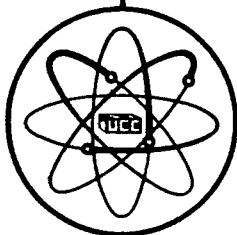Y-1280
Health & Safety

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RADIOACTIVITY OF THORIUM AND FEASIBILITY OF IN VIVO THORIUM MEASUREMENTS

R. E. Cofield

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THORIUM MEASUREMENTS

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Oak Ridge, Tennessee

November 11, 1959

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ABSTRACT

Tabulations of the radioactivity in the thorium series de-
cay chain and a discussion of the radioactivity variations
possible in non-equilibrium conditions of the decay chain
are presented. Experimental data indicate that the daugh-
ters of thorium within the human lung may be measured by
in vivo gamma spectrometry and related to Th^{232} content
for thorium quantities as small as 9 mg, if the activity ratio
of Th^{232}:Th^{228} is known for the deposited material. Two
crystal gamma coincidence measurements of pairs of the
simultaneously emitted gamma quanta from the daughters
are suggested as a promising approach to thorium in vivo
measurements.

INTRODUCTION

This report is a summary of the radioactive properties of thorium, the implications of variations in the decay chain equilibrium of thorium, and the feasibility of in vivo thorium measurements by gamma spectrometry. The material has been prepared as a guide for technical persons involved in processing of thorium or in the health physics aspects of thorium processing. Some of the evaluation has been taken from a feasibility study of in vivo uranium and thorium measurements supplemented by experimental measurements of the human gamma spectrum and of thorium samples in phantoms of the human body. The data regarding the radioactivity of the thorium decay chain have been summarized from standard tables of the isotopes and from actual searching of the scientific literature. The conclusions regarding the decay chain under non-equilibrium conditions have been made by application of standard texts on radioactive equilibrium, with particular reference to Friedlander and Kennedy.[4]

RADIOACTIVITY OF THORIUM AND DECAY DAUGHTERS

THE THORIUM SERIES

The term "thorium" usually refers to the isotope Th^{232}, having an isotopic abundance usually described as 100% of the Th isotopes. Chemically, the element is a metal of the Type 5 rare earth series; radioactively, Th^{232} is an alpha emitter with a half-life of 1.39×10^{10} years and, therefore, a specific activity of 246 alpha dpm (disintegrations per minute) per milligram.

This isotope is the parent of a heavy element, naturally radioactive decay series referred to as the thorium series or the 4n series. The designation "4n" arises from the fact that the masses (m) of all of the isotopes found in the series fit the formula:

$$m = 4n$$

where n is an integer.

This decay series is shown in Figure 1. It should be noted that the 4n series includes not only the parent Th^{232} but also another thorium isotope, Th^{228}.
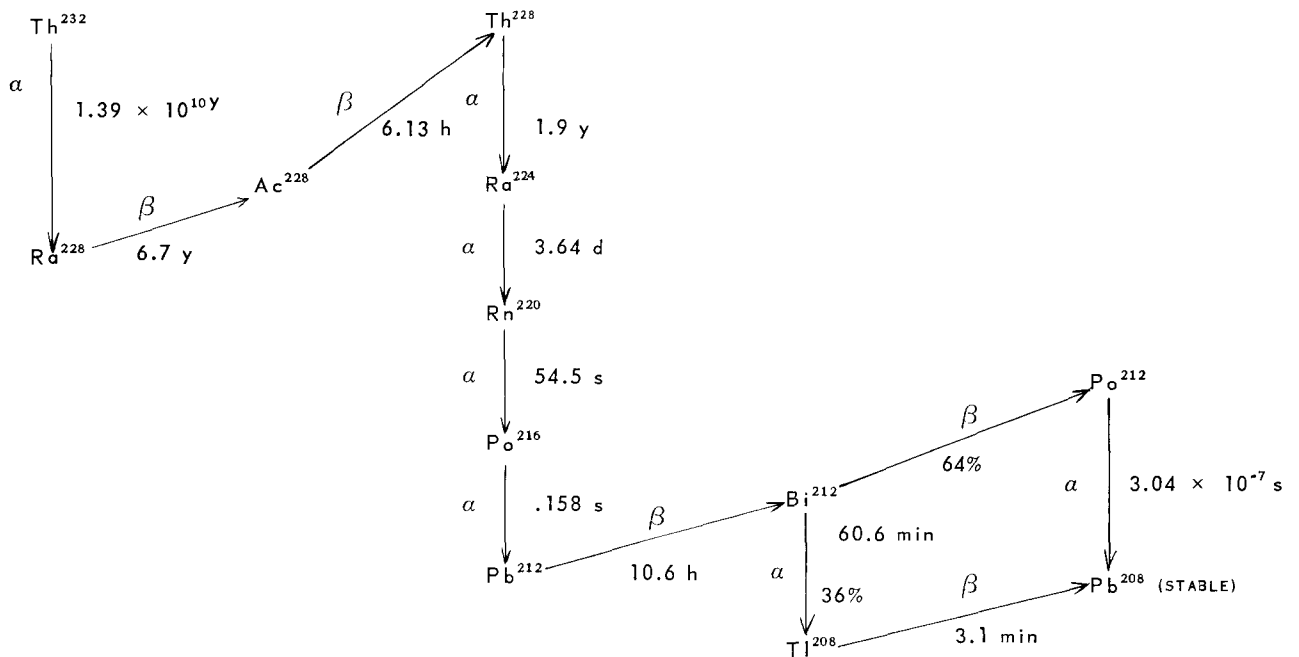

Figure 1. Decay scheme for Th^{232} -- thorium or 4n series

Other thorium isotopes exist – some from other natural decay series, some from the decay of artificially produced isotopes. Only two, Th^{230} and Th^{229}, have half-lives long enough to be of importance in process materials. The latter is the daughter of U^{233} in the artifically produced Np^{237} series $(4n+1)$ and would not be expected in natural materials. Th^{230} is the only long-lived daughter of naturally occurring isotopes, and traces are to be expected in thorium derived from uranium-carrying ore. If the original mineral contained 1% U, 1.5% of the thorium activity in the purified material would be contributed by the Th^{230} left by the uranium. Because of the small relative magnitude of this activity, no consideration of thorium isotopes outside the 4n series will be given.

In natural thorium minerals, all of the decay chain shown in Figure 1 will be present. Because of the long half-life of Th^{232}, its activity will be unchanged in any time period observed. The decay rate of Th^{232} determines the rate of formation of all of the succeeding daughters and, therefore, their concentration and activity. Since all of the daughters have much shorter half-lives than does Th^{232}, a secular equilibrium exists in which the number of disintegrations from each daughter is equal to the number of disintegrations of the Th^{232}. Table I lists all of the members of the 4n series, the half-life, and energy and abundance of radioactivity to be expected from the series. The abundances are given in relation to the Th^{232} alpha activity. The gamma abundances have been corrected for internal conversion within the atom to provide an actual indication of the number of gamma quanta that should be available for gamma spectrometry.

DECAY CHAIN NON-EQUILIBRIUM

The secular equilibrium described above will be disrupted by processing of the natural material to provide pure thorium. Two aspects of the matter require consideration: (1) the purified thorium, and (2) the separated daughters.

Purified Thorium

Although Th^{228} could not be separated from Th^{232} by any ordinary chemical or physical means, all of the daughter products not isotopic with thorium could be removed as chemical impurities. The ratio of Th^{232} and Th^{228} count rates would remain 1 for an instant. However, since the half-life of Th^{228} is much shorter than that of Th^{232}, the decay of Th^{228} would cause an appreciable reduction in the amount present; and the activity of Th^{228} would decrease with time, while that of Th^{232} would remain effectively constant. The daughters of Th^{228}, limited only by the 3.64 day half-life of Ra^{224}, would return to secular equilibrium with Th^{228} (as indicated in Figure 2) and would follow the Th^{228} activity, barring further chemical removal.

