

Characterisation of an Historic Radioactive Article at Manchester University





Prepared for	The University of Manchester
Prepared by	Serco
Your Reference	AHH738630
Our Reference	TCS/RS/ENV/NP8483/TR01 Iss01
Classification	SERCO COMMERCIAL
November 2010	

Design | Advise | Integrate | Deliver



Title	Characterisation of an Historic Radioactive Article at Manchester University				
Prepared for	The University of Manchester				
Your Reference	AHH738630				
Our Reference	TCS/RS/ENV/NF	28483/TR01 Iss 01			
	To minimise our sustainable sour	impact on the environment, Serco uses pa ces	aper from		
Contact Details	Serco Thomson House Birchwood Park Risley Warrington Cheshire WA3 60 United Kingdom	GA			
	T +44 (0) 1925 2 F +44 (0) 1925 2 E technical.servio	52525 54571 ces@serco.com			
	www.serco.com/	tcs			
	Name	Signature	Date		
Author(s)	A Frith C McKay	OSE Walth	18/11/2010		
Reviewed by	N Reeves	Mars	19/11/2010		
Approved by	A Frith	AFE.	19/11/2010		



Executive Summary

The decommissioning of the refurbishment area in the Rutherford Building (formerly Coupland 1) at the University of Manchester led to the discovery of a radioactive article in the undercroft beneath the former Cohen Lecture Theatre. The article itself is a crudely cast lead block with two notable features; a small engineered cylindrical hole in one face which is plugged by wooden dowling and a large uneven area on another face which is thought to be either a casting imperfection or where a void within the block has been backfilled. A comparison of the calculated mass of a solid lead cuboid of the external dimensions of the block with the measured mass of the block indicates the presence of a void of volume 205 cm³. This void is not accounted for by the cylindrical hole, which even if it were to extend throughout the block, would have a volume of approximately 20 cm³.

Initial characterisation [1] has shown that the radioactivity associated with the block is due to radium-226, that the block is surface contaminated and has suggested that up to 30 mCi (1.11 GBq) of Ra-226 may be present within the block. Serco Radiation Services have been asked to undertake further characterisation for the purposes of disposal etc. and, given the significant difficulties in transporting this potentially high activity article to a suitable facility for dismantling, have proposed a non-destructive method. This method is based on gamma spectrometry with a mathematical efficiency calibration, which although it requires certain assumptions on the distribution of Ra-226 and the internal structure of the block, is fit for the purposes of source disposal.

From the characterisation measurements made in this study, it can be concluded that the lead block is surface contaminated with Ra-226 and its decay products at an order of 100 Bq/cm² alpha (based on the gamma spectrometry assessment of faces 3 and 4 (Section 3.3.2)) and contains 36 \pm 4 KBg Ra-226.

None of the measurements made in this study, including the segmented gamma scanning, give any indication that a discrete source of Ra-226 (e.g. a pellet or capsule), of the activity which has been suggested, is present within the body of the lead block. Indeed, the findings are not inconsistent with the activity being the result of a contaminated void within the block.

As such, it must be questioned whether there is sufficient justification for treating this article or its contents as a sealed or closed source, whether The Radioactive Substances (Waste Closed Sources) Exemption Order 1963 could be applied to its disposal, and whether the costs associated with such a disposal route can be avoided. Given that these measurements indicate that the activity of the block is 4 orders of magnitude lower than that previously suggested [1], the University may wish to consider dismantling the block, removing contamination as far as is reasonably practicable, and disposing of waste via the Universities existing Permit for aqueous waste and solid Very Low Level Waste (VLLW). The VLLW limits of 40 KBq per item and 400 KBq per 0.1 m³ can be expected to accommodate such a disposal.



