Draft

REVIEW OF INTERNAL EXPOSURES TO THORIUM AND ITS PROGENY AT THE KANSAS CITY PLANT DURING MG-TH MACHINING OPERATIONS

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0 (Draft)	08/15/2014	Initial issue

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LIST OF ABBREVIATIONS AND ACRONYMS

AEC	Atomic Energy Commission
Bq	Becquerel
Ci/g	Curie per gram
cpm	count per minute
DU	depleted uranium
g	gram
KCP	Kansas City Plant
MPC	maximum permissible concentration
Mg	magnesium
m^3	cubic meter
mg/cc	milligram per cubic centimeter
mg/m^3	milligram per cubic meter
mr/hr	millirem per hour
µCi/cc	microcurie per cubic centimeter
$\mu Ci/m^3$	microcurie per cubic meter
µCi/ml	microcurie per milliliter
NBS	National Bureau of Standards
NIOSH	National Institute for Occupational Safety and Health
OCAS	Office of Compensation Analysis and Support
ORAUT	Oak Ridge Associated Universities Team
PER	Petition Evaluation Report
rem	Roentgen equivalent man
SC&A	S. Cohen and Associates (SC&A, Inc.)
SEC	Special Exposure Cohort
Sv	Sievert
Th	thorium

INTRODUCTION

This report specifically addresses the methods that NIOSH plans to use to reconstruct internal exposures associated with Mg-Th operations at the Kansas City Plant (KCP) from May 1, 1957, through April 30, 1979, and then separately from May 1, 1979, through May 31, 1984. Section 7.2.3.1 of the Special Exposure Cohort (SEC) Petition Evaluation Report (PER Petition SEC-00219, January 7, 2014) addresses this subject.

INTERNAL EXPOSURES TO TH-232

As explained in the SEC PER, there are limited air sampling data that might be used to estimate the airborne dust loading of alpha emitters during Mg-Th operations, and there are no bioassay data that can be used to reconstruct the internal exposures to thorium. The basic strategy adopted by NIOSH in the SEC PER for estimating the airborne dust loading of thorium and associated internal exposures during the Mg-Th operations is to use a combination of process knowledge and limits established and enforced on the maximum allowable airborne concentration of thorium. This report presents a review of the source documents cited in the SEC PER in order to assess the degree to which the protocols adopted by NIOSH for reconstructing internal exposures to thorium are scientifically sound and claimant favorable.

NIOSH proposes two different approaches to thorium dose reconstruction depending on the period. The first period would be from May 1, 1957, to October 31, 1959, and the second would extend from November 1, 1959, to April 30, 1979, the end of the Th-Mg alloy processing period. We will consider each of these in turn.

The May 1, 1957, to October 31, 1959 Period

NIOSH proposes to use a constant value of Th-232 in air of 9E-11 µCi/ml of air for this period. As stated on page 42 of the SEC PER:

> *In 1957 at the beginning of Mg-Th operations, KCP established engineered airborne limits and controlled these operations so as to not exceed 9E-11 µCi/ml (Th-232). KCP also instituted an industrial hygiene and fire protection limit of 0.1 mg/m^3 (thorium) (Thorium, unknown date).*

SC&A checked the reference (Thorium, unknown date) and confirmed that the KCP established strict controls for exposure to airborne toxins, primarily due to concern with respect to exposure to beryllium, and established a maximum permissible concentration (MPC) of 9E-11 µCi/ml of air. It certainly appears, therefore, that strict procedures were in place for maintaining exposures to Th-232 below this limit.

