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EXTERNAL RADIATION DOSE MEASUREMENTS FROM DEPLETED URANIUM

Introduction

Until now the laboratory has had only limited quantities of uranium on hand. Consequently, there has been little need to develop special instrument calibration and special handling procedures to measure and limit exposure. Now, with the anticipated arrival of large quantities of depleted uranium at the laboratory it is imperative that a program be developed to provide adequate safeguards for handling it. Such a program requires the capability of measuring exposure and contamination levels. The objective of the measurements reported in this note is to provide information which can be used in developing this capability.

Uranium Emissions

Uranium is a metal of density 19.05 g/cm^3 which is very active chemically, oxidizes easily in air and is pyrophoric in fine grain or dust form. The oxides, however, are relatively stable. Natural uranium is 99.3% ^{238}U, 0.7% ^{235}U and 0.006% ^{234}U.[1] Depleted uranium is the residue after much of the ^{235}U has been extracted by gaseous diffusion or other processes. However, both depleted and natural uranium are both over 99% ^{238}U. They will be considered identical for purposes of this report. One Curie of natural uranium is equivalent to 3000 kg (6615 lb) of natural uranium.[2]

Uranium contains traces of uranium-series radioactive daughters. A list of the uranium series and radiation emissions[3] is shown in Appendix A for easy reference. It is readily seen that uranium and its daughters provide a rich potpourri of α, β, γ emissions over a wide range of energies.

The primary contribution to dose at the surface is from the beta decay of 234 m Pa (E = 2.29 MeV). Alphas and low energy betas ($\sim$ 70 keV) do not penetrate the protective skin layers, .07 mm ($\sim$ 7 mg/cm^2) thick. A "rule of thumb" for beta ray surface dose rates with a 7 mg/cm^2 filter is 233 mrad/hr.[4] Our measured penetration gamma radiation dose rate near the surface is about 3 mr/hr (see below).

Contamination and Internal Exposure

Since the material is, by definition, evenly distributed, the contamination levels present depend primarily on the condition of the surface of the uranium. Unprotected surfaces have been shown to oxidize spontaneously in air. Badly oxidized surfaces yield considerable removable contamination (500 cpm α, 4000 cpm β in our tests). Surfaces prepared by galvanizing, acid washing, spray painting and electroplating were found to be free from removable contamination upon receipt. Preliminary tests indicate that cyclical heating to 60°C and cooling to -196°C does not visibly damage the surfaces. A small amount of contamination was produced in the liquid nitrogen bath. This activity (0.04 nCi) was detected on the inside of the dewar after evaporation of the cryogenic fluid. There was no significant removable activity detected on the plates after the testing.

The maximum permissible intakes (soluble form) are[5]

2.5 mg in one day by inhalation

150 mg in two days by ingestion

The maximum permissible body burden is 5 nCi

The most likely contaminants, however, would be (insoluble) uranium oxides for which the uptake from the GI tract is only 4% of soluble forms.

The maximum permissible concentrations in air (above natural background) of uranium and radon are:[2]

		Air Concentrations	
Nuclide	Solubility	Controlled area μCi/mℓ	Uncontrolled area μC/mℓ
U-natural	S	7×10^{-11}	3×10^{-12}
	I	6×10^{-11}	2×10^{-12}
Rn-222	S	1×10^{-7}	3×10^{-9}

Doses due to atmospheric radon are relatively small. For example, the average annual radon alpha dose to the lungs in the Chicago area where the average background concentration is about 300 pCi/m^3 (=0.3 pCi/ℓ = 3×10^{-10} μCi/mℓ) is 13 mrads.[1] It should be recognized that background radon concentrations vary considerably with time, location, weather, and the type and design of indoor enclosures.

Radon exposure from assembly of uranium components is expected to be low.[6] However, it is recommended that radon concentrations be monitored to verify this, at least initially, during major operations.

Uranium Sources

The uranium source material used for most of the measurements reported here was one of twenty-four 6 x 6 x 1 inch depleted uranium plates received from Argonne in December, 1974. Sometime previous to the measurements these plates had been coated with epoxy or varnish and wrapped in aluminum foil and vinyl plastic. However, we removed the plastic and foil before performing the measurements. A few measurements were also made using five 4 x 4 x 1/8 inch uranium plates coated with various materials obtained from Rocky Flats in January, 1984.