Contents

1	Introduction	5
2	Method	6
	 2.1 Relevant background for radium-226 2.2 Selection of faces for measurement 2.3 Surface contamination measurements on selected faces 2.4 Segmented gamma scanning of faces 2.5 In-situ HRGS with mathematical efficiency calibration 	6 6 7 7
3	Results	9
	 3.1 Surface contamination assessment 3.2 Segmented gamma scanning 3.3 In-situ HRGS 3.4 Further Work 	9 10 11 17
4	Conclusions	18
5	References	18

serco

Introduction

The decommissioning of the refurbishment area in the Rutherford Building (formerly Coupland 1) at the University of Manchester led to the discovery of a radioactive article in the undercroft beneath the former Cohen Lecture Theatre. The article itself is a crudely cast lead block with two notable features; a small engineered cylindrical hole on "face 6", which is plugged by wooden dowling, and a large uneven area on "face 5" which is thought to be either a casting imperfection or where a void within the block has been backfilled.



Figure 1. Left: Face 6 with the cylindrical hole plugged by wooden dowling. Right: Faces 1, 4 and 5. Face 5 has an uneven "backfilled" area.

Since its discovery on 03 May 2005 the source has been held in the secure storage facility within the University's Radiation Services Unit pending characterisation. Initial characterisation [1] has shown that the radioactivity associated with the block is due to radium-226 and its decay products, that the block is surface contaminated and has postulated that the block may contain up to 30 mCi (1.11 GBq) of Ra-226. Serco Radiation Services have been asked to undertake further characterisation for the purposes of disposal etc. and, given the significant difficulties in transporting this potentially high activity article to a suitable facility for dismantling, have proposed a non-destructive method based on gamma spectrometry.

Given the uncertainty over the internal structure of the block and the distribution of Ra-226 and attenuating material within, the characterisation method proposed was based on a two stage assessment process, specifically:

- Stage 1 a segmented gamma scanning of selected faces of the block
- Stage 2 in-situ HRGS with a mathematical efficiency calibration

Given reports that the surface of this block was contaminated and that this could, depending on its significance, affect the above measurements, the two stage characterisation was preceded by an initial assessment of surface contamination.

This report presents the methods used, the results obtained and a discussion of the results. The reference numbers used by this report to identify individual faces of the block are those which have been marked on the block by others.



2 Method

2.1 Relevant background for radium-226

Although the primary radiation emitted from Ra-226 is alpha radiation, this is accompanied by the emission with a 32.8% probability of a characteristic 186-keV gamma photon. In relation to the assessment of Ra-226 contained within the lead block, the usefulness of the gamma emissions from Ra-226 is limited by the fact that this is the only gamma ray emitted by Ra-226 with a significant emission probability and that it is of relatively low energy. As the wall thickness of the lead block is significant but unknown, this mono-energetic gamma cannot be used to infer the wall thickness nor is it sufficiently penetrating to make measurements with high sensitivity.

However, the radioactive decay of Ra-226 results in a number of short-lived decay products two of which emit a number of gamma rays with moderate to high energy. These radionuclides are Pb-214 and Bi-214 and their most abundant gamma radiations are summarised in Table 1. Although these radionuclides are useful for differential attenuation correction to infer the wall thickness, care must be taken in using them to infer the activity of the parent Ra-226. If the Ra-226 was present within a gas tight vessel, the short-lived decay products would quickly reach equilibrium with their parent. However, that the immediate decay product of Ra-226 is a noble gas, radon-222, its retention within the lead block cannot be assumed.

Radionuclide	Gamma Energy (KeV)	Emission Probability
Pb-214	295	19.2%
	352	36.9%
Bi-214	609	46.9%
	1120	15.5%
	1765	16.2%

Table 1. A summary of the most abundant gamma ray emissions of Pb-214 and Bi-214.

2.2 Selection of faces for measurement

The count rate of a gamma detector passed at a constant distance over the surface of an extended article containing a gamma emitting radionuclide will vary with both the distribution of activity within the article and the distribution of attenuating material between the source and detector.

As the segmented gamma scanning work is concerned with determining the distribution of Ra-226 within the block, this should not be done on any surface of the block which has obvious features or flaws which would vary the attenuation. Similarly, the limited range of geometries that are available within the gamma spectrometry software package which will undertake the mathematical efficiency calibration makes it desirable to avoid surfaces with flaws or features.

As such, the large face 3 and its opposite face 4 were selected for measurement.

2.3 Surface contamination measurements on the selected faces

As the gamma spectrometry measurements would also be sensitive to surface contamination, the Serco characterisation work was preceded by a detailed surface contamination assessment on these two selected faces.