As the activity of Th^{228} was decreasing due to decay, it would also be in the process of being built up through the decay of Th^{232}. The total Th^{228} activity (and hence the activity of its daughters) would decrease for a while, since the rate of build-up (limited by the 6.7 year half-

Table I

Radioactivity of Thorium 232 Decay Series

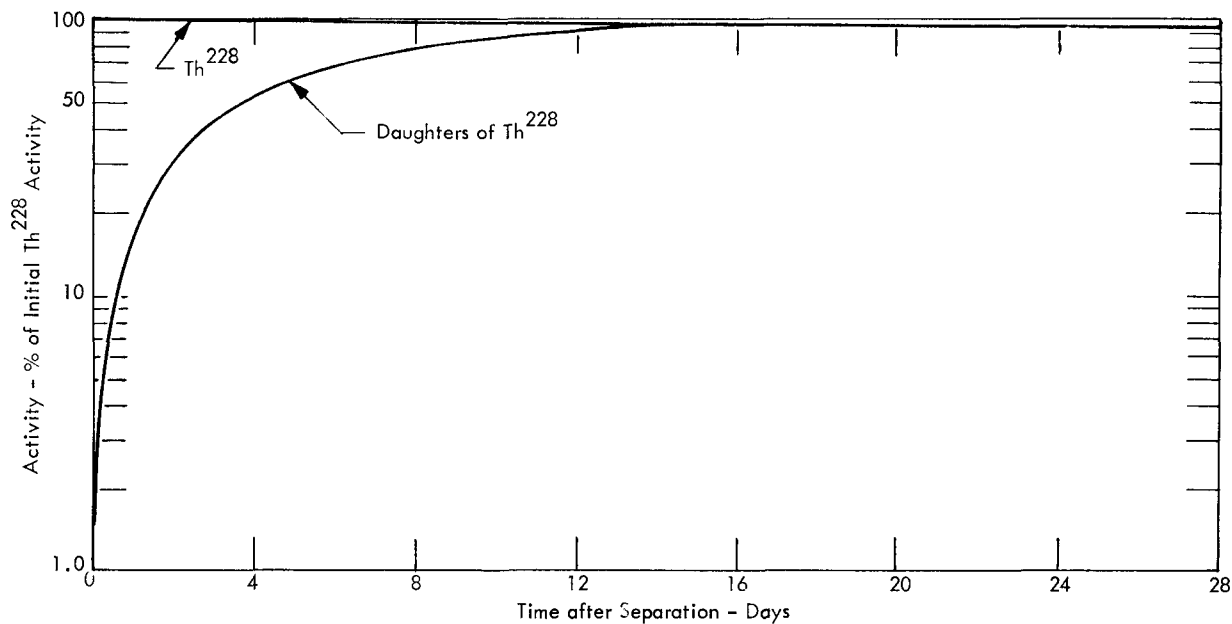| Isotope	Half-life	Alpha		Beta		Gamma	
		Energy (Mev)	Abundance $\alpha/Th^{232}\alpha$	Energy (Mev)	Abundance $\beta/Th^{232}\alpha$	Energy (Mev)	Abundance $\gamma/Th^{232}\alpha$
Th^{232}	1.39 x 10^{10} y	3.98	1.00	--	--	--	--
Ra^{228}	6.7 y	--	--	0.012	1.00	--	--
Ac^{228}	6.13 h	--	--	0.46	0.13	0.058	0.003
				0.66	0.08	0.129	0.034
				1.15	0.53	0.209	0.04
				1.72	0.07	0.270	0.03
				1.85	0.09	0.328	0.032
				2.18	0.10	0.338	0.092
						0.779	0.008
						0.790	0.045
						0.831	0.016
						0.908	0.25
						0.963	0.19
Th^{228}	1.9 y	5.338	0.28	--	--	0.084	0.02
		5.423	0.72				
Ra^{224}	3.64 d	5.194	0.004	--	--	0.241	0.046
		5.448	0.046				
		5.681	0.95				
Rn^{220}	54.5 s	6.282	1.00	--	--	--	--
Po^{216}	0.158 s	6.775	1.00	--	--	--	--
Pb^{212}	10.6 h	--	--	0.331	0.88	0.115	0.02
				0.569	0.12	0.176	0.01
						0.239	0.80
						0.300	0.05
Bi^{212}	60.6 m	5.603	0.0037	2.250	0.64	0.040	0.013
		5.622	0.0005			0.720	0.126
		5.765	0.0068			0.800	0.109
		6.047	0.238			1.03	0.037
		6.086	0.092			1.34	0.034
						1.61	0.047
						1.81	0.047
						2.20	0.018
Po^{212}	3.04 x 10^{-7} s	8.78	0.66	--	--	--	--
Tl^{208}	3.1 m	--	--	1.8	0.36	0.277	0.035
						0.510	0.087
						0.582	0.28
						0.859	0.05
						2.62	0.347

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Figure 2. Growth of Daughters in Th^{228}

life of Ra^{228} between Th^{232} and Th^{228} in the series) would be lower than the rate of decrease (determined by the 1.9 year half-life of Th^{228}).

When these two rates of change and their sums are calculated and plotted, the result may be illustrated as in Figure 3. This graphical presentation indicates that the total activity of Th^{228} in thorium freshly purified from the natural ore drops to a minimum of 56% of its equilibrium value after 4.8 years. The total activity then gradually rises until the initial equilibrium value is reached (97% of initial activity after 35 years, 100% at 67 years).

Of course, if by intention or chance, the material described by Figure 3 were subjected to chemical or physical processes that resulted in removal of the radium daughters from the series, the entire process would begin again - gradually increasing the Th^{232}:Th^{228} activity ratio, beyond that predicated by the single purification assumed in Figure 3. Theoretically, it should be possible to ultimately reduce the Th^{228} activity to an inconsequential fraction of the Th^{232} activity.

This behavior of the Th^{232} decay series is of considerable importance so far as radioactivity measurements are concerned. It indicates that unless a large amount of information about
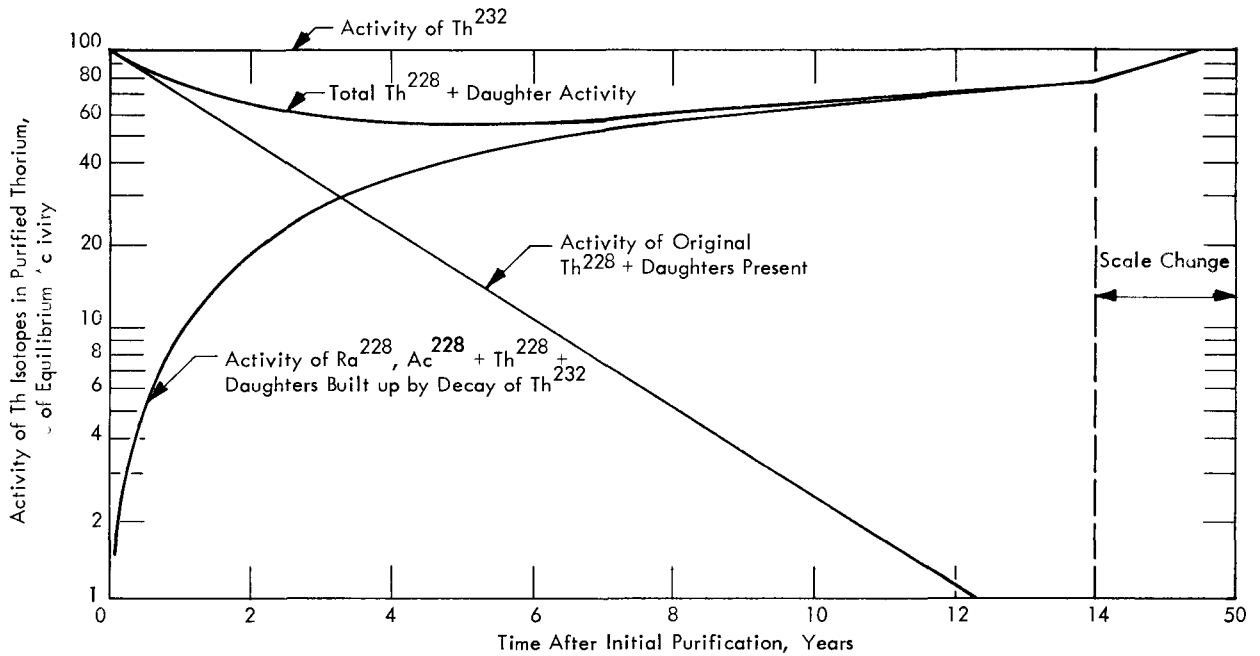
Figure 3. Variation of Activity in Natural Thorium After a Single Purification