The SEC PER further argues that, since the nominal content of the Mg-Th alloy was 3% of thorium by weight, if the Th-232 were present airborne at the MPC, the dust loading of Mg-Th would be 27.3 mg/m^3, an extremely high dust loading that might create breathing difficulties for persons exposed to this airborne dust loadings for prolonged time periods. The specific activity of Th-232 is 1.1E-7 Ci/g. Hence, the MPC is associated with 9E-11 µCi/ml × 1/1.1E-7 Ci/g × 1E-6 Ci/µCi × 1E3 mg/g = 0.818 E-6 mg/cc of Th-232. Assuming that the Th-232 is only 3% of

the Mg-Th alloy, and that all of the radioactivity in the air is due to Th-232, the dust loading of the alloy would be 27.3 mg/m^3. This would mean that the chronic concentration of Th-232 in air could not exceed 9E-11 µCi/ml of air if the limit were maintained in practice.

We have several concerns that need to be addressed by NIOSH before we can conclude that the 9E-11 µCi/ml limit and associated 27.3 mg/m^3 argument are well founded:

1. The document to which NIOSH refers is undated. Actually, the date cannot be determined from the available version because the first page is missing (Thorium, unknown date). As a result, it is not possible to establish the date on which the 9E-11 µCi/cc limit was established. Specifically, one cannot say if the starting date was set "at the beginning of Mg-Th operations" in 1957, as claimed by NIOSH (SEC PER 2014, p. 42).
2. It is unclear from the document whether the 9E-11 µCi/cc was applicable at all. The document does indeed give a limit for "[a]irborne contamination thorium-232 – 9 × 10^{-11} microcuries per cubic centimeter of air;" however, the phrase "232 – 9E-11" has a manual strike out across it, so that the phrase as it stands is, "[a]irborne contamination thorium-~~232 – 9 x 10^{-11}~~ microcuries per cubic centimeter of air." A handwritten note that is rather faded and rather difficult to read appears next to the stricken phrase. So far as it is legible, SC&A believes it reads as follows: "For [illegible] – [illegible] × 10^{-12} [exponent unclear] µCi/cc for soluble material and [illegible] for insoluble. For soluble natural thorium (MPC) [illegible] is 2 × 10^{-12} µCi/cc. For insoluble natural thorium 6.9 × 10^{-12} µCi/cc" (Thorium unknown date, pdf p. 2). It is not clear when the handwritten edits to the document were made. A change in the external dose limits similarly made in longhand indicates that the edits might have been made in 1959, sometime after the issuance of the National Bureau of Standards Handbook 69 (NBS 1959).[1] However, even if true, this does not solve the problem of when the document was written and adopted at the KCP.
3. While the deleted phrase refers to thorium-232, the handwritten note refers to natural thorium, which would include thorium-228 and possibly other decay products. Though the original phrase referred to thorium-232, no measurement protocol or assumptions about equilibrium of thorium-232 with its decay products are described. Therefore, it is not clear if the limit of 9E-11 µCi/cc was applied to gross alpha or to natural thorium, including thorium-228, or to natural thorium with other decay products besides Th-228. If the MPC was applied to thorium-232 alone, it is not apparent what assumptions about equilibrium or lack thereof were made.
4. The document "Thorium, unknown date" discussed strict air pollution controls for beryllium, which it noted was much more hazardous than thorium. In that context, it concluded that, "…any potential hazard from thorium could be adequately controlled" (Thorium unknown date, pdf p.2). However, there are no air monitoring data for thorium in the 1957–1959 period. Rather, there are depleted uranium air monitoring data for the 1958–1970 period (ORAUT 2006, p. 19). These data are not relevant for addressing the

[1] The change in the document reduces external dose to 3 rem per quarter, 12 rem per year, and 5 rem per year long-term average over several years. These limits were introduced in NBS 69 (NBS 1959, pp. 4–5).

issue of actual Mg-Th alloy dust concentrations. We know that maximum permissible air concentrations were established in Atomic Energy Commission (AEC) facilities, but it was also known that they were often exceeded, especially in the 1950s and 1960s.