As direct measurement of beta / gamma surface contamination would be affected by the transmission of gamma radiation from activity within the block, the surface contamination assessment was purposely limited to alpha activity. This was undertaken using the alpha output of a dual phosphor surface contamination probe, a Thermo DP2/4A coupled to a Thermo Electra ratemeter / scaler timer. This system was demonstrated to be insensitive to beta / gamma



radiation, either from the surface or from within the block, by placing a sheet of paper between the block and the probe and observing the count rate reduction to zero.

In order to conduct a surface contamination survey with a spatial resolution appropriate to an article who longest side measures 15.7 cm, a paper mask was used to limit the measurement area to a 2 cm x 2 cm window. This mask was used to calibrate the Electra / DP2 with respect to a traceable Am-241 surface contamination source and the determined response factor of 0.98 counts per second per Bq/cm² was used to convert the count rate in each 4 cm² measurement area to Bq/cm².

This factor converts the alpha count rate to a gross activity (Bq/cm²) using an Am-241 alpha counting efficiency. In addition to alpha particles from Ra-226, the measurement will also include a contribution from its alpha emitting decay products.

2.4 Segmented gamma scanning of faces

An IRAS Depth Profiler[™] was modified to allow the lead block source to be placed on the carriage beneath a 2 mm collimating slit.

A low resolution gamma spectrometry system, based on a 2"x2" Nal(TI) scintillation detector coupled to an Ortec spectrometer system was energy calibrated, and a "region of interest" was marked around the Ra-226 photopeak and the most significant peaks from Pb-214 and Bi-214 (Table 1). The detector was placed within the collimated shield.

Initial scoping measurements were made for 10 seconds for each 2mm segment on faces 3 and 4, and on other faces as time permitted. These were to be followed by further measurements with a count time which would permit suitable statistical precision.

2.5 In-situ HRGS with mathematical efficiency calibration

2.5.1 Overview of measurement technique

The measurement technique employed in the assay of the lead block source was in-situ high resolution gamma spectrometry combined with point kernel numerical integration software which permitted a mathematical efficiency calibration.

The identification of gamma emitting radionuclides using an energy calibrated high resolution gamma spectrometer is straightforward, as the high resolution is generally sufficient to resolve interferences with the characteristic photopeaks of the radionuclide of interest. However, converting the count rate in the photopeak to an activity or activity concentration requires an efficiency calibration and a suitable background spectrum.

The determination of an efficiency calibration normally involves the preparation of a traceable measurement reference of the same geometry and materials as the source. For in-situ gamma spectrometry, where a large range of articles may be encountered, this is impracticable. Instead, a detector which has been characterised by the manufacturer and which has an efficiency calibration for a point source, is used with a point kernel numerical integration mathematical method to extrapolate the point source efficiency to one which takes account of the geometry and construction of the article being measured.

The initial method proposed was to use differential attenuation of the moderate to high energy gamma photons emitted by Pb-214 and Bi-214 to infer the wall thickness. This inferred thickness would then be used to measure Ra-226 directly, thus avoiding any disequilibrium issues between Ra-226 and its decay products. However, as discussed in Section 3.3.1, the low probability that the 186 KeV gamma from Ra-226 would penetrate the lead wall of the block combined with the lower than anticipated activity, meant that reliance had to be placed on inferring the Ra-226 activity from the activity of its decay products.



2.5.2 The measurement system

Using an Ortec GEM-FX8530P4 High Purity Germanium (HPGe) detector, of 55% relative efficiency, measurements of the gamma spectrum from faces 3 and 4 were acquired for approximately 2 hours on each face. The detector was located approximately 35cm from the source so as to minimise any geometrical effects of the source being a finite size rather than a point, whilst still maintaining a sufficiently high geometric efficiency. The measurement configuration is shown in Figure 2.



Figure 2. HPGe detector an Ra-226 Block source during measurement of face 4

The ambient background was accounted for by means of a 'peak based correction' method which utilised a 2 hour background measurement taken immediately after and in the same position as the sample measurements.