the chemical and metallurgical history (and hence the conditions of the radioactive equilibrium) of the thorium in question is available, the concentration of Th^{232} cannot be measured by the activities of the daughters, even Th^{228}. The matter is of particular importance in any plan to measure Th^{232} body burden by in vivo gamma spectrometry, since Th^{232} itself does not have gamma rays suitable for such a measurement.

Separated Daughters

The daughters may be either completely separated from the parent thorium or concentrated in greater than equilibrium amounts in the presence of thorium. The latter might be expected to occur in some processing step, as an accumulation of the daughters in slag or in surface layers during casting of thorium. The behavior of the daughters free of thorium is simpler and will be considered first.

The decay scheme of Figure 1 indicates that the controlling half-lives are those of Ra^{228} and Ra^{224}. Separated from its Th^{228} parent, Ra^{224} would decay with a 3.64 day half-life. If all of the daughters were still present in the separated daughter system, their activities would follow that of the Ra^{224}. If absent, the daughters would quickly come into a transient equilibrium with the decaying Ra^{224}, controlled by the 10.6 hour half-life of Pb^{212}, as shown in Figure 4. The relative activities of this subseries would appear essentially unchanged from
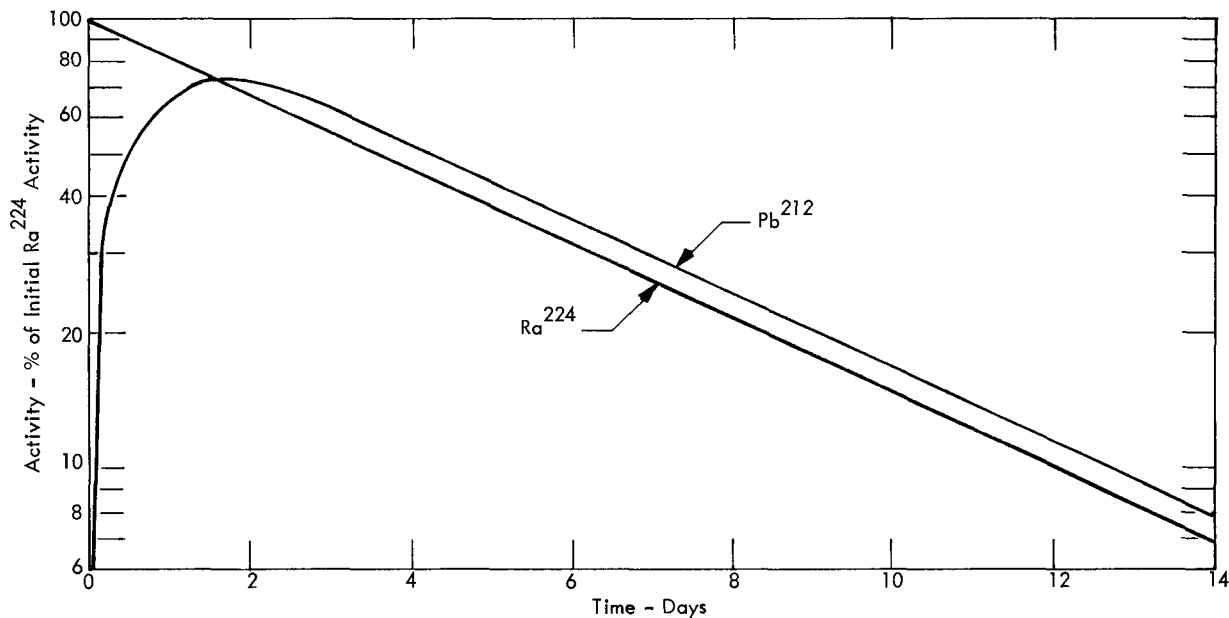
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Figure 4. Growth of Pb^{212} in Ra^{224} to Equilibrium

their ratios when the Th parents were present, and the separation would only be apparent because of the short half-life or by comparison with the activities of Th or the Ra^{228}.

The Ac^{228} will quickly come into equilibrium with its parent Ra^{228} and would follow that isotope in its decay characterized by 6.7 year half-life. This decay subseries would feed the Ra^{224} subseries described above; but the feeding has to be through Th^{228}, whose relatively long half-life (1.9 yr) limits its activity build-up and hence limits the production of new Ra^{224}. This approach to transient equilibrium between Ra^{228} and Th^{228} and its daughters is shown in Figure 5. It will be noted that 73 days (0.2 yr) would be required for the activity of Th^{228} and hence the Ra^{224} subseries to reach about 7% of the original equilibrium activity. In this period of time, the original Ra^{224} present and its daughters would be essentially all decayed, as indicated in Figure 4.

In summary, the activities of the daughters completely separated from the thorium parents would be as follows:

1. The original activity of Ra^{224} and its daughters would rapidly decay away with a half-life of 3.64 days.

2. The activity of Ra^{228} and Ac^{228} would decay with a characteristic 6.7 year half-life.
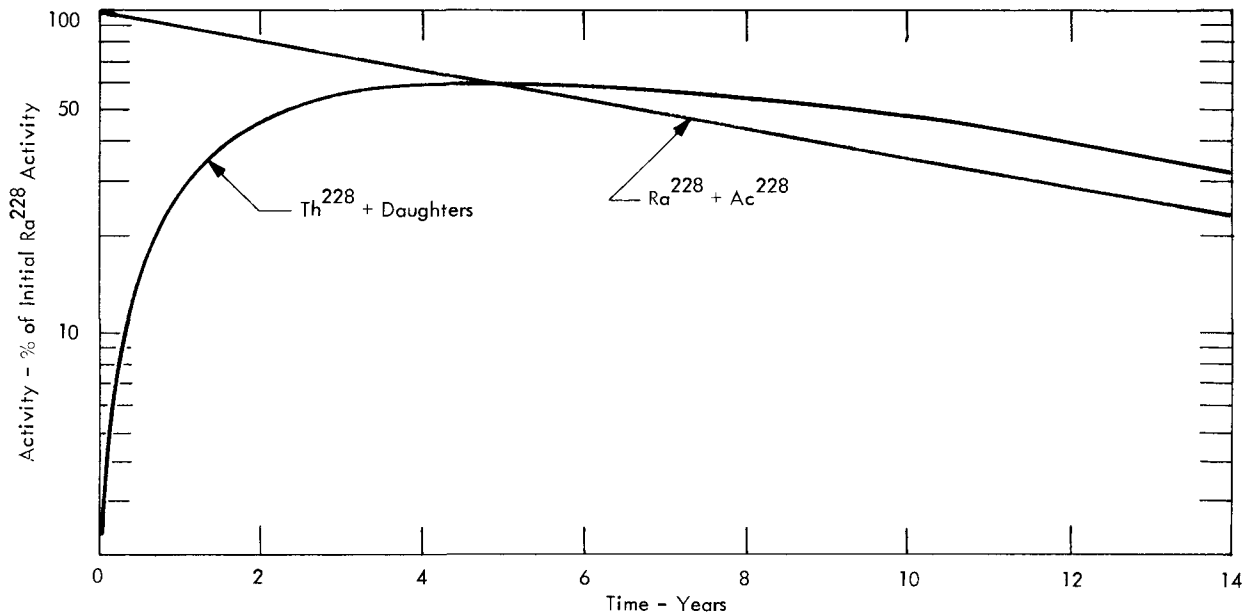
Figure 5. Growth of Th^{228} in Ra^{228} to Transient Equilibrium