5. If the MPC was anything other than for thorium-232 alone, the presence of decay products, notably thorium-228, would need to be taken into account. Freshly separated thorium will include equal activities of Th-232 and Th-228; furthermore, Ra-224 3.66 day half-life and its progeny will grow in quickly. Hence, a gross alpha air sample will contain other alpha emitters, all with much higher specific activities than Th-232. Therefore, the mass of Th-232 in a gross alpha sample would be considerably lower than 0.818E-6 mg/cc. In turn, this means that the mass loading of Mg-Th alloy dust including Th-228 associated with 9E-11 µCi/ml of gross alpha would be much lower than 27.3 mg/m^3. Of course, this would also mean that the concentration of Th-232 in air would be lower than the 9E-11 µCi/ml limit. NIOSH's conclusion that the 9E-11 µCi/ml is bounding is based on the inference of total mass loading of 27.3 mg/m^3 and the notion that continuous exposure to dust at this level is the maximum tolerable concentration. If other alphas, along with Th-232 were being counted, the mass loading would be much lower and an inference that the air dust concentration was at the tolerable upper limit for continuous exposure would no longer be valid.

For workers involved in machining operations, the SEC PER states that exposure duration will be assumed to be 2,000 hours per year. If 9E-11 µCi/m^3 were a Th-232 limit, it would be very claimant favorable to assume that workers involved in Mg-Th machining operations were continuously exposed to airborne Th-232 at that level, but it is premature to make that assumption at the present time. There are simply too many questions around the value of 9E-11 µCi/cc for it to be used in any dose reconstruction, much less to show that it is a bounding value.

The SEC PER further states the following:

> *NIOSH will assume that the air concentration for general laborers was half the concentration of that of the operators. The supervisors of the operators and general laborers had half the concentration of the general laborers' concentration. All other worker types, such as those performing primarily administrative and clerical duties with no reason to enter the restricted, radiological areas, had 10% of the supervisors' concentration.*

As discussed with respect to uranium exposures, a certain degree of judgment will be required by the dose reconstructor when determining exposure duration for a given worker. This is an implementation issue that is best evaluated as part of dose reconstruction reviews, and is not considered an SEC issue. In addition, as with uranium exposures, the SEC PER states that internal exposures associated with the inadvertent ingestion of Th-232 will be derived using the guidance in OCAS-OTIB-009 (OCAS 2004). SC&A concurs with this approach for reconstructing internal exposures associated with the ingestion pathway based on our previous review of OTIB-009.

The November 1, 1959, to April 30, 1979 Period

NIOSH proposes to use a constant value of 3E-11 µCi/ml of Th-232 for this period (SEC PER 2014, p. 43). This is based on a document that specifies this value as an air concentration limit (Mg-Th 1957–1970, pdf pp. 13 and 16).

However, an examination of the document reveals the matter to be more complex. The document in question states the following:

> *Personnel exposure limits have been established for thorium related to its chemical toxicity and radioactivity as follows:*
>
> *(1) 0.1 milligrams of thorium per cubic meter of air.*
> *(2) 3 × 10^{-11} microcuries per milliliter of air.* [Mg-Th 1957–1970, pdf p. 16]

One issue is that if both values relate to Th-232, they are incompatible. The first, 0.1 mg/m^3, translates into 1.1E-11 µCi/ml, which is much lower than the radioactivity limit specified. It is possible that the mass limit was different and stricter than the radioactivity limit. However, we should note that the maximum permissible air concentration established in the National Bureau of Standards Handbook 69 for thorium-232 was 1E-11 µCi/cc (NBS 1959, p. 82), which is close to the value of 0.1 mg/m^3. It appears, therefore, that 3E-11 µCi/ml may have been a gross alpha limit, though the quote indicates it may well have been a thorium limit. In the latter case, it is unclear if thorium-228 was included or if the limit was for thorium-232 only.