2.5.3 Energy and peak shape calibration

Detector energy and peak shape calibration were performed using a traceable 37KBq Eu-152 source, with peaks selected over the wide range of gamma ray energies associated with this radionuclide.

2.5.4 Point source efficiency calibration

The point source efficiency calibration is undertaken by the detector manufacturer (Ortec), using a traceable mixed radionuclide standard, and fit to a 6th order polynomial function of the natural logarithm of efficiency. This provides a reliable efficiency response calibration for photons over the energy range of 60kev to 1800keV.

The point source efficiency calibration of this characterised detector is used as an input for the software programme which mathematically extrapolates this efficiency to account for extended source material and geometry factors, and those of attenuate materials between the source and detector.

2.5.5 Development of mathematical calibration model.

For the purposes of mathematical modelling, it is assumed that a point of Ra-226 is located within a cavity at the centre of the lead block. Differential attenuation of the emitted photons depending on their energy allows, by comparison with the known relative emission probabilities of the gamma rays, an estimate of the thickness of the attenuating lead. In turn, once this is known, the activity of the individual radionuclides can be estimated.



3 Results

3.1 Surface contamination assessment

The measurement of surface contamination with alpha emitting radionuclides on faces 3 and 4 yielded a mean surface contamination of 13 ± 28 Bq cm⁻² (± 1 SD) with a maximum of 254 Bq cm⁻² averaged over 4 cm². The spatial distribution of this contamination is shown in Figure 3 and Figure 4 for faces 3 and 4 respectively. Using this data, the total alpha activity associated with faces 3 and 4 is 2.04 KBq and 2.90 KBq respectively. Both the spatial distribution and the total alpha activity are approximate on account of the face not being an integer multiple of the 4 cm² mask.

Figure 3The alpha surface contamination distribution over face 3 (Bq cm⁻²)

0.31	0.78	2.83	44.58	22.53	11.21	5.15	0.44
0.42	7.27	18.45	23.25	35.80	7.64	3.32	0.90
0.90	3.73	73.15	22.84	24.78	11.51	2.97	1.63
2.01	8.90	9.92	11.51	5.61	12.74	4.47	6.40
4.04	8.46	20.90	19.27	8.34	10.80	5.68	3.81
2.66	5.44	9.51	14.17	3.88	2.81	2.17	0.87





2.63	13.14	4.43	1.58	2.88	2.86	0.83	3.08
15.59	4.08	6.51	4.42	4.67	4.24	8.85	7.37
6.53	3.58	7.19	4.78	5.15	5.00	30.19	44.78
5.34	6.23	10.49	7.25	5.68	254.10	14.37	6.48
1.36	20.39	18.55	19.68	14.57	74.68	27.13	7.22
0.42	4.00	6.31	8.59	5.35	5.24	3.88	3.55

Figure 4The alpha surface contamination distribution over face 4 (Bq cm⁻²)



Any comparison between these measurements and those made by Dr Neil Todd, University of Manchester [1] during the initial characterisation work, must take into account that the Radiation Services Unit have undertaken some work to remove surface contamination since the initial study. Although the criterion which differentiates "fixed" and "non-fixed" surface contamination is ill-defined, we anticipate that the use of physical and chemical decontamination techniques has removed the majority of non-fixed surface contamination, and that what remains is reasonably well attached to the lead block. Of course, as the contents of the block are apparently leaking, non-fixed surface contamination may be replenished.

3.2 Segmented gamma scanning

Initial segmented gamma scanning with a short count time of 10 seconds and a slit width of 2 mm was undertaken on Faces 3 and 4. Gross photopeak count rates of between 0 and 0.6 cps were observed in the 609 KeV photopeak. No pattern was observed in these count rates across the face, nor would it be expected with the small number of counts recorded.

In order to obtain some data with higher statistical precision, a measurement was made on Face 3 for a period of 1000s at 40 mm, 80 mm and 120 mm, i.e. at a distance of quarter, half and three quarters the length of the side,. The resulting net peak counts are presented in Table 2.