3. New Th^{228}, Ra^{224}, and daughters would be created in the decay of Ra^{228}, but would not become significant until after the original Ra^{224} had essentially disappeared. These new activities would never be higher than about 60% of the same activities in the un-separated condition.

The discussion above assumes that no Th^{232} is present with the daughters. In the case of enrichment of the daughter activities in portions of the thorium to greater than equilibrium amounts, all of the daughters would be continually replenished by the thorium; and the decay of the daughter activities would only occur down to equilibrium amounts. Effectively, Figures 4 and 5 have to be considered as added to Figure 3, resulting in longer apparent half-lives for the daughters. As the daughter enrichment varied from complete separation to no enrichment, the apparent half-lives of the daughters would vary from those described by Figures 4 and 5 to the condition of no apparent change in daughter activity.

EXPERIMENTAL ALPHA AND GAMMA SPECTRA

The complexity of the decay scheme possibilities discussed above, and the large number of radiations indicated by Table I, suggest that separate measurements of radiation intensity at

each energy line or radiation spectrometry would be more meaningful than gross radiation counting. The second best alternative to radiation energy spectrometry would be separate alpha and beta counting of samples and comparison of the alpha-to-beta ratios.

The energy distribution of the alpha activities may be determined by counting the samples in the Frisch grid ionization chamber, which produces electrical pulses proportional in voltage to the energy of the alpha particle causing the ionization. The technique requires careful preparation of very thin samples in which essentially no collisions of the alpha particles occur. Such collisions of the alpha particles result in a "smearing" of the alpha lines, reducing the resolution of a line and its independence of other alpha lines. Figure 6 is the alpha spectrum of an electroplated thorium sample counted in a Frisch grid chamber operating with a 200 channel pulse height analyzer to sort and store the alpha energy distribution. The various alpha lines may be identified as those described in Table I, with the addition of the 4.6 Mev line, probably contributed by residual Th^{230} left in the thorium by the uranium originally present with the thorium ore. The relative heights of the Th^{232} and Th^{228} lines suggest that this sample has a $Th^{232}:Th^{228}$ activity ratio of 1.89. The activities of Rn^{220}, Bi^{212}, Po^{212}, and Po^{216} have the proper magnitudes relative to each other; but are slightly lower than that of the Th^{228}, probably because some of the Rn^{220}, an inert gas, is continually lost from the sample.
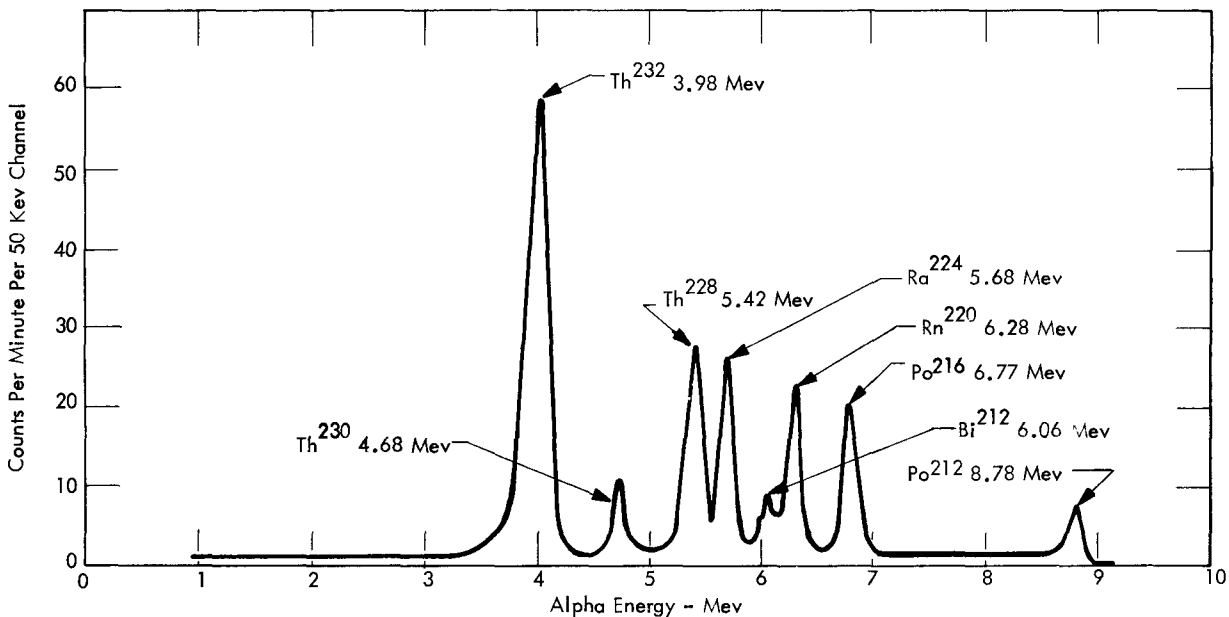

Figure 6. Alpha Spectrum of Thorium and its Daughters - Plated Sample

The gamma energy distribution may be best determined by NaI gamma spectrometry. Figure 7 is the gamma spectrum to 3 Mev of a thorium series sample taken with a 1-1/2" x 1" NaI (TI) detector and a multichannel pulse height analyzer. Energies of the photopeaks have been marked, and tentative assignment of the line to thorium series isotopes has been made on the basis of Table I. It should be noted that although less sample preparation is usually required for the gamma spectrometry than for alpha spectrometry, the gamma spectrum is more ambiguous than the alpha spectra because of the partial absorption of some gamma quanta (or Compton effect) in the small crystal and because of the interferences from the complex gamma emissions in the thorium series.