NIOSH cites gross alpha air concentration measurements in the Main Manufacturing Building between 1958 and 1971 and a thorium-related test measurement in 1970 as its basis for the choice of 3E-11 µCi/ml:

> *KCP performed gross alpha fixed-filter air monitoring from 1958 through 1971 in the Main Manufacturing Building and maintained operations at: 2.85E-12 µCi/ml (average measured level for the period) and <8.55E-11 µCi/ml (maximum measured level for the period) (ORAUT-TKBS-0031, pdf p. 20).* [ORAUT 2006]
>
> *In 1970, KCP performed an evaluation of Mg-Th machining operations in the Model Shop and validated that their process did not generate airborne radioactivity. As part of this test, each machining station was evaluated with breathing-zone air samplers running while performing the machining operation. KCP machinists maintained airborne levels during this test at background (0 cpm recorded) for long-lived activity, and <3.22E-9 µCi/ml for short-lived activity. Direct-probe surveys of the Mg-Th are shown as: 0.3-1.2 mr/hr for beta-gamma radiation, and 250–700 cpm alpha (Mg-Th, 1962–1975).* [SEC PER 2014, p. 43]

SC&A has some concerns about this reasoning. First, the air concentration measurements in the Site Profile (ORAUT 2006, p. 19) that NIOSH cited do not refer to thorium or gross alpha, but to

depleted uranium (DU).[2] Presumably, gross alpha measurements were made in DU processing areas and hence the radioactivity was attributed to DU. In that case, they would have no relevance for making any conclusions about thorium air concentrations. Specifically, NIOSH has not provided any evidence that any of these air samples were taken at times and in areas of Mg-Th alloy processing.

NIOSH's second rationale concerns a single test done in 1970. SC&A evaluated the document in which this test is described (Mg-Th 1962–1975, pdf pp. 16–20). While the test results do indeed show that the counts of alpha activity after several days were zero, the sensitivity of the test was not high. Specifically, samples were taken over brief periods (5 minutes) and counting was also done over brief periods (2 to 5 minutes). Actually, these counting times were for the count 1 hour after the sample was taken. The counting times several days after the samples were taken are not provided. Only a result "0 cpm" is stated in each case.

It is evident from the 1-hour results that a considerable amount of thorium-related short-lived activity was present in the air. In most cases, the sample volume was 14 liters. The counting efficiency was estimated at 0.3. With these parameters, the test would register a single count in 5 minutes at long-lived alpha air concentrations of 2E-11 µCi/ml. Background counts are provided; they are between 0 and 3 cpm. The latter would correspond to a thorium-232 plus thorium-228 count of about 6E-11µCi/ml, which is well above the value that NIOSH proposes to use as a constant for the 1959–1979 period.

Finally, using a constant value of 3E-11 µCi/ml assumes that the regulatory limits were generally met at KCP. There are many of examples in the nuclear weapons complex when that was not the case in the 1950s and 1960s, and even into the 1970s. NIOSH's assumption is at present without substantial empirical foundation, since there appear to be no thorium air samples for 1959–1969 and 1971–1979. The one test during a short period in the 1970s is lacking in sensitivity to determine what value would be suitable even for that year, let alone any other year.

We may conclude that:

- The short-lived radionuclide counts show a definite presence of thorium-related decay products.
- The counting times for long-lived radionuclide counts are not provided. If they were similar to the counting times for short-lived radionuclides, the method was quite insensitive and may not have registered amounts that are of the same order of magnitude as the constant value that NIOSH proposes to use.

There appear to be no thorium air monitoring data from 1971 onward. At least NIOSH cites none.

In sum, the 1959–1979 period value for thorium-232 air concentration is based on a single test measurement in 1970. This test was inadequate at best for indicating conditions, even during the

[2] The quote cites page 20 of the Site Profile (ORAUT 2006). However, that page does not contain air concentration measurements, which are on p. 19. SC&A assumes this is a typo in the PER.

time of the test, in so far as the constant value that NIOSH proposes to use is concerned. No data have been provided for the 1971–1979 period. The data for the 1959–1970 period that NIOSH has cited do not refer to thorium, but to DU and are, therefore, not relevant to thorium dose reconstruction.

