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*Draft*

**ADVISORY BOARD ON  
RADIATION AND WORKER HEALTH**  
*National Institute for Occupational Safety and Health*

**A FOCUSED REVIEW OF OPERATIONS AND THORIUM EXPOSURES  
AT THE DOW CHEMICAL COMPANY MADISON PLANT**

**Contract No. 200-2004-03805  
Task Order No. 5  
SCA-SEC-TASK5-0057**

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<p>S. COHEN &amp; ASSOCIATES:</p> <p><i>Technical Support for the Advisory Board on Radiation &amp; Worker Health Review of NIOSH Dose Reconstruction Program</i></p>	Document No. SCA-SEC-TASK5-0057
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## 1.0 BACKGROUND

On November 27, 2006, the law firm of Simmons-Cooper, LLC, filed a Special Exposure Cohort (SEC) Petition with the National Institute for Occupational Safety and Health (NIOSH) Office of Compensation and Support (OCAS) in behalf of workers at the Dow Chemical Company (DOW) plant in Madison, Illinois. On December 14, 2006, the Department of Health and Human Services published a notice in the Federal Register (71 FR 75258) indicating that the federal government had made a decision to evaluate the Dow employees petition to determine if the group should be included in an SEC. The period of employment for the proposed SEC was defined as January 1, 1957, through December 31, 1960. The period was chosen because during that timeframe, Dow did some uranium fabrication work related to Atomic Energy Commission (AEC) programs.<sup>1</sup> For the defined period, it is possible that some Dow workers received exposure to uranium (and daughter products) from the AEC uranium fabrication work, and to thorium from commercial activities related to the routine commercial melting, casting, and fabrication of Mg-Th alloys.

As required by both Energy Employees Occupational Illness Compensation Program Act of 2000 (EEOICPA) and Title 42, Part 83 of the *Code of Federal Regulations* (42 CFR Part 83), NIOSH issued *SEC Petition Evaluation Report for SEC-00079* (NIOSH 2007) on April 13, 2007. In its evaluation, NIOSH decided that both uranium and thorium doses need to be considered during the time period of January 1, 1957, through December 31, 1960. The evaluation concluded the following:

*NIOSH has documented herein that it cannot complete the dose reconstructions related to this petition where doses resulted from exposure to thorium-containing materials. The basis of this finding is specified in this report, which demonstrates that NIOSH does not have access to sufficient information to estimate with sufficient accuracy either the maximum radiation dose incurred by any member of the class or to estimate such radiation doses more precisely than a maximum dose estimate. Members of this class at the Dow Chemical Company site in Madison, Illinois, may have received unmonitored internal and external radiological exposures from thorium radionuclides at the plant. NIOSH lacks sufficient information, which includes sufficient personnel and workplace monitoring data and radiological source term information, to allow NIOSH to estimate the potential total internal thorium exposures to which the proposed class may have been exposed.*

*With the data currently available to NIOSH, it is feasible to reconstruct with sufficient accuracy the external and internal doses resulting from exposure to uranium metal during the Dow Madison AWE operational period (January 1, 1957*

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<sup>1</sup> On March 15, 1957, Subcontract No. 25034 was implemented between the Uranium Division of Mallinckrodt Chemical Works and the Madison Division of Dow Chemical Company to do research and development work on gamma phase extrusion of uranium metal. No contractual evidence of prior work has been developed. However, a Record of Contact prepared by Spectrulite Consortium, Inc., quotes a former Dow employee as stating, "The work was done in the plant extrusion department beginning in 1955 for a period of about two years—may have been longer" (Young 1988).

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*through December 31, 1960), and during the residual radiation period (January 1, 1961 through December 31, 1998). NIOSH also considers the reconstruction of medical dose for Dow Madison workers to be feasible.*

At a meeting of the Advisory Board on Radiation and Worker Health (Advisory Board) in Mason, Ohio, on February 9–11, 2007, SC&A, under its continuing support to the Advisory Board, was tasked to review the SEC petition and the NIOSH evaluation of the petition. Subsequently, at the Advisory Board Meeting in Denver, Colorado, on May 2, 2007, SC&A was asked to evaluate thorium exposures from 1961 forward at the Dow (Madison) plant and to review newly available material on the Madison site. This material consisted of approximately 700 pages of information obtained by NIOSH from Dow Chemical Company and made available on the “O” drive on May 1, 2007. This report to the Advisory Board describes relevant activities at the Dow Plant for the defined time period and SC&A’s attempt to further clarify thorium exposures from 1961 forward.

The material that follows explicitly addresses exposures to thorium. With respect to exposures to uranium, SC&A concurs with NIOSH’s conclusion that uranium exposures to workers during uranium operations and exposures to residual uranium subsequent to the termination of uranium operations at Dow Madison can be reconstructed in a scientifically valid and claimant-favorable manner. The basis for this conclusion is our understanding that the nature of the uranium operations that took place at Dow Madison during the covered period was similar to the uranium metal-working operations that took place at numerous other DOE/AWE facilities. We have reviewed numerous reports addressing these types of operations, including the reports by Harris and Kingsley (1959), Adley et al. (AEC 1952), and documentation in support of the *Site Profile for Simonds Saw and Steel* ORAUT-TKBS-0032 (ORAUT 2005) and the recently issued *Site Profiles for Atomic Weapons Employers that Worked Uranium and Thorium Metals* (Battelle-TBD-6000 2006). We believe that these reports, taken in combination, are comprehensive in describing the range of operations and the range of exposures that workers might have experienced during uranium metal-working operations at DOE/AWE facilities during the early years. We believe that the material contained in these reports can be used to place plausible upper bounds on the uranium exposures experienced by workers at the Dow Madison facility.

On August 6, 2007, while this report was being prepared, NIOSH issued *Addendum to Dow Madison (SEC-00079) Special Exposure Cohort Evaluation Report* (referred to here as the Addendum). The Addendum provides a review of approximately 700 pages of Dow documents that were placed on the “O” drive. SC&A has had an opportunity to briefly review the Addendum, and has thoroughly reviewed the Dow documents, and concurs with the factual material provided. SC&A also concurs with the Addendum conclusion that upper-bound external exposures to thorium at the Dow Madison facility can be reconstructed, based on a combination of a limited amount of external exposure data, and, more importantly, knowledge of the upper-bound exposures associated with working in close proximity and in contact with thorium metal and metal alloys, as determined using external exposure models, such as MCNP.

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## 2.0 OPERATIONS AT DOW - MADISON

During the period 1957 through 1960, and presumably before and after that period, Dow (and successor owners of the plant) produced magnesium alloys containing thorium for commercial customers.<sup>2 3</sup> Dow noted in its Annual Report for 1957 that, “Among promising **new** products are our **new** thorium-containing alloys which have greatly improved strength retention at elevated temperatures. They are being used principally in missiles and high-speed aircraft where they have distinct advantages over other metals in the 400 to 700 degree range” [emphasis added]. Some thorium sampling data from as early as September 1955 are available (Levy 1959).

Dow applied to the NRC for exemption from the licensing requirements under 10 CFR 20 (or 10 CFR 40) for Mg-Th alloys requiring less than 4% Th. [A draft exemption letter was prepared by D. Cunningham of NRC on July 12, 1957, supporting this application (Cunningham 1957).] On July 14, 1958, AEC License No. C-2782 was amended to exempt Dow from the requirements of 20.203(e)(2) and 20.203(f)(2) of 10 CFR Part 20.