Common Methods of Measuring Uranium Emissions

Uranium can be detected with a variety of instruments. Any thin window ($\sim$ 7 mg/cm^2) GM tube or ion chamber is suitable. Accurate absorbed beta dose rate measurements, however are more complex. The use of a GM type instrument is generally not recommended. A thin window ion chamber is the instrument of choice. Such an instrument can be calibrated to a standard gamma source (Cs-137) and then used with a beta correction factor. A typical correction factor for a Cutie Pie type ion chamber used for beta dose measurements is 1.25.[7]

Another common method of calibrating an ion chamber is to take the surface beta dose reading from a uranium slab as a standard (233 mrad/hr on contact). An Elron or similar GM instrument calibrated to uranium betas may prove useful under limited conditions.

Dose Measurements Using Fermilab Instruments and Dosimeters

Preliminary measurements were made using a variety of survey instruments including the FNAL minimeter, Ludlum 14-C geiger counter, Eberline frisker, Thyac (both GM and scintillator probe) and Eberline PRM4B alpha meter. Two types of hand and foot monitors were tested as well. The dose rate meters and hand and foot monitors were all calibrated using Cs-137. The count rate instruments were pulser calibrated and source checked.

Two types of dosimeters supplied by Landauer were used for this test. The standard P1 type film badge comes in a multi-filtered holder which has an "open" window of 25 mg/cm², a plastic filter of 325 mg/cm², an aluminum filter of 375 mg/cm² and a lead/tin filter of 1660 mg/cm². The films are calibrated to Sr-90 for beta exposure and to Cs-137 for gamma exposure. LiF TLD rings were also used to estimate the probable extremity dose. The ring dosimeter is filtered only by the label, which is nominally 25 mg/cm², and the calibration is based on Cs-137.

The following tables summarize the results obtained during the preliminary measurements. These measurements were made at the surface of the 6 x 6 x 1 inch slab of uranium coated with epoxy.

Instrument	Surface Reading
Minimeter (pancake)	off-scale (> 50,000 cpm)
Minimeter (dose-rate)	6 mR/hr
Ludlum (T-probe)	8 mR/hr
Eberline (proportional ctr)	250 c/m (alphas)
Eberline (pancake)	off-scale (> 50,000 cpm)
Thyac (scintillator)	off-scale (> 800,000 cpm)
Thyac (GM-probe)	6 mR/hr with beta shield
Thyac (GM-probe)	60 mR/hr without beta shield
Elron (with absorber)	30 mR/hr
Elron (w/o absorber)	70 mR/hr
Cutie Pie	60 mR/hr

The Elron absorber was later determined to be 191 mg/cm^2.

The two types of hand and foot monitors available at the laboratory were checked for sensitivity at the 1 nCi level using a uranium-contaminated wipe as a source. The Technical Associates monitor did not detect the source, but the Fermilab-modified Wm. B. Johnson Associates monitor did detect it, both on the hand and the foot detectors.

Three P1 film badges and three U3 TLD rings were exposed on contact with the epoxy coated uranium surface for a period of 22.33 hr. The following results were returned by Landauer:

Films	Gamma	Beta*
1	140	2310
2	140	2190
3	140	2340
Average mrem/hr	6.27	102.1

*^{90}Sr equivalent

Rings	Total $\gamma + \beta$
1	2090
2	2070
3	2190
Average mrem/hr	94.8

Upon receipt of a shipment of 1/8" thick plates from Rocky Flats, surface measurements were made using an Elron (absorber removed). The surface reading varied from 23 to 45 mr/hr, depending on the preparation of the uranium surface. These plates are not expected to emit as much radiation as an infinitely thick slab.

Surface Preparation	Surface Readings
Zn-galvanized	23 mR/hr
Spray painted	31 mR/hr
Electroplated	45 mR/hr

The worst case preparation, electroplating, gives a 1 foot gamma plus beta reading of 2.0 - 2.5 mR/hr.

There is a discrepancy between the observed surface readings and the expected "rule of thumb" beta absorbed dose rate of 233 mrad/hr. This is primarily due to the strong dependence of the instrument response on beta energy and measurement geometry. The unknown thickness of the coating may contribute appreciably to the discrepancy as well.

