBq <sup>109</sup>Cd source used previously in the XRF of Hg and consisting of the planar HPGe detector system (Grinyer, 2008).

## References

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# Bioassay sample above the internal investigation level at your facility

by **Emélie Lamothe**, Health Physics Specialist

Welcome back. I trust you had a nice relaxing summer. If your garden is like mine this year, your cornucopia runneth over. Canning and pickling will surely increase your sweating and liquid consumption. And what could be a better lead-in to last issue's question?

## Last Issue's Question

While reviewing bioassay sample results, you found a 70 µCi/L result, which is above the internal investigation level at your facility. What can you say about the tritium biological half-life for this worker?

Some data:

Day	Bioassay Result (MBq/L)
0	2.9E+06
1	2.3E+06
2	1.8E+06
3	1.4E+06
4	1.1E+06
5	9.1E+05
6	7.2E+05
7	5.7E+05
10	2.9E+05
14	1.1E+05
16	7.2E+04
20	2.8E+04
30	2.8E+03

## This issue's question

A CNSC inspector is coming to your site. Whose Thermo Luminescent Dosimeter (TLD) should the inspector wear?

Have fun! Remember, this column's for you. Send your answers and suggestions for future issues to the CRPA Secretariat or [eslamothe@hotmail.com](mailto:eslamothe@hotmail.com).

## Answer

Within a few hours of an uptake, tritium in the form of tritiated water (HTO) will be evenly distributed throughout the body's fluids and will be cleared from the body with the same turnover rate as water. A person's biological half-life for tritium will vary significantly due to variations in bodily excretion rates, environmental temperature and humidity levels, and fluid intake.

At any given time, the amount of HTO remaining in the body can be calculated as follows:

$$N(t) = N_0 e^{-\lambda t} \quad (1)$$

where

$N_0$  = initial amount taken into the body

$\lambda$  = the biological elimination rate (s<sup>-1</sup>), which can be calculated as

$$\lambda = \left( \frac{0.693}{t_{1/2}} \right) \quad (2)$$

and  $t_{1/2}$  is the biological half-life.

Working with equations (1) and (2), we can now calculate the biological half-life of HTO as follows:

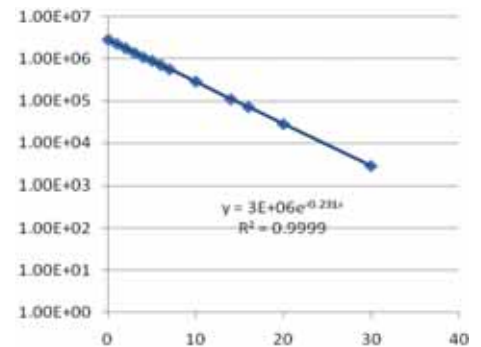
$$\frac{N(t)}{N_0} = e^{-\lambda t}$$

$$-\ln \left( \frac{N(t)}{N_0} \right) = \left( \frac{0.693}{t_{1/2}} \right) t$$

$$t_{1/2} = \left( \frac{(0.693 t)}{\ln \left( \frac{N(t)}{N_0} \right)} \right) \quad (3)$$

For the bioassay results given above, equation (3) gives a  $t_{1/2}$  of about 3 days, which is shorter than the typical 10 days for tritium.

Another way of determining the biological half-life is to plot the data on semi-log graph paper.



In this particular case, the time (x-axis) at which the original concentration (y-axis) has decreased by one-half is about 3 days. 🍀

## Activated Thulium Source

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