THORIUM CONCENTRATION IN THE MG-TH ALLOY

NIOSH assumes that machining of Mg-Th alloy was limited to 3% Mg-Th alloy. During the Work Group meeting held on June 10, 2014, SC&A raised the question whether the KCP might have actually fabricated Mg-Th alloy in addition to simply machining the alloy. SC&A raised the question because, if Mg-Th alloy was fabricated at the KCP, it would likely have involved using a master alloy that had much higher concentrations of thorium. During the meeting, NIOSH indicated that they have documentation indicating that only 3% Mg-Th alloy (e.g., HK31 alloy) was handled at KCP, and that they would modify the SEC PER to that effect.

However, a compilation of Mg-Th documents (Mg-Th 1957–1970) indicates that several different alloy compositions were processed:

- A 3.5% Th concentration is given in an attachment to an October 1959 memorandum (mg-Th 1957–1970, pdf p. 14)
- A 4% concentration is given in a 1960 document (Mg-Th 1957–1970, pdf p. 9)
- A 2% concentration is mentioned for 1970 (Mg-Th 1957–1970, pdf p. 2)

It is important for NIOSH to determine the proportions of thorium-232 in the Mg-Th alloy, especially for the 1957–1959 period, but also in the latter period.

INTERNAL EXPOSURES TO THE PROGENY OF TH-232 AND TH-228

The above discussion appropriately addresses exposures to Th-232, but we also raise possible issues associated with the progeny of Th-232. The SEC PER is silent regarding possible exposures to the progeny of Th-232. In general, all of the Th-232 progeny are present in equilibrium in thorium ore, as shown in Figure 1:

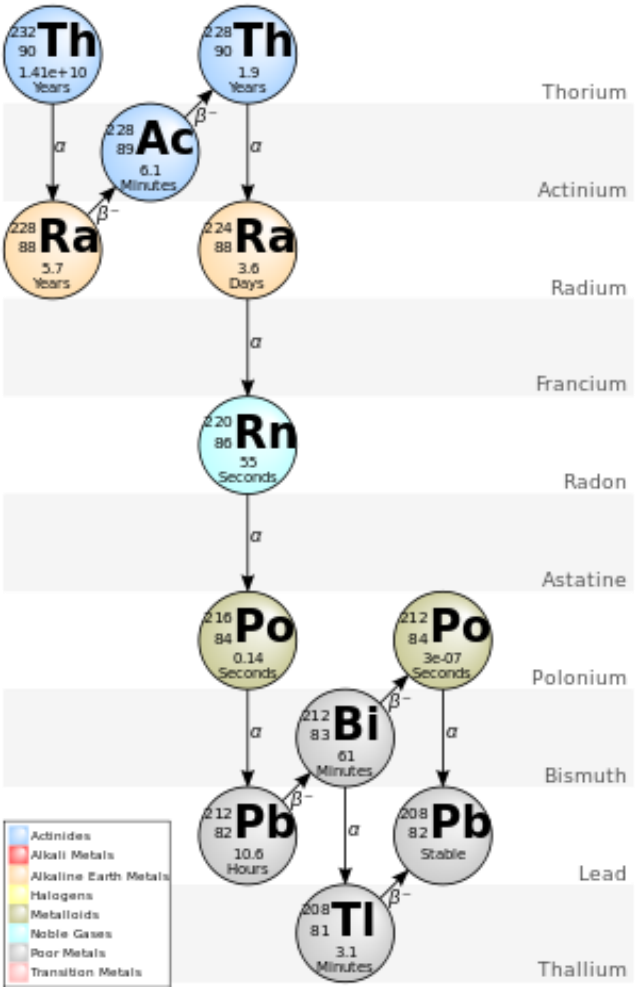

Figure 1. Thorium-232 Decay Series

When thorium is chemically separated from its ore, the product includes equal activities of Th-232 and Th-228, and Mg-Th alloy prepared from freshly separated thorium will not contain any of the non-thorium progeny. As time passes, the Th-228 begins to decay with its 1.9 year half- life and its progeny grows in. In addition, as Th-232 decays, all of the progeny begin to grow in. Hence, the mix of radionuclides in the alloy depends very much on the age of the thorium in the alloy.