FEASIBILITY OF IN VIVO THORIUM MEASUREMENTS

EFFECTS OF DECAY CHAIN VARIATIONS

The discussion of the radioactivity and non-equilibrium possibilities of the thorium series has many implications regarding the feasibility of personnel monitoring of thorium process workers by measuring the thorium within the body.
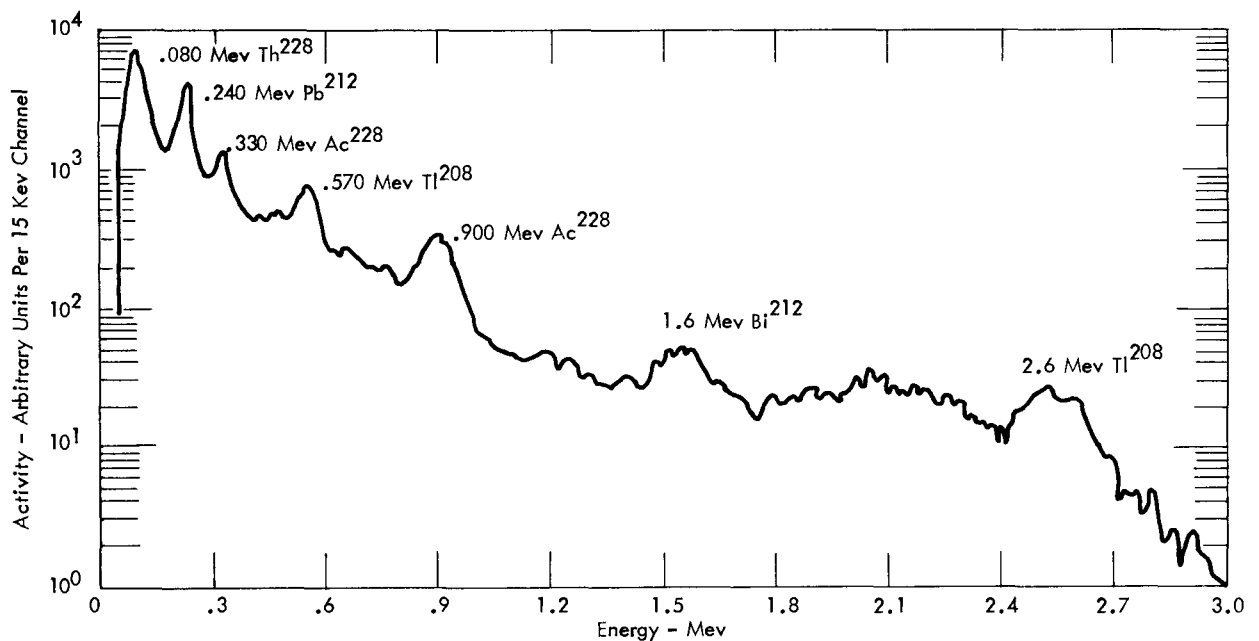

Figure 7. Gamma Spectrum of Thorium Series Sample, 0-3 Mev.

In the first place, such in vivo monitoring for any radioactive material must be dependent upon measurement of the electromagnetic radiation (essentially the gamma rays), since the alpha and beta particles do not emerge from the body wall. This fact means that such monitoring cannot measure Th^{232} itself, since the Th^{232} does not have gamma radiation.

The daughter series has several gamma rays of sufficient energy to pass out of the body with very little loss. To relate such measurements of the daughter gamma activity to Th^{232} content, knowledge of the activity ratios described by Figures 2, 3, and 4 is required. In a practical sense, so long as Th^{232} is considered the critical isotope, the activity ratio of Th^{232}: Th^{228} (as described by Figure 3) or variations thereof caused by repeated purification is the important one. This conclusion is based on Figure 2, which indicates that the daughters of Th^{228} which would be used for in vivo measurements are at 90% of the equilibrium ratio in 12 days after purification. Thus, even if thorium were taken into the body immediately after purification, rather good measurements could be made of Th^{228} after about 10 days by measuring the daughters.

For such an in vivo measurement, the 2.62 Mev gamma line of Tl^{208} would be quite suitable because its energy is above that of the natural body activities (Cs^{137} at 0.662 Mev and K^{40} at 1.45 Mev) which might interfere with less energetic gamma lines. Alternatives might be the 240 Kev activities from Pb^{212} and Ra^{224} or the 700 to 800 Kev activities primarily from Bi^{212}. The 900 Kev activity from Ac^{228} would be a measure of Ra^{228}, but Ra^{228} is not a suitable indicator of Th^{228} or Th^{232}. It must be emphasized that such measurements of the Th^{228} daughters could only be related to Th^{232} activity in the body through knowledge of Th^{232}:Th^{228} activity ratio, presumably by continual analysis of the process or environmental material to which persons are exposed.

A second complication is the possible fractionation, or separation from the thorium, of the daughter isotopes by the metabolic processes in the human body. The isotopes are different chemical entities and could be expected to be handled in different ways by the body if in a chemically tractable form. It is known that the radium daughters of <u>injected</u> thorium may be stored in the inorganic portion of the bone or may be excreted; while the thorium is deposited in the liver and spleen, or in the periosteum and endosteum of the bone. [1] However, it is to be expected that the greatest hazard in metallurgical processing of thorium would be inhalation of insoluble thorium metal or insoluble compounds such as oxide. The existence of thorium in such a physical and chemical state would be expected to minimize the removal of thorium daughters from the parent thorium and the consequent introduction of additional error of underestimating the Th^{232} content by measuring the daughters and applying too small a Th^{232}:Th^{228} ratio.

Third, the possibility of loss of some of the daughter activities from physical as well as chemical action must be recognized. One of the daughters, Rn^{220}, is noble gas and may be lost from the material in the body. This loss would probably depend upon particle size (i.e., the

ratio of volume to surface area) of the inhaled material. The very short half-life of Em^{220} should also help minimize this loss.

The inclusion of the effect of the 80 Kev activity of Th^{228} in estimating the in-excess-of-normal activity in the body would possibly reduce the error due to loss of Th^{228} daughters from thorium by those mechanisms described.

The measurement of the 900 Kev line from Ac^{228} would certainly be important if the inhaled materials were enriched in the radium daughter activities.

PROBABLE EFFECT OF THORIUM ON THE IN VIVO SPECTRUM

Experiments have been performed to demonstrate the possible effect of small amounts of thorium in the human lung on the normal in vivo gamma spectrum and to compare the probable minimum detectable amount of thorium with the maximum permissible body burden (MPBB) of insoluble thorium.

Most of the work has been done in relation to the statements of MPBB for insoluble thorium with the lung as the critical organ made in the 1953 ICRP recommendations [5] *and described in the in vivo uranium and thorium measurement feasibility study.* This MPBB of 0.02 µc amounts to 74 a dps of Th^{232} or 18 mg of thorium metal. The more recent recommendations of NCRP and ICRP indicate a lack of clarity as to whether natural thorium should be treated as a plutonium class hazard or as a natural uranium class hazard. [6,7] Thus, a new maximum permissible concentration in air is given as 4×10^{-12} µc/cc, a value about 13% of the 1953 ICRP value. A footnote comments on the uncertainty regarding thorium hazards and remarks that the old values are recommended, pending further evidence which may require the new, lower values. [7] It is recommended that exposures to natural thorium be kept as low as operationally possible until the situation is clarified. The new recommendations do not prescribe MPBB for insoluble inhalation hazards. [6,7] It is important to remember in reading the following discussion that the 18 mg of thorium discussed represents a uranium-type hazard and may be too large.

The data presented here have been obtained by determining the spectra of known amounts of thorium in the standard Masonite chest phantom used for calibration in the Y-12 In Vivo Radioactivity Measurement Facility. [2] The Th^{232}:Th^{228} activity ratio has not been measured for the sample material used, but the metal was typical turnings of thorium used for development projects in Y-12. The Th^{228} activity is almost certainly less than 70% of the Th^{232}, and quite possibly less than 50%.