Absorption Measurements for Uranium

The attached graph shows measured detector responses at the uranium surface as a function of absorber thickness for uranium. Some scatter in the data is to be expected because a mix of Al, Fe, and plastic absorbers were used in the measurements. In all measurements the absorbers were sandwiched between the surface of the 6 x 6 x 1 inch uranium and the detector.

Measurements were made with both a Cutie Pie ionization chamber and an Elron geiger counter (window absorber removed). The Cutie Pie beta component was corrected by the factor 1.25 (See earlier note). These instruments were previously calibrated with a ^{137}Cs (ε = 662 keV) gamma source. There is no reason why a GM Counter calibrated to ^{137}Cs should yield correct beta dose rate measurements. However, it turns out fortuitously that there is a rough correspondence between the actual beta dose rate and the dose rate readings made with a gamma-calibrated 1" diameter by 3" long GM tube.[8]

A close correspondence exists between the Elron and Cutie Pie responses to the beta component of the radiation, according to the data. The data indicate that betas from the uranium are essentially ranged out with about 800 mg/cm² plus the unknown epoxy thickness, so meter responses beyond this thickness can be attributed to X- and gamma radiation. Elron and other GM-type detectors as well at the P1 film dosimeter measured a contact gamma component of about 6 mr/hr. This is in contrast to the Cutie Pie which measures the X- and gamma radiation to be about 3 mr/hr. This discrepancy is attributed to the over-response of GM-type instruments and film (calibrated to ^{137}Cs) to low energy X-rays produced in the high - Z uranium material. The Cutie Pie measurement of 3 mr/hr is taken to be the more accurate of the measurements. This gamma component is shown as the dashed line in the figure.

The 6 x 6 x 1 in. uranium was also measured with the Elron & Cutie Pie through 1060 mg/cm² absorber at a distance of 1 ft.

	"Contact"	1 ft
Cutie Pie	3 mr/hr	.4 mr/hr
Elron	6 mr/hr	.6 mr/hr

Thus, the gamma component decreases by about a factor of 10 at a distance of 1 ft for the 6 x 6 x 1 in. uranium plate.

Tests of radiation absorption by several types of gloves were measured with the Elron. The results are listed below:

	Dose Rate (mr/hr)	Effective Thickness (mg/cm²)
without absorber	70+	-
leather, 1 layer	35	150
leather, 2 layers	20	270
rubber, 2 layers	45	80
cloth, 2 layers	49	60

The effective absorber thicknesses listed in the last column were read from the curve in the figure opposite the corresponding transmitted dose rate values. Clearly, gloves do offer significant protection from beta exposures.

One additional test was made in order to gain some idea of the radiation absorption properties of epoxy coatings. 6.25 mg/cm² Al foils were covered with one-, two-, and three coats of epoxy, and their thicknesses were determined by measuring mass and area. The coated foils were measured for absorption with an Elron.

Absorber	Thickness mg/cm² Total	Epoxy	Elron (incl. foils) mr/hr
2Al foils	12.52	--	70
2Al + 1 Epoxy	43.44	30.93	54
2Al + 2 Epoxy	62.83	50.32	48.5
2Al + 3 Epoxy	80.94	68.43	47
4Al + 6 Epoxy	174.69	149.68	28

The average thickness of one coat of epoxy is approximately 25 mg/cm². A layer of epoxy or varnish having a thickness in the range of these tests would have an appreciable effect on the dose rate and could account for much of the discrepancy between the expected 233 mr/hr and measured 70 mr/hr at the surface. Of course, one could not reliably extrapolate to "zero thickness" of epoxy even if the thickness were known.

SUMMARY

All instruments and dosimeters tested were capable of detecting radiation from uranium. Pancake instruments and the most sensitive hand and foot monitor can detect at the 1 nCi level. The Cutie Pie ionization chamber, with appropriate correction factors for betas, is the preferred instrument for measuring the dose rate of betas.

Epoxy-coated uranium was found to have deteriorated appreciably over a time span of several years. The resulting surface oxides pose a potentially serious contamination problem. However, only very minor contamination was found during tests of five uranium plates recently prepared and sent from Rocky Flats.