Commercial use of Mg-Th alloys declined in later decades. ASM International observed the following in a 1998 publication (ASM 1998):

*Although thorium-containing alloys have found applications in missiles and spacecraft (Ref 14), they have lost favor because of environmental considerations and are generally considered obsolete. In Britain, for example, alloys containing as little as 2% of thorium are classified as radioactive materials that require special handling, thereby increasing the cost and complexity of manufacturing them. However, many thorium-containing magnesium parts are still in use, and some placement castings are still being produced; hence, some information on these alloys continues to be covered in various articles in this Handbook.*

A layout of the Dow Madison plant is included as Figure 1 (Cottrell and Williams 1990). Key locations include Building 7, where melting and casting were done; Building 6, where extrusion fabrication was done; and Building 5, where sheet and plate were rolled. A waste disposal area was located north and east of Building 7. Processing of magnesium-thorium (Mg-Th) alloys occurred in all of these major locations. Uranium work for the AEC was limited to Buildings 5 and 6. Experimental uranium extrusion work was done in Building 6 and uranium rod straightening was done in Buildings 5 and 6. There are unconfirmed reports that some uranium rolling was done in Building 5. One document also mentions Buildings 152 and 356 (Silverstein 1956b). Interviews with former employees could not identify these buildings and it is possible that they were at another Dow plant site.

<sup>2</sup> According to Affidavit No. 9 (Dowpet 2006), work on Th-containing materials continued from 1951 through about 1998.

<sup>3</sup> Commercial customers are those who purchase standard mill products based on normal terms and conditions. Commercial customers may be government entities.

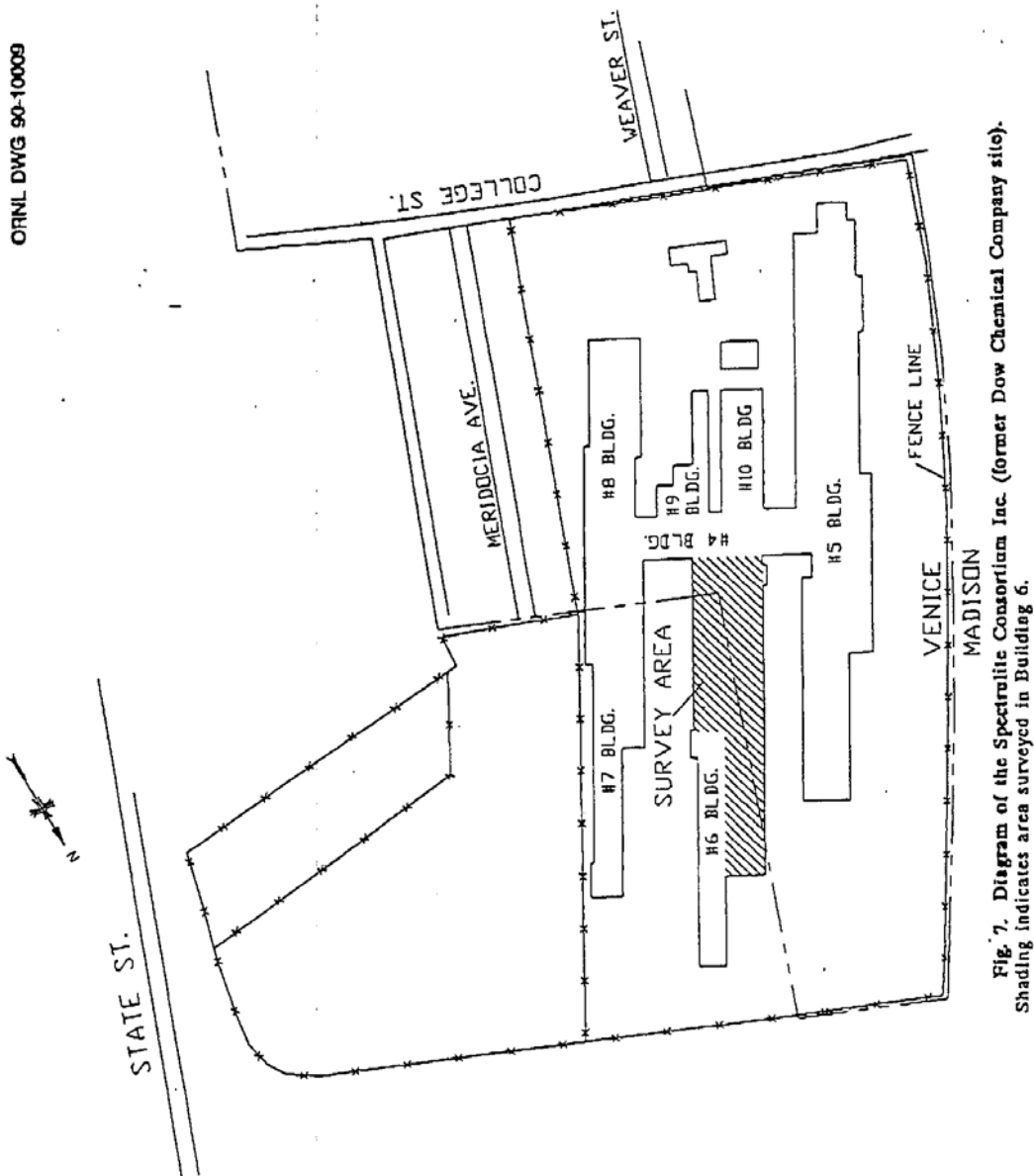


Fig. 7. Diagram of the Spectralite Consortium Inc. (former Dow Chemical Company site). Shading indicates area surveyed in Building 6.

**Figure 1. The Dow Madison Plant**

Thus, to the extent that workers did not move from one production area to another during the 1957–1960 timeframe due to job changes or nature of the job, workers in melting and casting (Building 7) would not have received significant uranium exposure. Workers in Buildings 5 and 6, where uranium extrusion and, presumably, rod straightening were done, would clearly have received both uranium and thorium exposures during this period.

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Interviews with former employees indicate that some worked their entire careers in the casting department, while some worked in several areas within the plant.

## 2.1 Radioactive Materials Handled

Alloys are identified by a code, which contains two letters and two numbers. The letters identify the metals in the alloy and the numbers the amount of metal, in percent, in the alloy. For magnesium-base alloys, H in the alloy descriptor indicates the use of thorium as an alloying agent, K indicates the use of zirconium as an alloying agent, and M indicates the use of manganese as an alloying agent. Alloy HK31 has a nominal composition of 3% Th and 1% (actually 0.7%) Zr. Worker Affidavit No. 2 specifically mentions alloys HK31 and HM21 (Dowpet 2006; see also Affidavit No. 25). Other thorium-bearing alloys processed at the Madison Plant included HZ32A and ZH62A (Silverstein 1957a). The *Dow Product Standards* (July 1, 1968) list only three thorium-containing alloys; HK31A, HM21A, and HM31A (Dow 1968). See Attachment A for additional details on Mg-Th alloys. Additional alloys were identified in the NIOSH Addendum.

In addition, according to Affidavit No.1 (Dowpet 2006), uranium was processed on rolling mill #7 in the 1950s. While it is not totally clear from the affidavit, it appears that this was a one-time operation involving a single slab of uranium. No confirmation of uranium rolling has been uncovered to date in other documents. The affidavit also suggests that uranium rods were straightened either on the rolling mills or via extrusion. This testimony was from a second worker who started work in 1961. Further discussion of uranium rod straightening is provided in Dow 2007. The extrusion and straightening of uranium rods is elucidated in SRDB Ref ID 3804.pdf, which is an excerpt from the closeout report for the FUSRAP Madison site. The report indicates that extrusion and straightening of uranium rods were conducted under contract to Mallinckrodt in the late 1950s and early 1960s. According to worker interviews, three methods of rod straightening were attempted in Buildings 5 and 6, but none were successful and the material was returned (Dow 2007). According to the FUSRAP report, "Records suggest that only a small quantity of uranium was involved in these operations." The full text of SRDB Ref ID 3804.pdf, included here as Attachment B, describes Building 6 where the extrusion work was done.