Figure 8 shows a typical 0-1.6 Mev in vivo gamma spectrum of the human body in the chest position used in examinations for uranium. [2,3] To this spectrum has been added the spectrum of 18 mg of thorium metal in the standard chest phantom. Thus, it might be expected

that a person carrying a lung burden of 18 mg of thorium having the same parent: daughter relationship as the sample would have a gamma spectrum like the sum curve in Figure 8. It may be noted that there is an appreciable general elevation of all features of the spectrum, including the Cs^{137} and K^{40} photopeaks, which is not the case with uranium in the in vivo spectrum. Of particular interest is the small peak at 900 Kev, where no naturally present body activities occur. Unfortunately, this peak is probably due to Ac^{228}, signaling the presence of Ra^{228} rather than necessarily Th^{232}. In most cases, however, the appearance of this peak would be a qualitative indication of the presence of some thorium and would certainly serve to insure special attention to a subject yielding this spectrum.
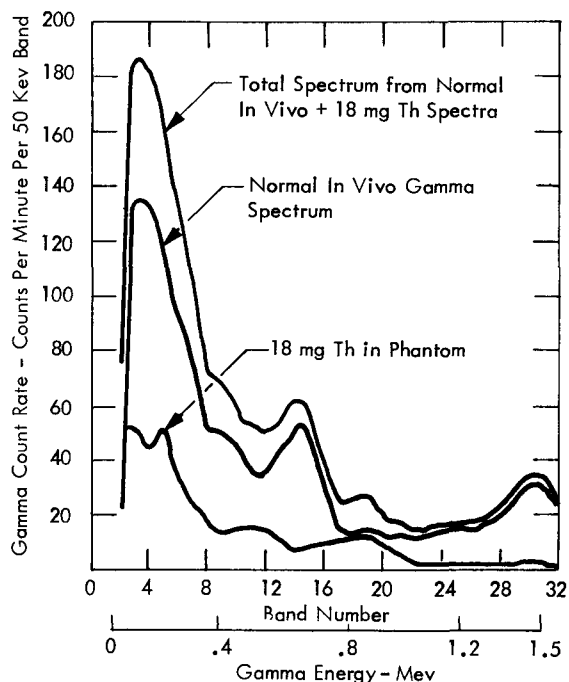

Figure 8. In Vivo Spectra With and Without Thorium

The effect on the parameters normally calculated in evaluating a possible uranium lung burden are tabulated from these spectra in Table II. It may be noted that although the thorium spectrum changes the activity level in the K^{40}, Cs^{137}, and M_{290} (265-315 Kev) regions used in the spectral prediction equations used for uranium,[3] the estimated normal N_{90} and N_{186} activities (90 and 186 Kev regions) are still less than the total measured spectral activity in these regions when thorium is present. Thus the application of the normal uranium measurement parameters would detect a difference in the spectra containing 18 mg of thorium. Applying the usual limits of error in the measurement and prediction of the N_{90} and N_{186} activities ($\pm$ 22 and $\pm$ 13.1 cpm, respectively), it is apparent that about 9 mg of this type of thorium would represent the limit of detection using the present spectral prediction equation for the normal activity in the 186 Kev region. Certainly in any actual program of measuring thorium in vivo, spectral prediction equations more directly suited to thorium would be developed. Figure 8 and Table II at least serve to indicate that the presence of 18 mg of Th of a typical type would certainly be detected even in a routine IVRM examination. An IVRM examination on employees before exposure to thorium hazards would be very helpful.

The spectra of Figure 8 cover the standard energy range taken in an IVRM examination for uranium lung burden. Figure 9 represents the same type of spectra taken for a larger energy range, 0 - 3.2 Mev. Here the effects of the higher energy gamma rays can be noted in the regions where the body has no normal activities of any consequence.

Table II

Spectral Evaluation Calculations Based on Simulations of
Thorium* and Uranium* in the In Vivo Spectrum

Parameter	Normal Spectrum (cpm)	Normal Spectrum + Th (cpm)	Normal Spectrum + Depleted U (cpm)	Normal Spectrum + Enriched U (cpm)	Normal Spectrum + Depleted U + Th (cpm)	Normal Spectrum + Enriched U + Th (cpm)
(1) Measured count rate, 65-115 Kev	134.6	186.5	286.9	169.5	338.8	221.4
(2) Measured count rate, 165-215 Kev	108.6	160.9	149.1	139.5	201.4	191.8
(3) Measured count rate, 265-315 Kev	74.1	99.1	87.2	74.2	112.2	99.2
(4) Measured count rate, 835-985 Kev	41.4	72.6	50.2	41.4	81.5	72.6
(5) Apparent Cs^{137} count rate, 585-735 Kev	149.3	173.0	158.3	149.3	182.0	173.0
(6) Apparent K^{40} count rate, 1.25-1.55 Mev	131.4	140.0	135.8	131.4	144.4	140.0
(7) Spectral slope in 186 Kev region	20.2	35.3	42.5	48.9	57.6	64.0
(8) Predicted count rate, 65-115 Kev	138.9	167.5	153.9	139.0	182.5	167.6
(9) Predicted count rate**, 165-215 Kev	113.9	136.1	125.4	113.9	148.4	136.7
(10) Excess count rate, 65-115 Kev	-4.3	19.0	133.0	30.5	156.3	53.8
(11) Excess count rate, 165-215 Kev	-5.3	24.8	23.7	23.6	53.0	55.1
(12) Excess count rate, 835-985 Kev	--	31.2	8.8	0.0	40.1	31.2
(13) Apparent Cs^{137} excess	--	23.7	9.0	0.0	32.7	23.7
(14) Apparent K^{40} excess	--	8.6	4.4	0.0	13.0	8.6
(15) Excess count rates ratio, 65-115/165-215 Kev	--	0.77	5.6	1.3	2.95	0.98

*In all cases, the spectrum of the amount considered as MPBB was added: 18 mg thorium, 0.275 mg enriched uranium, 49.6 mg depleted uranium.

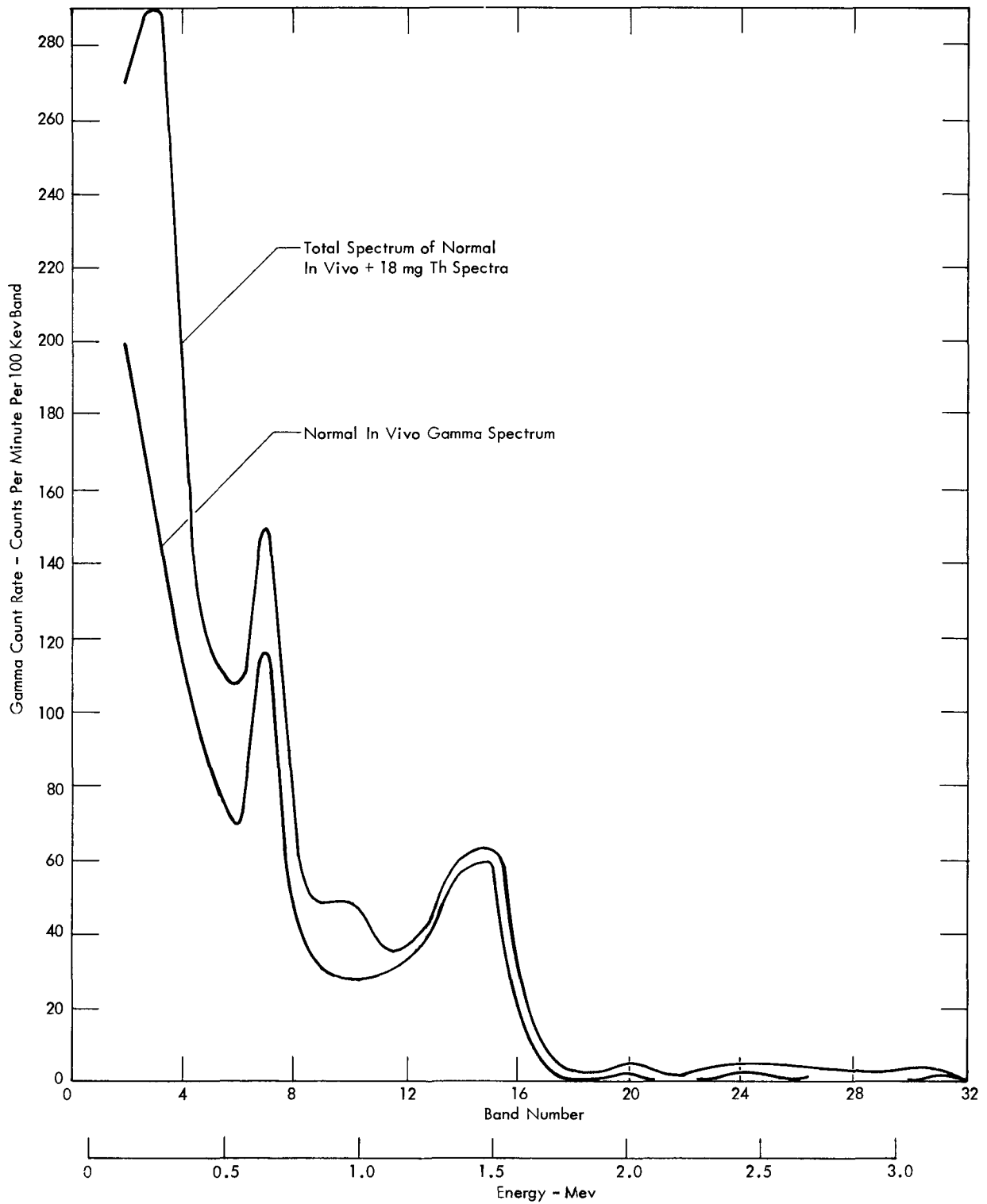**As calculated by spectral prediction equations, see Reference 3.