An absorption curve is provided which shows that all betas are absorbed by about 800 mg/cm^2 absorber. Gloves were found to reduce the dose rate appreciably.

The surface beta dose rate on bare uranium could not be determined because of the unknown thickness of the Epoxy coating. The measured surface gamma dose rate is about 3 mr/hr. It is recommended that a bare uranium calibration source be acquired for use as a standard for calibrating instruments used to measure beta dose rates.

ACKNOWLEDGEMENT

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REFERENCES

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2. DOE Order 5480.1, Attachment XI-1, p. 12.

3. Radiological Health Handbook, 1970 ed., p. 112.

4. Reference 3, p. 204.

5. ICRP Publication 6.

6. "Recommendations for the Safe Use of Uranium at CERN". CERN Safety Note No. 7.

7. Technical Specifications, 2590 Series Cutie Pie, Nuclear Chicago.

8. Reference 1, p. 236.

Uranium Series (4n + 2)*

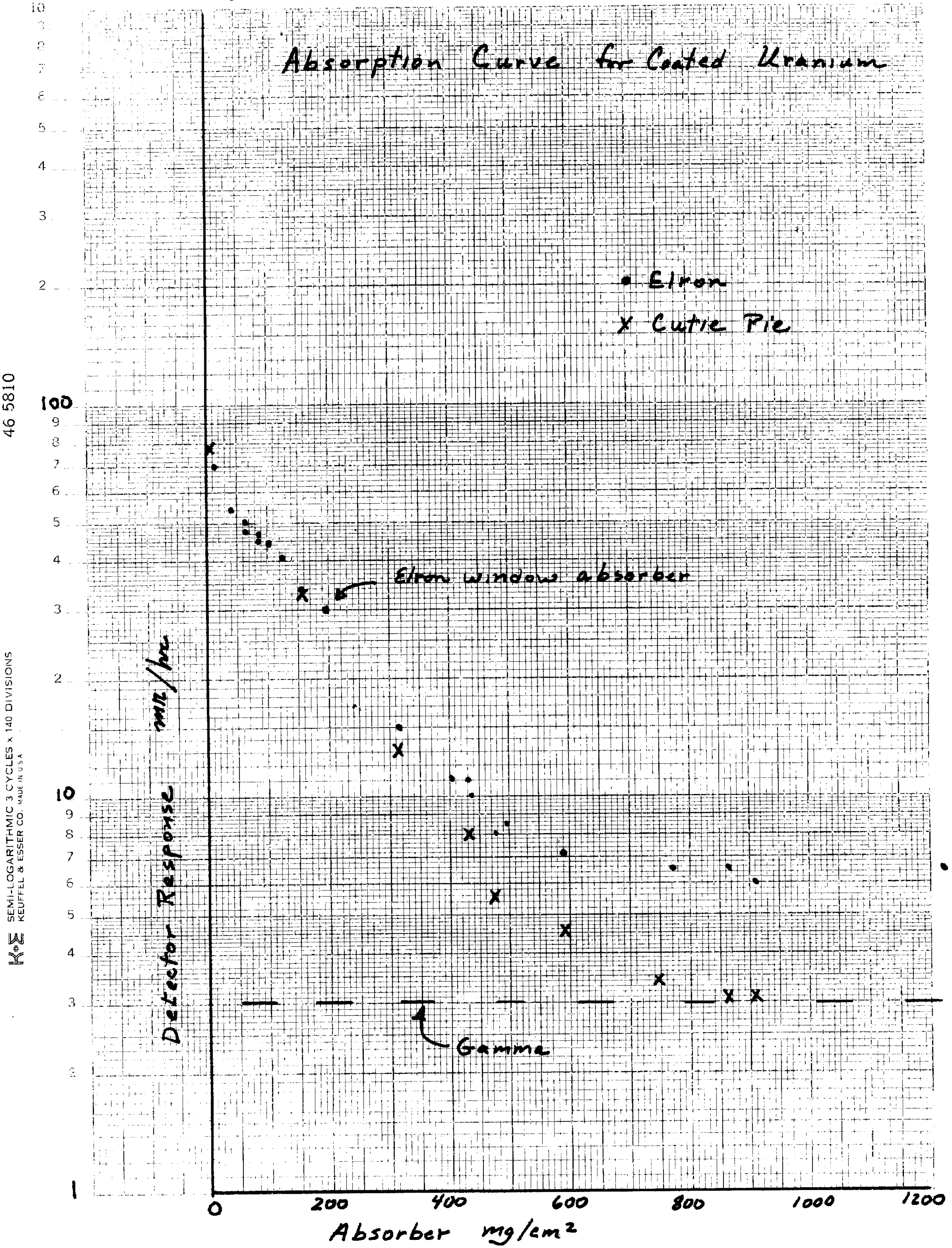
Nuclide	Historical name	Half-life	Major radiation energies (MeV) and intensities† α	β	γ
$^{238}_{92}U$	Uranium I	4.51×10^{9}y	4.15 (25%) 4.20 (75%)	---	---
$^{234}_{90}Th$	Uranium X$_1$	24.1d	---	0.103 (21%) 0.193 (79%)	0.063c‡ (3.5%) 0.093c (4%)
$^{234}_{91}Pa^{m}$	Uranium X$_2$	1.17m	---	2.29 (98%)	0.765 (0.30%) 1.001 (0.60%)
99.87% / 0.13%					
$^{234}_{91}Pa$	Uranium Z	6.75h	---	0.53 (66%) 1.13 (13%)	0.100 (50%) 0.70 (24%) 0.90 (70%)
$^{234}_{92}U$	Uranium II	2.47×10^{5}y	4.72 (28%) 4.77 (72%)	---	0.053 (0.2%)
$^{230}_{90}Th$	Ionium	8.0×10^{4}y	4.62 (24%) 4.68 (76%)	---	0.068 (0.6%) 0.142 (0.07%)