		Net Peak Count Area (in 1000 s)				
Assessed location		Ra-226	Pb-214	Bi-214		
		(186 KeV)	(352 KeV)	(609 KeV)		
Face 3 – Lengthways	40 mm	47 ± 24	-8 ± 32	-20 ± 33		
	80 mm	-5 ± 26	-37 ± 35	58 ± 27		
	120 mm	-11 ± 27	-50 ± 34	48 ± 24		

Table 2 Net peak counts at several positions along Face 3.

Given the failure to detect a peak in most cases and, even when a positive net peak area has been reported, the large uncertainty associated with that measurement, it became apparent that the activity contained within the block was insufficient for its distribution to be mapped within reasonably practicable counting periods. As a consequence, the aim of trying to determine the distribution of Ra-226 within the lead block was abandoned, and the mathematical model used for the efficiency determination (Section 3.3) is left with an assumption of a point source at the centre of the block.

Notwithstanding this, this data does demonstrate the absence within the block of a localised source of Ra-226, with a relatively high activity such as initially suggested.

3.3 In-situ HRGS

3.3.1 Deep Activity

The background corrected peak areas observed for peaks associated with the gamma rays from Ra-226 and it's daughter products are presented in Table 3. As can be seen from Figure 5 the improved ability of the HPGe detector to distinguish peaks from the background, compared to the Nal detector, combined with the absence of the slit collimation, allowed the radium and its daughter products to be easily detected.

Nuclide	Peak Energy (keV)	Net Area Face 3 (counts)	Net Area Face 4 (counts)	Nuclide	Peak Energy (keV)	Net Area Face 3 (counts)	Net Area Face 4 (counts)
PB-210 ¹	10.8	0	0	BI-214	806.8	1316	170
PB-214 ¹	10.8	0	0	BI-214	934.24	2318	677
PB-210	46.32	1930	1659	BI-214	1120.28	7278	825 ^D
PB-214	53.2	480	427 ^D	BI-214	1154.9	1289	693
PB-214	74.81	8534 ^D	5442 ^D	BI-214	1238.42	2678	479
PB-214	77.11	6469 ^D	3869 ^D	BI-214	1281.67	356	-228
PB-214	87.2	2694 ^D	1400 ^D	BI-214	1377.81	1723 ^D	945
PB-214	89.8	197 ^D	-15 ^D	BI-214	1401.5	664 ^D	160 ^D
Po-214	100	2211	535 ^D	BI-214	1407.98	1081 ^D	244 ^D
RA-226	185.95	3168	2102	BI-214	1509.44	1194	248
PB-214	241.92	4898 ^D	1816 ^D	BI-214	1661.39	830	483
PB-214	295.42	12051	3580	BI-214	1729.02	1401	272
PB-214	352.06	22441	4580	BI-214	1764.21	5682	1223

Table 3.	Ra-226 Peaks observed during measurments
----------	--



Nuclide	Peak Energy (keV)	Net Area Face 3 (counts)	Net Area Face 4 (counts)	Nuclide	Peak Energy (keV)	Net Area Face 3 (counts)	Net Area Face 4 (counts)
Rn-222 AP	511.21	1139	68	BI-214	1846.73	971	328
BI-214	609.5	26849	5612	BI-214	2117.66	624	260
BI-214	665.45	862	-1593 ^D	BI-214	2202.79	1634	532
BI-214	768.49	3152	469	BI-214	2445.95	-322	288
PB-214	785.19	992	-138				

¹ gamma-ray below low level discriminator and so were not observed. ^D Peak overlaps with another peak, its area has been deconvoluted. ^{AP} includes an unquantified contribution from the 511 KeV annihilation peak.

As Bi-214 emits multiple gamma rays with energies ranging from 0.6 to 2.5 MeV, it is the ideal radionuclide to use for differential attenuation analysis. Using a least square regression fit, weighted by the uncertainty, the thickness of lead between the source and the detector has been estimated:

- Measurement of face three indicates an effective thickness of lead between the source and the detector of 0.76cm, with an estimated Bi-214 activity of 40.4(±2.1) kBq.
- Measurement of face four indicates an effective thickness of lead between the source and the detector of 1.76cm, with an estimated Bi-214 activity of 32.1(±1.9) kBq.