Figure 9. In Vivo Spectra, With and Without Thorium, to 3 Mev

Figures 8 and 9 are the 32 band spectra easily prepared from the 8 channel accumulations from the 256 channel pulse height analyzer. For the resolution presently obtained on the 9" diameter x 4" thick NaI crystal on the IVRM detector (using a single 5" diameter photomultiplier), such spectra are probably satisfactory. For thorium, gamma energy resolution will probably be more important than with uranium. Figure 10 reproduces the spectra of Figure 8 in a channel-by-channel spectral graph for better resolution. An additional and very promising approach, on which no experimental work has yet been done in Y-12, would be the use of two NaI detectors and the measurement of coincidences for some of the daughter products' gamma rays which are emitted simultaneously. Use of coincidence techniques, of course, serves very effectively to eliminate interferences from other isotopes and should reduce the minimum detectable amount in the body considerably. A good choice for a coincidence pair would be the 580 Kev and 2.6 Mev gamma lines of Tl208.

EFFECT OF URANIUM AND THORIUM TOGETHER IN THE IN VIVO SPECTRUM

Some interest has been expressed in the possibility of differentiating between thorium and uranium lung burdens in the human body. Figure 11 illustrates the spectra, made up by summing the proper normal in vivo and chest phantom calibration spectra, for the following conditions.
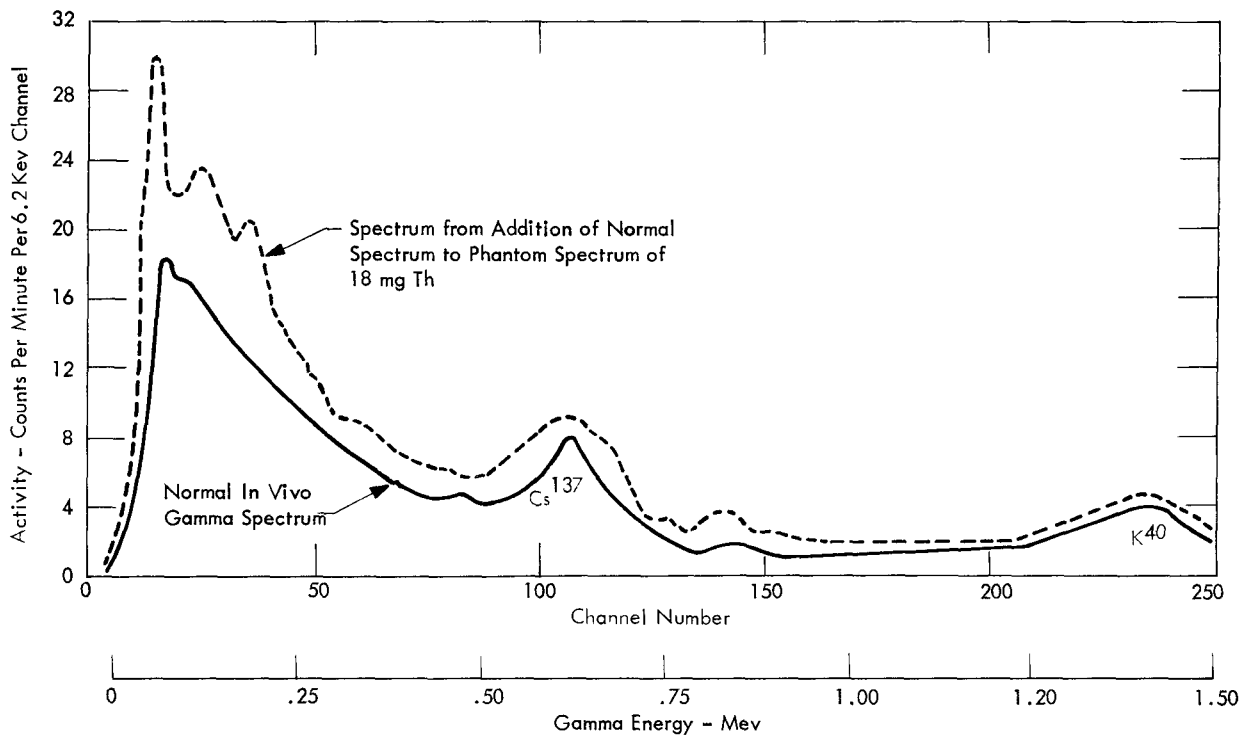