According to Worker Affidavit No. 2 (Dowpet 2006), the thorium was received in barrels and stored in the back of Building 7. This building contained the pot room, where various magnesium alloys, including Mg-Th alloys, were melted. Thorium was initially received as notched bars and later as pellets. According to Affidavit No. 9, the thorium bars came from England. (This is probably an error, since the AEC License specified import from Dominion Magnesium Limited in Canada). Silverstein (1957a) mentions that Dow also made a master alloy or "hardener" containing 25% thorium. However, it is not clear if Silverstein's comment is related to Dow (Madison) or to Dow (Bay City). In its report of an inspection conducted by the AEC on August 4, 1960, the AEC inspector notes that thorium pellets, obtained from Canada, were stored within a fenced-off area in an isolated section of the Melt Room Building (USAEC 1960). The fenced-off area had radiation warning signs and symbols. It is noted in the AEC report that 80 tons of thorium metal had been used through mid-1960 at the Dow magnesium

foundry.<sup>4</sup> The inspector stated that “no items of noncompliance were observed or otherwise noted” during his visit to the plant in 1960. It is noted in the 1960 report that a prior inspection had been made from April 20–22, 1959.

Details on the inventory of Th-bearing materials at various times, based on information contained in the annual AEC license applications, is included in Table 1. The 1957 license application includes an estimate of annual requirements for 135,000 lb of thorium metal and 30,000 lb of ThF<sub>4</sub> or ThO<sub>2</sub>. By the time of license renewal for 1960, the estimated requirements for ThF<sub>4</sub> or ThO<sub>2</sub> had dropped to 2,000 lbs, suggesting that the process of reducing thorium salts to make an Mg-Th hardener was being phased out. Also note the small quantities in inventory in 1956, suggesting that commercial manufacture of Mg-Th alloys on a significant scale had not begun.

**Table 1. Thorium Inventory at Dow Madison<sup>5</sup>**

Description	Grade	Th Content (%)	Quantity (lb)
October 31, 1956 Inventory			
Th metal – sintered pellets	Pure	97	4,766
ThF <sub>4</sub>	Commercial	71	22,702
October 31, 1957 Inventory			
Th metal – sintered pellets	Pure	97	6,254
ThF <sub>4</sub>	Commercial	71	10,542
October 31, 1958 Inventory			
Th metal – sintered pellets	Pure	97	2,426
ThF <sub>4</sub>	Commercial	71	9,700
Th scrap	Commercial	3	4,799
Th scrap	Pure	100	1,312
October 31, 1959 Inventory			
Th metal – sintered pellets	Pure	97	12,580
ThF <sub>4</sub>	Commercial	71	9,480
Th scrap	Commercial	3	1,012
Th scrap	Pure	100	114

## 2.2 Alloy Melting and Casting

The Mg alloys were melted in cast iron pot furnaces. It is noted in the final affidavit in Dowpet 2006 (the “Dow Employee Outreach Meeting” Affidavit, pg. 6) that some of the melting pots were 6,000 lbs, but it was later verified (Dow 2007) that this refers to the load in the pots. Apparently, the thorium was weighed on a scale and then introduced into the magnesium melt via a basket that permitted the thorium to be forced under the melt surface. According to

<sup>4</sup> It is not clear whether this is 80 tons of Th metal or 80 tons of Th-containing material. Elsewhere in USAEC 1960, the following breakdown is provided: 64 tons as pellets from Canada, 13 tons in the form of scrap from Consolidated Edison Company, and 3 tons of master alloy containing 25%–40% Th as notched ingots from Canada.

<sup>5</sup> It should be noted that estimates for annual thorium usage provided by worker testimony are much greater than estimates in this table (see discussions in Dow 2007).

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Saunders (1960), the maximum frequency of weighing pellets during a casting run (using a scoop) is once per hour requiring 1 to 3 minutes. "The frequency of casting runs may be as high as once per month, extending to a maximum of five continuous days per run."

A "setting" for melting Mg-Th alloys consisted of a group of 10 melting pots. When alloying was completed in the required number of pots, the molten alloy was pumped to a continuous casting machine, where it was solidified and cut into slabs or billets. The slabs were then scalped prior to movement to one of the fabrication areas. Scalping involved removing about 3/8 in of material from the slab surfaces, leaving a relative smooth, clean surface for subsequent fabrication.

Based on the description in Affidavit No. 2, it appears that thorium was introduced directly into the melt. However, according to the generic description in Albert (1966, Chapter 3, Section 8, pg. 42), a master alloy containing equal parts of Th and Mg is first prepared, and this master alloy is used to obtain the correct composition in the Mg-Th alloy. Albert further notes that the maximum Th concentration in Mg-Th alloys is 4%. The use of master alloys is not mentioned in the Affidavits, but is mentioned in Dow internal documents (Silverstein 1956b, Silverstein 1957a). It is possible that master alloys were used at Dow plants other than the Madison location.

Hardener production at Dow involved the dissolution of ThF<sub>4</sub> powder in a bath of molten KCl salt, and then reacting this mixture with molten magnesium (Silverstein 1957a). Originally, operations were conducted in Building 152 under a steel hood, where the airflow rate ranged from 50 to 100 linear feet per minute (lfm) and averaged 72 lfm. In some cases, two or three floor fans were added to increase the airflow in the vicinity of the reaction vessels to 80 to 200 lfm, averaging 125 lfm at the hood (Silverstein 1956b). Air-sampling results are included in Table 2. Since workers from the Dow Madison plant could not identify Building 152, it is possible that hardener was produced at another Dow facility. As noted by Silverstein (1956b), Sample I-1 was a worst-case measurement:

*The actual hazard to workers would be less than indicated since workers normally would not be exposed continuously to concentrations shown, and would not occupy positions as close to the operations as the sampling locations.*

Concentrations of radioactivity have been added in brackets in Table 2 by the authors of the current review, based on the tolerance conversion factors presented in footnote a to Table 2.

**Table 2. Air-Sampling Measurements during Mg-Th Hardener Production (Silverstein 1956b)**

Sample Number	Date	Description	Sampling Time & Volume	Analysis <sup>a</sup>
I-1	4-11-56	Taken near two pots during the addition of ThF <sub>4</sub> to molten KCl (1400°F). ThF <sub>4</sub> dropped from hopper into stirred KCl. Position of sample was somewhat nearer the pot than the operator normally stands.	10.0 min. (0.85 m <sup>3</sup> )	Thorium is 10 times tolerance [1.1 × 10 <sup>-13</sup> Ci/L], MsTh is 10 times tolerance [1.2 × 10 <sup>-13</sup> Ci/L], short-lived alpha activity is 3,000 times tolerance [3 × 10 <sup>-8</sup> Ci/L]
I-2	4-11-56	Taken near workbench about 30 ft. from pot. 20 min. after ThF <sub>4</sub> addition completed.	10.0 min. (0.85 m <sup>3</sup> )	Short-lived alpha activity is 50 times tolerance [5 × 10 <sup>-10</sup> Ci/L]. Th and MsTh are 1.5 times tolerance [1.8 × 10 <sup>-14</sup> Ci/L for MsTh and 1.6 × 10 <sup>-14</sup> Ci/L for Th].
I-3	4-12-56	Near workbench about in the center of the work area after ingots of Mg-Th hardener were poured; cleanup was in progress during sampling.	10.0 min (0.85 m <sup>3</sup> ).	Short-lived alpha activity 75 times tolerance [7.5 × 10 <sup>-10</sup> Ci/L]. Long-lived activity 70% of tolerance (Th and MsTh) [0.84 × 10 <sup>-14</sup> Ci/L for MsTh and 0.77 × 10 <sup>-14</sup> Ci/L for Th]
I-4	4-12-56	Near workbench 5 min. after ThF <sub>4</sub> addition had begun.	5.0 min. (0.425 m <sup>3</sup> )	Short-lived alpha activity 8 times tolerance [8 × 10 <sup>-11</sup> Ci/L]. Long-lived activity is 40% of tolerance (Th and MsTh) [0.48 × 10 <sup>-14</sup> Ci/L for MsTh and 0.44 × 10 <sup>-14</sup> Ci/L for Th].

a – Continuous exposure tolerances: mesothorium (MsTh) (Ra-228) – 1.2 × 10<sup>-14</sup> Ci/L of air; short-lived alpha products (radon and thoron) – 10<sup>-11</sup> Ci/L of air (Silverstein 1956b). Silverstein quotes the tolerance for Ra-228 as 1 × 10<sup>-14</sup> Ci/L of air in Silverstein 1957a. These are internal Dow standards. He also quoted the permissible level for thorium as 1.1 × 10<sup>-14</sup> Ci/L (Silverstein 1957a).