$^{226}_{88}Ra$	Radium	1602y	4.60 (6%) 4.78 (95%)	---	0.186 (4%)
$^{222}_{86}Rn$	Emanation Radon (Rn)	3.823d	5.49 (100%)	---	0.510 (0.07%)
$^{218}_{84}Po$	Radium A	3.05m	6.00 (~100%)	0.33 (~0.019%)	---
99.98% / 0.02%					
$^{214}_{82}Pb$	Radium B	26.8m	---	0.65 (50%) 0.71 (40%) 0.98 (6%)	0.295 (19%) 0.352 (36%)
$^{218}_{85}At$	Astatine	~2s	6.65 (6%) 6.70 (94%)	? (~0.1%)	---
$^{214}_{83}Bi$	Radium C	19.7m	5.45 (0.012%) 5.51 (0.008%)	1.0 (23%) 1.51 (40%) 3.26 (19%)	0.609 (47%) 1.120 (17%) 1.764 (17%)
99.98% / 0.02%					
$^{214}_{84}Po$	Radium C'	164μs	7.69 (100%)	---	0.799 (0.014%)
$^{210}_{81}Tl$	Radium C"	1.3m	---	1.3 (25%) 1.9 (56%) 2.3 (19%)	0.296 (80%) 0.795 (100%) 1.31 (21%)
$^{210}_{82}Pb$	Radium D	21y	3.72 (.000002%)	0.016 (85%) 0.061 (15%)	0.047 (4%)
$^{210}_{83}Bi$	Radium E	5.01d	4.65 (.00007%) 4.69 (.00005%)	1.161 (~100%)	---
~100% / .00013%					
$^{210}_{84}Po$	Radium F	138.4d	5.305 (100%)	---	0.803 (0.0011%)
$^{206}_{81}Tl$	Radium E"	4.19m	---	1.571 (100%)	---
$^{206}_{82}Pb$	Radium G	Stable	---	---	---

*This expression describes the mass number of any member in this series, where n is an integer.
Example: $^{206}_{82}Pb$ (4n + 2)......4(51) + 2 = 206

†Intensities refer to percentage of disintegrations of the nuclide itself, not to original parent of series.

‡Complex energy peak which would be incompletely resolved by instruments of moderately low resolving power such as scintillators.

Data taken from: Table of Isotopes and USNRDL-TR-802.


Absorption Curve for Coated Uranium
• Elron
X Cutie Pie
Elron window absorber
Gamma
Detector Response mR/hr
Absorber mg/cm²
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