Figure 10. In Vivo Gamma Spectra With and Without Th
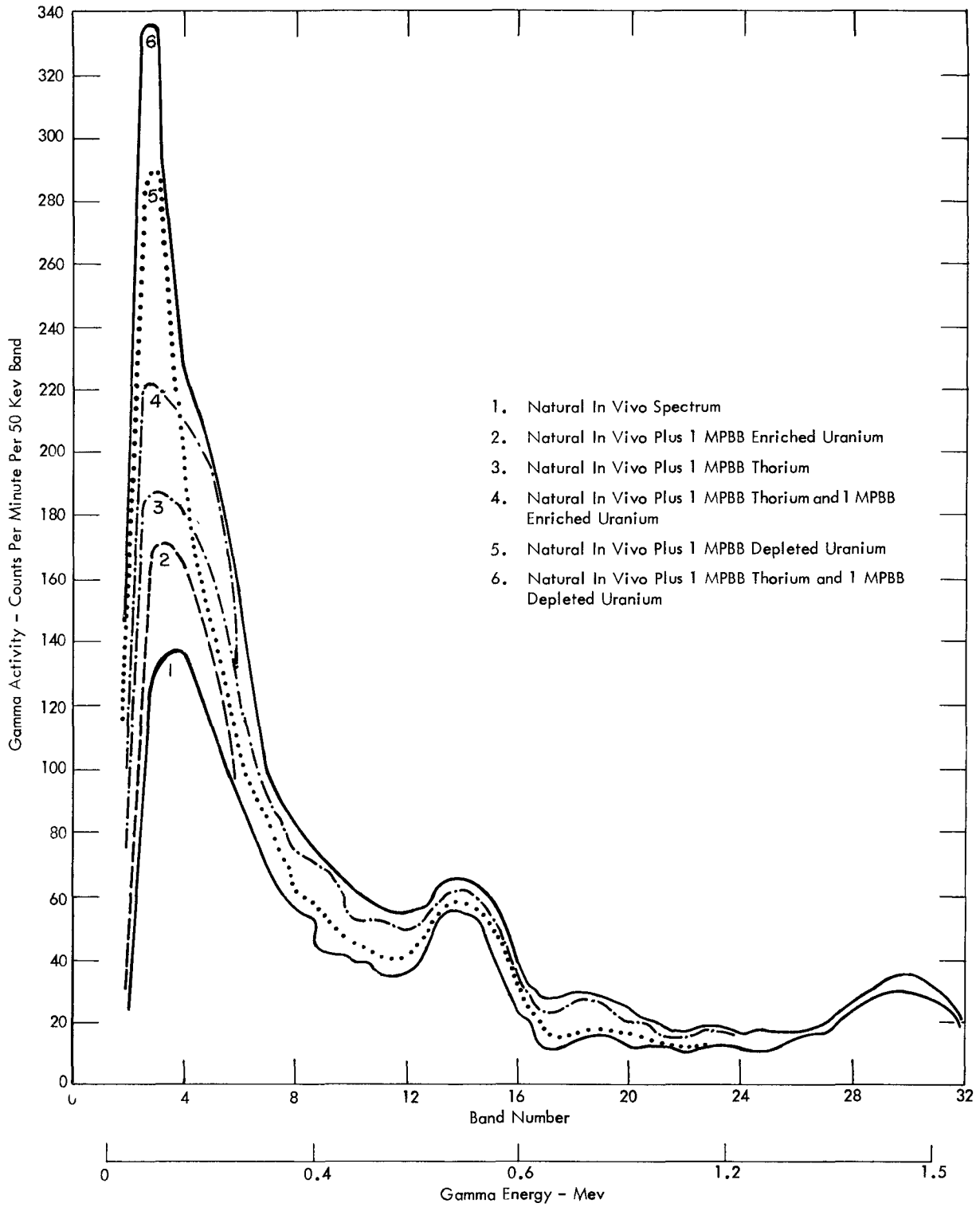
Figure 11. In Vivo Spectra With and Without Thorium and Uranium Combinations

1. Natural In Vivo Spectrum
2. Natural In Vivo Plus 1 MPBB Enriched Uranium
3. Natural In Vivo Plus 1 MPBB Thorium
4. Natural In Vivo Plus 1 MPBB Thorium and 1 MPBB Enriched Uranium
5. Natural In Vivo Plus 1 MPBB Depleted Uranium
6. Natural In Vivo Plus 1 MPBB Thorium and 1 MPBB Depleted Uranium

1. Normal in vivo gamma spectrum.

2. Normal in vivo gamma spectrum plus spectrum of 1 MPBB of enriched uranium.

3. Normal in vivo gamma spectrum plus·spectrum of 1 MPBB of thorium.

4. Normal in vivo gamma spectrum plus spectra of 1 MPBB enriched uranium and 1 MPBB thorium.

5. Normal in vivo gamma spectrum plus spectrum of 1 MPBB depleted uranium.

6. Normal in vivo gamma spectrum plus spectra of 1 MPBB depleted uranium and 1 MPBB thorium.

Various parameters, including those used in routine IVRM examinations for uranium lung burden, have been tabulated for all of these spectra in Table II.

On the basis of Table II and Figures 8, 9, and 11, the following items should be useful in determining if the excess activity in the lower energy channels were due to thorium or to uranium:

1. If thorium were present, lines at energies higher than those of the normal body activities would be observed between 1.45 and 2.6 Mev. If Ra^{228} were present with the thorium, an excess activity at 900 Kev (Parameter 12, Table II) would be observed.

2. The excess count rate in the 90 Kev region (Parameter 10) is much higher than that in the 186 Kev region (Parameter 11) for depleted uranium; but very nearly equal to the latter for thorium, as indicated by Parameter 15. Further study and data will be required to determine if the ratios for thorium, enriched uranium, and thorium plus enriched uranium could be differentiated.

3. In the uranium measurements, a rather definite relationship between excess count rate in 165-215 Kev (Parameter 11) and spectral slope (Parameter 7) has been found,[3] apparently according to the equation:

$$\text{Excess count rate (165-215 Kev)} = 1.1 \ (\text{Slope-19 cpm})$$

The data for thorium and for thorium-uranium combinations in the in vivo spectrum do not fit this equation. This is to be expected, since the spectral slope relates to the activity in the 215-265 Kev region, which receives counts from the 240 Kev activity of the Pb^{212} daughter of thorium. The relationship of spectral slope to excess count rate would probably differentiate the amounts of uranium and thorium in the body.

4. If careful IVRM examinations were made on employees before they begin work in thorium processing and periodically thereafter (in order to follow the natural body spectrum changes), the appearance of either thorium or uranium or both could probably be evaluated by comparison.

In general, it might be stated that very small quantities of either thorium or uranium can be detected in the lung. If thorium as represented by the gamma activity of the daughters were present, it is not likely that it would be identified as uranium alone. Similarly, if only uranium were present it would not be identified as thorium. However, if both uranium and thorium were present, the presence of thorium would be recognized while the recognition and certainly the quantitative estimation of the amount of uranium would be difficult. The importance of the ability to differentiate between uranium and thorium in the body depends upon the amount of thorium established as MPBB. If the 18 mg level used in the phantom experiments reported here is selected as the MPBB, the minimum detectable amount using the present spectral prediction equations for either thorium or uranium is about 50% MPBB; and the excess count rate per MPBB is essentially equal for the two materials. Under these conditions, there might be no need for separation of possible thorium exposures from possible uranium exposures, since a predetermined count-rate-in-excess-of-normal would be established as the action level regardless of which isotope was involved. On the other hand, if the 18 mg value is scaled down to 13% - as implied by the treatment of thorium as a plutonium-type hazard - it is not certain that the MPBB of 2.3 mg of thorium could even be detected in the body. Certainly, more rigorous treatment of the data than that considered above, and perhaps even a new approach in counting - such as the use of two crystals and perhaps coincidence counting - would be required. Under this condition, it would be very important that possible thorium exposures be kept free of possible uranium exposures, since the presence of 1 MPBB of insoluble uranium would almost certainly obscure the presence of several MPBB of thorium.

CONCLUSIONS

The decay scheme of the thorium series is complex and may provide many ratios of Th^{232} and daughter activities. A table of the radioactivity types and abundances that might be expected is provided. The decay chain has been studied and several critical aspects discussed in order to clarify the radioactivity patterns that might be expected under processing conditions.

In vivo gamma counting is a suitable method for detecting thorium and its daughters within the human lung. Quantitative estimation of Th^{232} in the body cannot be made without knowledge of the relative abundance of Th^{232} and its daughter Th^{228}. This abundance ratio would have to be known from analysis of the process or environmental material to which persons were exposed.

On the basis of the probably typical thorium metal used in phantom experiments, the application of nothing more than the routine evaluation now given in vivo spectra for the presence of uranium would measure as small a quantity of thorium as 9 mg.

Although no experimental data have been taken, use of two detectors for measurement of

coincidences between some of the simultaneously emitted gamma quanta appears to be a promising approach for reducing the limit of detection in the body.

The data presented suggest that the presence of either thorium or uranium alone, or of thorium, if thorium and uranium together were in the lung, would be relatively simple to detect by IVRM techniques. The quantitative estimation of uranium-thorium relative abundances, particularly in the case of enriched uranium, would be more difficult but not impossible.

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