While not shown in Table 2, Silverstein (1957a) stated that use of local exhaust that drew air at 150 lfm across the face of the pot reduced the long-lived alpha and beta emitters to 1/10 of the tolerance level (presumably this was 1.2 × 10<sup>-15</sup> Ci/L for Ra-228 and 1.1 × 10<sup>-15</sup> Ci/L for Th). Short-lived alpha radioactivity ranged from 10<sup>-14</sup> to 10<sup>-12</sup> Ci/L, values well below the tolerance limit of 10<sup>-11</sup> Ci/L.

The above discussion of hardener production is taken from documents, as referenced, from those available on the “O” drive for Dow (Madison). Further review of this material, and in light of the information gathered during the workers’ meeting in June 2007 (Dow 2007), SC&A is of the opinion that thorium hardener was not produced at Dow (Madison), but was probably used in the production of thorium alloys at Dow (Madison). Therefore, the data presented on hardener production may be irrelevant to this site, but it is included should further evidence come to light indicating differently.

While it is questionable that thorium hardener was produced at Dow (Madison), according to worker testimony (Dow 2007), thorium hardener was used in the production of thorium alloys. However, because Silverstein 1956b describes a sampling location in Building 152, which is associated with Dow (Bay City), the data presented in this reference are likely from Bay City. Without evidence to the contrary, we believe these data are relevant to the melting process at

Dow Madison. Data on the use of hardener in melting of Mg-Th alloys are presented in Table 3 (Silverstein 1956b). The samples described in Table 3 do not involve the production of Mg-Th hardener as summarized in Table 2; rather they involve the use of the hardener in producing Mg-Th alloys, such as HK31. Samples in Table 3 were taken on settings<sup>6</sup> in the SE corner of Building 152. (Since these samples were taken in Building 152, these measurements are believed to be from Bay City.) All of the samples in Table 3, except II-3 and II-6, were taken near two settings that had local exhaust ventilation away from the operator's normal position with 90 lfm of airflow. Sample II-3 was representative of normal airflow in the melting room averaging 30 lfm. Sample II-6 was taken over a setting with no local exhaust system, but the airflow was vertical at 40 lfm. (Concentrations of radioactivity have been added in brackets in Table 3 by the authors of the current review, based on the tolerance conversion factors presented in footnote a to Table 2.)

**Table 3. Levels of Radioactivity Associated with Melting Alloys Using Mg-Th Hardener**  
(Silverstein 1956b)

Sample Number	Date	Description	Sampling Time and Volume	Analysis
II-1	5-4-56	Taken over a pot of molten Mg to which 57.2 lb of Mg-24.5% Th hardener had been added. Hardener had been made from cell Mg and ThF <sub>4</sub> 3 weeks before. 1,290°–1,400°F; normally held at 1,400°F.	5 min. (0.424 m <sup>3</sup> )	5 times tolerance for short-lived alpha material [ $5 \times 10^{-11}$ Ci/L]. No thorium (Th) or mesothorium (MsTh).
II-2	5-7-56	Taken over a pot of molten Mg at 1400°F immediately after 53.4 lb of Mg-23.7% Th hardener had been added. Pot was stirred during sampling. Hardener was 3 weeks old.	5 min. (0.424 m <sup>3</sup> )	4 times tolerance for short-lived alpha material [ $4 \times 10^{-11}$ Ci/L]. No Th or MsTh.
II-3	5-7-56	Taken near workbench 20 ft. from alloying pots. 143.4 lb of Mg-23.7% Th hardener, 3.5 weeks old, was alloyed at 1,400°F. Melt was transferred to casting pot and then to a billet mold.	6.5 hr. (33.0 m <sup>3</sup> )	5 times tolerance for short-lived alpha materials [ $5 \times 10^{-11}$ Ci/L]; 50% of tolerance for Th and MsTh. [ $0.6 \times 10^{-14}$ Ci/L for MsTh and $0.55 \times 10^{-14}$ Ci/L for Th].
II-4	4-10-56	Taken near pot in which 43.5lb of Mg-25%Th hardener (3.5 weeks old) was alloyed with Mg at 1,350°F.	4.0 min. (0.340 m <sup>3</sup> )	Short-lived alpha at tolerance [ $1 \times 10^{-11}$ Ci/L], ½ tolerance for MsTh, no Th. [ $0.6 \times 10^{-14}$ Ci/L for MsTh].
II-5	4-15-56	Taken near pot in which 29 lb of three-week old Mg-25% Th hardener and 20 lb of ten-month old Mg-27% Th hardener were alloyed with Mg at 1,330°F.	5.0 min. (0.425 m <sup>3</sup> )	Short-lived alpha 50% of tolerance [ $0.5 \times 10^{-11}$ Ci/L]. MsTh 1.8 times tolerance [ $2.2 \times 10^{-14}$ Ci/L]. No Th.

<sup>6</sup> A setting is a group of melting/alloying pots with interconnected piping that permitted the molten metal to be transferred from one vessel to another and to the continuous casting machine.

**Table 3. Levels of Radioactivity Associated with Melting Alloys Using Mg-Th Hardener (Silverstein 1956b)**

Sample Number	Date	Description	Sampling Time and Volume	Analysis
II-6	4-17-56	Over an open pot (1.5 ft above) in which 16 lb of HK31 scrap (14 months old, 3.14% Th ) was melted.	9.0 min. (0.765 m <sup>3</sup> )	Short-lived alpha 18 times tolerance [ $1.8 \times 10^{-10}$ Ci/L]. MsTh 65% of tolerance [ $0.8 \times 10^{-14}$ Ci/L].
II-7	5-3-56	Near two pots in which 142.5 Lb of Mg-25% Th (3 weeks old) were alloyed with Mg at 1,400°F	10.0 min. (0.85 m <sup>3</sup> )	Short-lived alpha three times tolerance [ $3 \times 10^{-11}$ Ci/L]. No Th or MsTh.

Additional details on the Mg-Th alloy melting process are included in the report of the AEC inspection in August 1960 (USAEC 1960). That information is excerpted as Attachment C to this report.

The Affidavits do not provide much discussion about the use of fluxes or cover gas over the melt surface to minimize oxidation. When magnesium becomes molten, it tends to oxidize (burn) and explode, unless care is taken to protect the molten metal surface against oxidation. The following is noted on one website (<http://www.key-to-metals.com/Article78.htm>):

*Molten magnesium alloys behave differently from aluminum alloys, which tend to form a continuous, impervious oxide skin on the molten bath, limiting further oxidation. Magnesium alloys, on the other hand, form a loose, permeable oxide coating on the molten metal surface. This allows oxygen to pass through and support burning below the oxide at the surface. Protection of the molten alloy using either a flux or a protective gas cover to exclude oxygen is therefore necessary.*

Worker 20 in the “Dow Employee Outreach Meeting” Affidavit indicated that SO<sub>2</sub> was used in the pot room presumably as a cover gas over the melt.

Affidavit “Pot Room Conditions” states that exhaust fans existed in the pot room, but the fans often could not keep up with the smoky conditions in the area. (The worker who made this comment began work in casting in 1960, but it did not state whether exhaust fans were in use as of that date.) This problem was at its worst when alloying additions were made. Affiant No.1 in the “Dow Employee Outreach Meeting” Affidavit stated that there were eleven 4-ft ventilation fans in the pot room. Further discussion of pot room ventilation is contained in Dow 2007, and indicates that smoky conditions were often present in both the pot room and in the casting area. Testimony presented in this meeting describes the ventilation fans as being located at high points in the buildings, often inoperable, and ineffective. Albert (1966, Chapter 10, Section 7) states that canopy hoods are used over the Mg-Th melting pots, ensuring that there are no significant levels of airborne thorium or thoron.<sup>7</sup> This presumably generic description is at odds with that in

<sup>7</sup> This conclusion is based on a 1964 personnel communication between Albert and E.R. Averill of the Continental Insurance Company of New York, NY.

the AEC inspector’s report of August 1960 (see Attachment C, Section 15). Both of these descriptions are at odds with Silverstein (1957a), who stated, “Although no local exhaust ventilation is employed, the ceilings in the alloy area are 65 feet high and the natural ventilation is excellent.”