The deviation of the observed gamma ray relative intensities from the predicted relative intensities is shown in Figure 6 along with the effect of mathematically compensating for the lead attenuation.





Figure 5. Measured spectrum from face 3 with peaks corresponding to Ra-226 decay chain highlighted.





Figure 6. Deviation of relative peak intensities from theoretical prediction for (left) face 3 and (right) face 4.

It should be noted that the effective attenuation thicknesses of 0.76cm for face 3 and 1.76cm for face 4 implies the existence of a cavity 5.5cm in width within the 8-cm wide block. It would also imply that this cavity is not centrally located within the block.

With a known mass of 14.5kg and a density for lead of 11.7g/cm³ it can be inferred that a cavity of approximate 205cm³ (including the cylindrical hole) is present within the block. With a known width of 5.5cm the mean cross-section for the other dimensions should be 37cm².

As can be seen from Figure 7, the probability of a 186-keV Ra-226 gamma photon traversing the 0.76cm of lead is only 0.002%. This means that the Ra-226 within the block could not be measured directly, but could only be inferred from the measurement of daughter products which emit high energy gamma rays, such as Bi-214 and Pb-214.

The result of the activity calculations from gamma rays with energies in excess of 400keV are presented in Table 4. By assuming that Ra-226's daughter products are in secular equilibrium, then the Ra-226 activity is estimated to be 36 (± 4) kBq. This is the weighted mean of the Bi-214 activities reported in Table 4, which due to the multiple gamma rays from Bi-214 are the most reliable assessment.

Table 4. Measured nuclide activities with gamma rays >400keV

Nuclide	Face 3	Face 4
Bi-214	40.4(±2.1)kBq	32.1(±1.9)kBq
Pb-214	62.7(±9.2)kBq	<29.0kBq [†]

No activity detected, value reported is the minimum detectable activity for a 95% confidence level.

It should be noted that Rn-222 emits a 510-keV gamma ray, albeit with a low emission probability (0.076%), which is sufficiently penetrating to be observed. However this gamma ray could not be reliably separated from the 511-keV annihilation peak that was also observed.





Figure 7. Attenuation Coefficient for photons traversing 0.76cm of lead with a density of 11.7g.cm³. Photons less than 280keV have a negligible probability of penetrating 0.76cm of lead.

3.3.2 Surface Activity

As "low energy" gamma rays cannot pass out of the lead block, the fact that the 186-keV gamma ray and any other gamma rays observed with energy less than 280keV have been detected implies that they arise solely from surface contamination on the block. By assuming a uniform "active layer" of contamination on each face (as shown in Figure 8) the surface contamination has been calculated from the observation of gamma rays below 280keV and is presented in Table 5.



Figure 8. Conceptual model of surface contaminated lead block



Isotope	Face 3	Face 4
Optimum Active Layer Depth (mm)	0.1	0.0
Po-214	<7.1kBq [†]	<3.7kBq [†]
Pb-214	7.23(±0.26)kBq	2.12(±0.12)kBq
Ra-226	7.65(±0.83)kBq	4.82 (±1.3)kBq
Mean	7.26(±0.01)kBq [*]	2.14 (±0.06)kBq [*]

Table 5. Estimated surface activities by HRGS

[†] Measured activity was less than the minimum detectable activity for a 95% confidence level. ^{*} Weighted mean

If it is assumed that the daughter products of Ra-226 are in equilibrium (not necessarily a valid assumption) then the alpha activity would be five times that of Ra-226. This would give an estimate of 36kBq alpha activity on face 3 and 11kBq on face 4, which equates to an alpha surface contamination of approximately 200Bq/cm² and 61Bq/cm² for faces 3 and 4 respectively.

The surface contamination for both faces measured using the HRGS system is considerably higher than that measured by the direct surface contamination probes (2.04 KBq on face 3 and 2.90 KBq on face 4). However, this is not unexpected for the following reasons:

To provide an optimum fit between predicted and observed relative peak intensities for Pb-214 it was necessary to describe the "surface" contamination as an active layer with a depth of 0.1mm for face 3, and 0.0mm for face 4. This implies that, for face 3, the contamination remaining after cleaning is not on the "true" surface of the block, but distributed within a very thin layer. The attenuation of alpha particles within this layer would be significant and would contribute to the discrepancy between the direct surface contamination measurements and the HRGS measurements. The alpha contamination probe would only detect alpha radiation emitted from the surface of this layer and would not detect alpha radiation from the rear of this layer.