The issue of the decay products of Th-232 present in air is also important from the point of view of dose estimation. Th-228 would likely contribute significantly to dose. In the process of separating thorium from its ore, the product will include equal activities of Th-232 and Th-228. As evident from Figure 1, the ratio of Th-232 to Th-238 will change in a complex manner as both the Th-232 and Th-228 decay. As it turns out, the inhalation dose conversion factor for the lung for Type M Th-228 is 1.81E-4 Sv/Bq, while for Th-232 it is 2.71E-5 Sv/Bq. Hence, if the contributions to internal organ dose from Th-228 are not included, the result could underestimate organ dose significantly, most importantly in the case of lung dose. Doses from other decay products may also contribute to dose, depending on the disequilibrium state of those products in

the air sample in question. In any case, ignoring decay products is not claimant favorable; they must be included whatever the approach chosen for reconstructing internal exposures to thorium associated with machining Mg-Th alloy. This subject was discussed during the June 10, 2014, Work Group meeting, and NIOSH agreed to address this issue in a future revision of the SEC PER.

Finally, page 39 of the SEC PER addresses exposure to thoron, stating that the airborne concentration of thoron (and the associated exposures) will be calculated based on the airborne concentration of Th-232. Page 44 of the SEC PER states that NIOSH will assume that the thoron concentration is 3.2E-9 µCi/ml, which basically assumes that thoron is at about 30% equilibrium with Th-232. The following explains why we believe this is a reasonable assumption.

One of the progeny, thoron, grows in quickly and can become airborne, because it is a noble gas. However, for it to become airborne as a separate radionuclide, it must escape from the alloy before it decays with its 55-second half-life. Hence, it is unlikely that much of the thoron will escape from the alloy before it decays (i.e., it is trapped in the relatively large mass of the alloy and decays before it has an opportunity to escape). However, the airborne particles of alloy dust generated during machining are very fine, and it is possible that some of the thoron could escape. Taking guidance from the behavior of radon (see Yu 1993), it is reasonable to assume that the emanation coefficient of thoron is about 30%. This means that when thoron's parent (Ra-224) decays, its recoil energy could result in thoron escaping from the small particle and becoming airborne. Hence, SC&A concurs with NIOSH's approach to addressing thoron exposure at the KCP.

CONCLUSIONS

NIOSH's approach to reconstructing internal doses associated with the machining of Mg-Th alloy at the KCP is based on assumptions that need to be clarified. Data and evidence are also lacking in certain cases.

Our overall conclusion is that the available data are insufficient to assign thorium doses to workers in the 1957–1979 period. Specifically:

1. For the 1957–1959 period, it is unclear whether the 9E-11 µCi/ml was a Th-232 limit, a natural thorium limit, or a gross alpha limit. This issue is central for determining whether a mass loading limit can be used for this period. Furthermore, there are no data to enable a determination of whether this limit was enforced and actual air concentrations of Th-232 remained generally below this limit.
2. The 1958–1970 air concentration data that NIOSH referred to are for DU (according to ORAUT 2006), not thorium. NIOSH has provided no evidence that any of these air samples related to Mg-Th processing.
3. For the period after 1959, it is unclear whether the limit of 3E-11 µCi/cc includes Th-228 and possibly other decay products of Th-232. The limit for Th-232 based on the lung as the critical organ set in NBS 69 was 1E-11 µCi/cc. NBS 69 was published in 1959.

4. NIOSH has not provided any air monitoring data for 1971–1979.
5. NIOSH refers to one thorium machining air concentration test. This test is inadequate to determine the value that should be used even for the year of the test, much less for any other year.
6. The issue of doses from the progeny of thorium needs to be addressed.
7. NIOSH needs to determine the various alloy compositions that were machined, and whether variation in thorium content may have made a difference to particulate generation during machining.

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