Under Spectrulite ownership,<sup>8</sup> several explosions occurred. It is not stated whether similar explosions might have occurred in the 1957–1960 time frame (Dowpet 2006, Affidavit – “Incidences involving explosions at the Spectrulite plant”). Worker interviews included in Dow 2007 indicate that explosions were not uncommon. Silverstein (1957a) indicates that Mg fires were monitored during the period of interest for the present report.<sup>9</sup> He provides information on three fires involving alloy HK31A. In the first fire, no thorium was found in the visible fumes or in the air a short distance away (breathing zone – 1 ft from fire). The air samples were collected and analyzed by three techniques to detect the presence of thorium; spectrographic analysis, x-ray diffraction, and x-ray fluorescence. Similarly, in the second fire, thorium was not detected in the fumes over the melt or at a distance of 5 ft from the fire. The absence of thorium was again based on chemical analyses (x-ray diffraction and x-ray fluorescence). Residue from the fire contained 10%–20% ThO<sub>2</sub>, as determined by x-ray diffraction. In the third fire, measurements taken from the fume indicated the presence of  $2 \times 10^{-9}$  µCi/ml of Ra-224 and  $2.5 \times 10^{-9}$  µCi/ml of Pb-212. At a distance of 15 ft from the fire, measurements indicated  $8 \times 10^{-12}$  µCi/ml of Ra-224 and  $2 \times 10^{-11}$  µCi/ml of Pb-212. Silverstein (1956b) summarizes results of additional air-sampling measurements made during Mg-Th alloy fires. Results are summarized in Table 4. (Concentrations of radioactivity have been added in brackets in Table 3 by the authors of the current review, based on the tolerance conversion factors presented in footnote a to Table 2.)

**Table 4. Results of Air Sampling Measurements Made During a Mg-Th Alloy Fire**

Sample Number	Date	Description	Sampling Time and Volume	Analysis
IV-1	5-8-56	Area sample 15 ft from a fire of 4 lb of shavings (14 months old). No ventilation other than normal currents in Building 152.	15.0 min (1.275 m <sup>3</sup> )	Short-lived alpha three times tolerance [ $3 \times 10^{-11}$ Ci/L]. No Th or MsTh
IV-2	5-8-56	In visible fume of the fire, 2 ft above the fire.	1.0 min (0.85 m <sup>3</sup> )	Short-lived alpha activity 100 times tolerance [ $1 \times 10^{-9}$ Ci/l]. MsTh 3.5 times tolerance [ $4.7 \times 10^{-14}$ Ci/L].

Worker 13 in the “Dow Employee Outreach Meeting” Affidavit (Dowpet 2006) stated that sludge, which collected in the bottom of the melting pots, was removed and placed in barrels and boxes. During the process, material would splash on the floor and fumes would rise up into the workers’ faces, making breathing difficult. This sludge was dumped on the property outside of Building 7. From time to time, workers were sent to pick through the sludge pile and recover

<sup>8</sup> As noted in Attachment B, Spectrulite purchased the facility in 1986. The magnesium assets were subsequently sold by the bankrupt company to Magnesium Elektron North America in 2003.

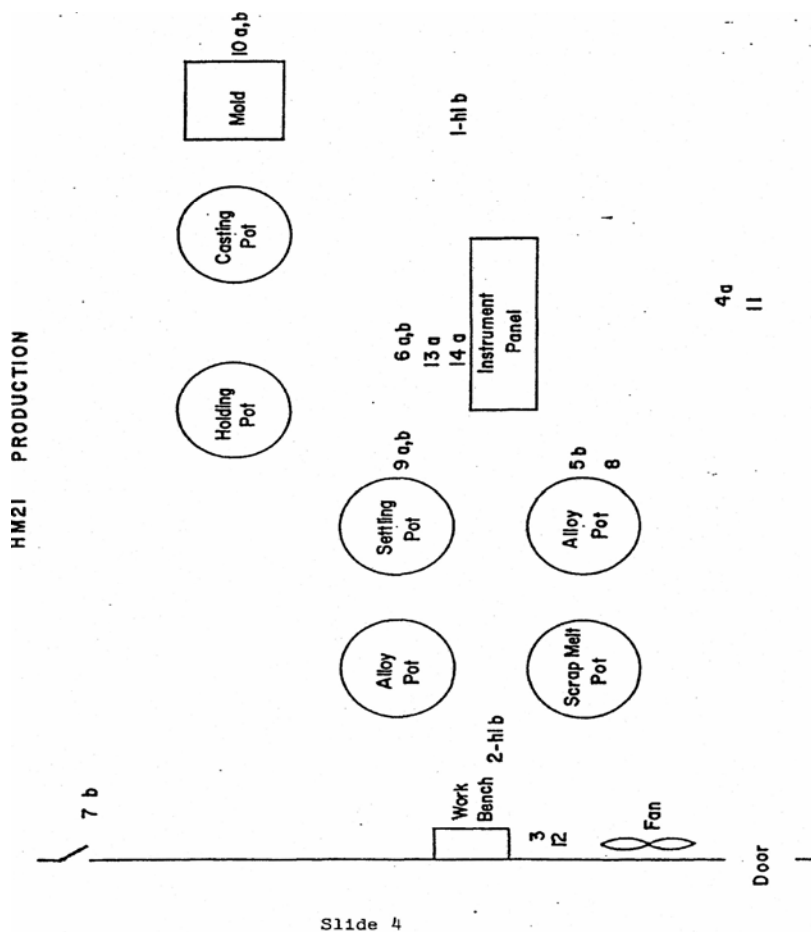
<sup>9</sup> It is not clear whether these were accidental fires that started during production operations or fires intentionally started to characterize a disposal method for Mg-Th alloy scraps and chips (Dow 1958).

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metal for remelting (Affidavit No. 2). Similar information was presented in the Dow 2007 meeting.

Silverstein (1956a, 1957a) described radioactive contamination measurements made during the production of 35,000 lb of alloy HM21. The area involved in making these measurements in Building 7 is depicted in Figure 2. The charge included cell magnesium and HM21 scrap from a previous run. After the magnesium and scrap had been melted, 190 lb of Th pellets and 150 lb of Mg-Th hardener were added to the melt. According to Silverstein (1956a), "Pots 32, 36, and 37 were used for melting in scrap and alloying. The melt was then pumped into settling pot 33, to holding pot 31, to casting pot 30. It was then pumped into the DC [Direct Chill] unit mold." Silverstein (1956a) further notes that normal ventilation conditions in the pot room were augmented by an open door and a circulating fan near Pot 36 (see Figure 2). Visible fumes rose vertically from the pots and returned to floor level 50 ft to 75 ft away.





TDCC 000379

**Figure 2. Pot Room Schematic**

**NOTICE:** This report has been reviewed for Privacy Act information and has been cleared for distribution. However, this report is pre-decisional and has not been reviewed by the Advisory Board on Radiation and Worker Health for factual accuracy or applicability within the requirements of 42 CFR 82.

The measurements made during the HM21 production run are summarized in Table 5 below.