A further contributory factor to the discrepancy is likely to be the assumption of secular equilibrium. For secular equilibrium, the noble gas Rn-222, which is the immediate decay product of Ra-226, must be retained by the surface. Examination of the measured activity of Ra-226 (above Rn-222 in the decay chain) and Pb-214 (i.e. below Rn-222 in the decay chain (Figure 9)) in Table 5 is a useful indicator of the retention of Rn-222 and the degree to which the decay chain below Rn-222 is supported. If Rn-222 was completely retained, it would be expected that the activities of Ra-226 and Pb-214 would be equal, as is the case for face 3 (within uncertainties). The data suggests that on face 4, where the contamination appears closer to the surface, a portion of the Rn-222 is escaping, whilst on face 3 much of it is trapped within the 0.1 mm active layer. As a consequence, for face 4 at least, the alpha activity predicted by the HRGS measurement and the assumption of secular equilibrium will be an over-estimate.

In saying this, care should be taken when considering the accuracy of the depth of the active layer derived from the mathematical models used for the HRGS calibrations. For simplicity, the models assumed that the surface contamination was uniform across the surface, however the direct measurements have shown that this is not the case. Further refinement of the mathematical models could marginally improve the accuracy of the results, but the current models were judged to be sufficient for the purposes of regulatory characterisation.

In summary, the alpha surface contamination measured directly with a contamination monitor is likely to result in an underestimate where the activity is distributed more than a few microns of the surface. The HRGS measurement of Ra-226 itself is likely to be reliable; however, using this to estimate the alpha surface contamination using an assumption of secular equilibrium is likely to result in an over-estimate.



3.3.3 Other Radionuclides

There were no other gamma emitting radionuclides, other than those present within the Ra-226 decay chain, that were observed above the background. This indicates that the source if comprised solely from the Ra-226 decay chain.

3.4 Further Work

The HRGS measurements have shown that the surface activity is not a small fraction of the total activity in and on the lead block. For the measurement of the deep activity, there will be a contribution from the surface contamination, meaning that the quoted activity is likely to be an overestimate. More detailed modelling of the activity distribution could be performed to more fully take account of the surface contamination and by consideration of the distribution observed in the alpha activity measurements performed with the DP2 probe. The effort required to undertake this may be considered disproportionate for the disposal of a contaminated lead block.



Figure 9 Decay series from radium-226



4 Conclusions

From the characterisation measurements made in this study, it can be concluded that the lead block found in the undercroft to the Cohen Lecture Theatre is surface contaminated with Ra-226 and its decay products at an order of 100 Bq/cm² alpha (based on the gamma spectrometry assessment of faces 3 and 4 (Section 3.3.2)) and contains 36 ± 4 KBq Ra-226.

None of the measurements made in this study, including the segmented gamma scanning, give any indication that a discrete source of Ra-226 (e.g. a pellet or capsule), of the activity which has been suggested, is present within the body of the lead block. Indeed, the findings are not inconsistent with the activity being the result of a contaminated void within the block.

As such, it must be questioned whether there is sufficient justification for treating this article or its contents as a sealed or closed source, whether The Radioactive Substances (Waste Closed Sources) Exemption Order 1963 could be applied to its disposal, and whether the costs associated with such a disposal route can be avoided. Given that these measurements indicate that the activity of the block is 4 orders of magnitude lower than that previously suggested [1], the University may wish to consider dismantling the block, removing contamination as far as is reasonably practicable, and disposing of waste via the Universities existing Permit for aqueous waste and solid Very Low Level Waste (VLLW). The VLLW limits of 40 KBq per item and 400 KBq per 0.1 m³ can be expected to accommodate such a disposal.

5 References

1. Report on radiological measurements made on 16th June 2010 at the Radiation Safety Unit from the large source found in the old Schuster attic. N Todd, 28 June 2010.