**Table 5. Air Sampling Measurements Taken During Melting and Casting of Alloy HM21 at Dow Madison (July 22–23, 1956)**

Sample Number	Time	Description	Short-lived Radioactivity (Thoron & daughters) (Ci/L)	Radium-228 (Ci/L)	Thorium (mg/m <sup>3</sup> )
M-1	1701–1715 7-22-56 1.2 m <sup>3</sup>	Breathing zone (BZ) of man cutting open 10 drums of HM21 heels with acetylene torch. HM21 was cast into drums on 2-11-56.	$6 \times 10^{-13}$	$12 \times 10^{-14}$ (10 x tolerance)	0
M-2	1800–1820 1.7 m <sup>3</sup>	Between pots 36 & 37, 8 ft. away from either pot. HM21 scrap (last melted on 2-11-56) was being melted, some burning of metal was observed. Workers occupied this area once every 5 or 10 min for less than one minute each time.	$4 \times 10^{-13}$	$3.5 \times 10^{-14}$ (3 x tolerance)	0
M-3	1842–1904 1.87 m <sup>3</sup>	At workbench across aisle from pots 36 and 37. Workers spent most of their time here or outside door during melting down of HM21 scrap.	$5 \times 10^{-13}$	0	0
M-4	2027–2140 6.2 m <sup>3</sup>	50 ft. south of operation near another group of settings on which men were working.	$5 \times 10^{-13}$	0	0.004
M-5	2214–2222 0.68 m <sup>3</sup>	BZ of worker alloying 70# of thorium pellets into 5000# of HM21 + cell Mg in pot 32. Pellets burned before being immersed.	$6 \times 10^{-13}$	$0.27 \times 10^{-14}$	0
M-6	0036–0056 7-23-56 1.7 m <sup>3</sup>	In front of instrument panel. Pot 32 was recharged with HM21 scrap, melt was pumped from pot 37 to pot 32, and from 33 to pot 31, during sampling period. Some burning occurred during pumping and also in pot 32 when scrap was added.	$8.7 \times 10^{-13}$	$0.27 \times 10^{-14}$	0.02
M-7	0220–0250 0.425 m <sup>3</sup>	Near washroom, about 100 ft. north of the group of settings in use.	$7 \times 10^{-13}$	$0.3 \times 10^{-14}$	0
M-8	0315–0320 0.935 m <sup>3</sup>	BZ of worker alloying 50# of thorium pellets in pot 32. No burning of the pellets was observed.	$6.2 \times 10^{-13}$	0	0

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**Table 5. Air Sampling Measurements Taken During Melting and Casting of Alloy HM21 at Dow Madison (July 22–23, 1956)**

Sample Number	Time	Description	Short-lived Radioactivity (Thoron & daughters) (Ci/L)	Radium-228 (Ci/L)	Thorium (mg/m <sup>3</sup> )
M-9	0624–0635 1.87 m <sup>3</sup>	BZ of man alloying 100# of hardener into pots 31 & 32, 50# had been alloyed into pot 30 just before sampling began. Hardener was last melted on 2-7-56.	$6.2 \times 10^{-13}$	$0.44 \times 10^{-14}$	0.01
M-10	0907–0929 1.78 m <sup>3</sup>	BZ of worker attending mold during casting of HM21.	$8 \times 10^{-13}$	$0.17 \times 10^{-14}$	0.011
M-11	1007–1028 1.78 m <sup>3</sup>	By billet caster 50 ft. south of operation. 70# of pellets were alloyed in pot 32, pot 32 was pumped into pot 31, pot 36 was mechanically sludged and pot 33 was hand-sludged during sampling period.	$6.0 \times 10^{-13}$	0	0
M-12	1032–1053 1.78 m <sup>3</sup>	By workbench across aisle from pot 37. Pot 33 was pumped over to pot 31, sludge was pumped from pot 32 into cake molds.	$7.5 \times 10^{-13}$	0	0
M-13	1107–1140 2.7 m <sup>3</sup>	By instrument panel. Sludge and metal were pumped out of pots 30 and 31 into cake molds.	$3.8 \times 10^{-13}$	0	0.006
M-14	1243–1310 2.3 m <sup>3</sup>	By instrument panel. Sludge and metal were pumped out of pots 30 and 31 into cake molds.	$3.8 \times 10^{-13}$	0	0.006

In a later survey made during the production of both HM21 and HK31, Silverstein (1957a) stated that **no** long-lived alpha or long-lived beta radiation was detected, and that short-lived alpha levels were in the range of  $10^{-10}$   $\mu\text{Ci/ml}$ .<sup>10</sup>

A similar set of measurements was made during the melting and casting of HK31 on December 15-16, 1959 (Dow 1959). Details included in Attachment F indicate that concentrations of thoron and daughters were about an order of magnitude lower for the 1959 measurements. The melting log from December 16, 1959, is summarized in Table 6. Pots 34,

<sup>10</sup> Silverstein (1957) is not specific in his definitions of short- and long-lived alpha and long-lived beta. He states on page 6 that short-lived alpha samples in Table 1 displayed the characteristic half-life of radon daughter products. This suggests that Ra-224 was either not considered to be one of the short-lived alpha products or it was not released in measurable quantities. It can be inferred from Silverstein's paper that Th-232 and Th-228 are the principle sources of long-lived alpha. It can also be inferred that Ra-228 is the primary source of long-lived beta. See Attachment E for details on the Th-232 decay chain.

35, 38, and 39 are not shown in Figure 2. It was noted in the melting log that the sludge recovery centrifuge operated on a 32-minute cycle throughout the day of December 16.

**Table 6. Melting Log for Alloy HK31**  
(December 16, 1959)

Time	Action
9:00	Add 38 lb sheet Th to Pot 38
10:00	Add 40lb Mg-Th master alloy to Pot 37
10:35	Add 65 lb of Th pellets to Pot 35
11:00	Clean (sludge) Pot 37
13:24	Pump Pots 32 and 33
13:26	Charge Pot 32
13:34	Charge Pot 37
14:00	Charge Pot 36
14:10	Add 95 lb Th pellets to Pot 38
14:25	Add 50 lb Th scrap to Pot 37
??	Add 9 lb Th to Pot 32

Silverstein (1957a) also described some measurements of radioactivity in samples taken over a pot during the melting of alloy HK31 at 1,400°F as a function of time between initial melting of the alloy and remelting of the scrap from the initial melt. Regardless of the age of the alloy, significant levels of activity from short-lived alpha emitters were measured, while no long-lived alpha emitters were detected above the pot for any of the samples. This suggests that Th-232 and Th-228 with half-lives of  $1.4 \times 10^{10}$  yr and 1.91 yrs are not transferred out of the melt, based on the detection limits existing in 1957. (The Th-232 decay scheme is included as Attachment E.) Activity over the melt from short-lived alpha emitters was reported by Silverstein to range from 0.5 to  $18 \times 10^{-7}$  Ci/m<sup>3</sup>. It is not known what short-lived alpha emitters were actually evolved during the melting process. The potential suite includes Ra-224, Rn-220, Po-216, Bi-212, and Po-212. It is likely that Rn-220, which is a gas, would evolve from the melt. Some fraction of the Po, which has a vapor pressure of  $10^4$ Pa at 1,346° F, might also be vaporized (<http://en.wikipedia.org/wiki/Polonium>).

Mechanisms for removal of the other short-lived alpha emitters (Ra and Bi) are not obvious. As shown in Albert 1966, Figure 2.6, the Th-228 decay chain reaches equilibrium after about 21 days, so that aging of the alloy for longer periods before remelting should not significantly alter the quantity of short-lived alpha emitters present. Samples of Mg-Th alloy aged 10 to 14 months before remelting showed some increase in the quantity of radioactivity from long-lived beta emissions. This may be the result of in-growth of Ra-228 from Th-232 decay (see Figure 3). Some of the Ra-228 could then escape from the melt and is captured in the sampler above the melting pot. However, the mechanism for such possible Ra emissions from the melt is not apparent. The information presented in this section indicates that Th-232 and Th-228 are not present in significant quantities in the air in the melting and casting area. Measurements

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provided in Table 5 indicate the presence of 4 to 20  $\mu\text{g Th}/\text{m}^3$  of air in those instances where long-lived alpha radiation is detected, but often no long-lived alpha is observed.<sup>11</sup>

### 2.3 Alloy Fabrication

Some of the Mg-Th alloys cast in Building 7 were fabricated into sheet via multiple passes on the rolling mills in Building 5. Apparently, up to seven mills were used with plate produced on mill #1 and sheets of various thickness produced on the other mills. At several stages in the size reduction process, the sheets were sanded to remove gouges or dirt and scale. Pickling was also used for surface cleaning (Dowpet 2006, Affidavit No. 2). Worker 15 in the “Dow Employee Outreach Meeting” Affidavit stated that on mill #1, there was no airborne dust. He opined that any generated dust would have been smothered by the mill coolant.

Measurements of dust levels from various fabrication operations associated with the production of Mg-Th alloys at Dow are presented in Figure 3 based samples collected with an electrostatic precipitator and analyzed spectrographically for thorium (Albert 1966).<sup>12</sup> Thorium concentrations as high as  $53.4 \mu\text{g}/\text{m}^3$  were measured in the dust from rotary filing operations. According to Silverstein (1957a), this high value was probably due to a large particle (called a brick) that was dislodged from the metal surface. He stated that such a particle would be of sufficient size as to not be breathed by the operator. In contrast, thorium concentrations of about  $1 \mu\text{g}/\text{m}^3$  were measured for rolling operations. Silverstein characterizes the samples used for Figure 3 as follows:

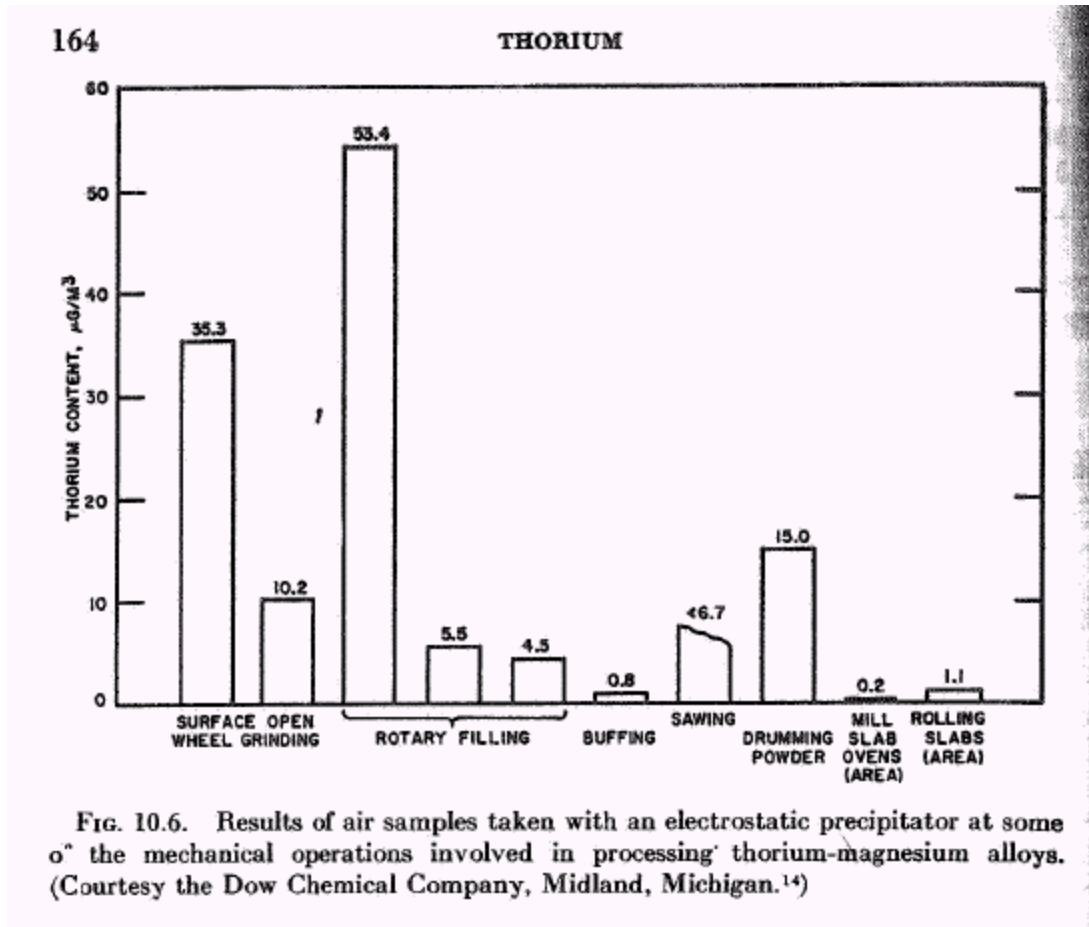
*All were breathing zone samples and the operation took place on equipment that is designed for safe operation with magnesium which means local exhaust and water traps to collect the magnesium fines,*

It should be noted in Figure 3 that the “Mill Slab Ovens” sample and the “Rolling Slabs” sample are based on area air samples, while the other samples in the figure are breathing-zone samples.

For thorium that is freshly chemically separated, Th-232 and the Th-228 would probably be in radioactive equilibrium. Thus, for a dust sample containing  $53.4 \mu\text{g}/\text{m}^3$  of newly-separated thorium, the equivalent activities for Th-232 and Th-228 would be  $5.9 \text{ pCi}/\text{m}^3$ . As shown in Figure 4 (Albert 1966, Figure 2.5), during the initial time period after separation, the rate of ingrowth of Th-228 from Ra-228 is less than the rate of decay of Th-228, and the Th-228 activity declines relative to that of Th-232, reaching a minimum of about 40% after about 6 years. After that, the ratio of Th-228 to Th-232 activity begins to increase and the two radionuclides are again in equilibrium after about 55 years.

<sup>11</sup> Based on the fact that the mass ratio of Th-228/Th-232 is  $1.34 \times 10^{-10}$  at equilibrium and the specific activities for Th-232 and Th-228 are  $1.1 \times 10^5$  and  $8.2 \times 10^{14}$  pCi/g, respectively, the Table 5 measurements can be converted to a range from 0.44 to 2.2 pCi/m<sup>3</sup> for Th-232 and for Th-228.

<sup>12</sup> This same figure appears in Silverstein (1957) as Slide V, and in compliance report\_0001.PDF as Exhibit B.



**Figure 3. Results of Air Samples Taken During Processing Thorium-Magnesium Alloys**  
(Albert 1966)

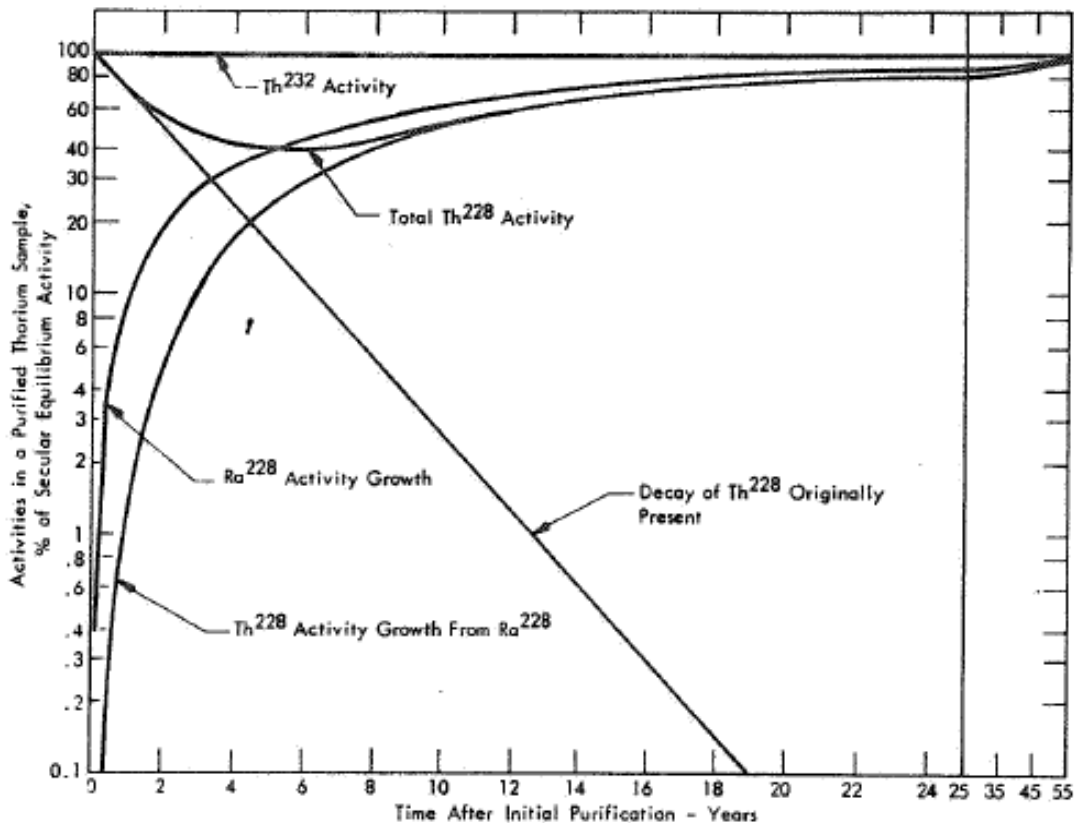
It is expected that dust particles collected for the measurements presented in Figure 3 would be magnesium particles with the intermetallic phase  $Mg_{23}Th_6$  precipitated within the grains and at the grain boundaries of the Mg crystals (see Attachment D for details).

Results from three air samples taken on March 14, 1957, in the operator's breathing zone during the hand-sanding of HK31 were as follows (Hoyle 1957):

- < 3.5 µg/m<sup>3</sup> Th
- < 7 µg/m<sup>3</sup> Th
- 21 µg/m<sup>3</sup> Th

These values are well below the thorium "tolerance limit" of 100 µg/m<sup>3</sup> used by Dow in 1957.<sup>13</sup> (See also Attachment F, Section F.11.)

<sup>13</sup> It appears that this level was reduced to 76 µg/m<sup>3</sup> by 1959 (Levy 1959).



**Figure 4. Variation of Activity in Natural Thorium after a Single Purification**  
 Courtesy of Union Carbide Corporation, Oak Ridge, Tennessee (Albert 1966)

Two additional air samples were collected on Millipore® filters on December 2, 1957, during the hand-sanding of HK31 with small air-operated vibratory sanders (Mitchell 1957). The sample showed significant alpha activity when counted on December 19, 1957. Assuming that all of the alpha activity was from Th-232, Silverstein (1957b) estimated that the thorium concentrations were 138 and 165  $\mu\text{g}/\text{m}^3$ . However, Silverstein opined that the thorium activity was less than 50% of the total, and said that spectrographic analyses were being conducted to verify the assumption. Although results of these spectrographic analyses were not included in the available documents, the Dow 1959 *Annual Industrial Hygiene Survey* (Levy 1959) quotes values of 18–36  $\mu\text{g}/\text{m}^3$  for vibrator-sanding of HK31 during December 1957. It is highly likely that these are the thorium contributions to the total alpha activities measured on December 19, 1957.

As described by Worker 21 in the “Dow Employee Outreach Meeting” Affidavit, the extrusion facility (Building 6) had six presses; two rated at 1,800 tons, two rated at 3,000 tons, one rated at 5,500 tons, and one rated at 11,500/13,500 tons. The affidavits provide very few details about the extrusion process, e.g., alloys, shapes and sizes, surface clean-up, etc. Alloy HK was extruded on the “heavy press” after 1961 (Affidavit No. 8), but it is not clear whether extrusions of Mg-Th alloys were made prior to 1961. The final affidavit (Worker 21, pg. 11) indicates that extrusion work on some (unspecified) alloys began in 1953. A worker in the Dow 2007 meeting

indicated that thorium alloys were being tested in 1953, as well. Other workers have indicated that thorium alloys were produced as late as the late 1980s (Dow 2007).

Pickling (chemical milling) was used to an unknown extent for surface treatment of mill products (Silverstein 1957a). The pickling tank was equipped with slot ventilation drawing about 50 lfm. Results from two samples taken during the processing of HK31 are presented in Table 7.

**Table 7. Air Sampling During the Chemical Milling of HK31**

Sample	Analysis
4 inches above tank in which HK31 was being acid pickled	$6.6 \times 10^{-11}$ Ci/m <sup>3</sup> short-lived alpha; no long-lived alpha or beta.
5 feet from pickle tank	$4.5 \times 10^{-11}$ Ci/m <sup>3</sup> short-lived alpha; $2 \times 10^{-12}$ Ci/m <sup>3</sup> long-lived beta; no long-lived alpha.

According to worker interviews (Dow 2007), pickling tanks were used in Building 6 and Building 7 at the Dow (Madison) plant.

With regard to uranium fabrication, Worker 14 in the “Dow Employee Outreach Meeting” Affidavit stated that the work for Mallinckrodt was done on the Number 7 press. Other workers (Dow 2007) have indicated that uranium rod straightening took place in Buildings 5 (rolling mill) and Building 6. Mallinckrodt provided their own handlers and operators, and they would “pretty much clean up their mess after they were finished.” Apparently, Dow workers were present, but not active participants. Subsequently, as part of FUSRAP clean-up activities at the Madison plant, ORNL conducted a survey in March 1989 of Building 6, where extrusion work was done. This survey was in response to uranium extrusion and rod straightening work done for the AEC by Dow under a 1957 contract (extrusion)<sup>14</sup> and a 1960 purchase order (rod straightening) from Mallinckrodt Chemical Company (Cottrell and Williams 1990). Eighteen dust samples were collected from overhead beams in Building 6 and analyzed for Ra-226, Th-232, and U-238. Ranges for the results are as follows:

- Ra-226: 0.22 to 1.3 pCi/g
- Th-232: 0.48 to 7.8 pCi/g
- U-238: 3.7 to 310 pCi/g

In spite of the fact that only limited processing of uranium was done in Building 6, while Mg-Th alloys had been processed there for more than 30 years, the uranium contamination was far greater. This observation is consistent with other evidence presented in this report indicating that thorium mobility is low. Based on the sampled area (about 200 cm<sup>2</sup>), the dust measurements were converted to dpm/100 cm<sup>2</sup> and compared with DOE guidelines. The Ra-226 contamination ranged from 40% to 46% of the DOE guideline, and averaged about 43%. The Th-232 contamination ranged from 9% to 100% of the guideline, and averaged 45%. The U-238

<sup>14</sup> The contract (No. 25034-M, March 15, 1957) provided for 12 monthly sessions of 28 hours each, including 6-hr setup, 16 hrs of extrusion, and 6 hrs for clean-up (Cottrell and Williams 1990). It is not known whether all 12 sessions took place.



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contamination ranged from 10% to 1,360% of the guideline, and averaged 250%.<sup>15</sup> The Cottrell and Williams report states that the Th-232 contamination found throughout the building is the result of separate licensed processes not connected with DOE (AEC) activities.

Silverstein (1957c) observed that, based on a film badge survey, the average weekly external exposure to workers involved in a variety of production operations including foundry activities, inspection, and shipping was less than 10 mR with the maximum exposure being about 30 mR. It is possible that the author is referring to the results documented in Appendix F, Table F-5, since these are the only film badge data provided by Dow to NIOSH.

External radiation exposure has been measured for a 2,000 lb<sup>16</sup> stack of Mg-3.5% Th alloy (HK31A) slabs (40 in by 30 in by 7 in). The  $\beta/\gamma$  dose at the surface was 4.2 mR/hr, 1.8 mR/hr at 1 ft, and declined to 0 at a distance of 7 ft from the alloy stack (Albert 1966, Figure 10.5; Silverstein 1957a, Slide 1). Dow estimated that a worker with hands in contact with and body 1 ft from the alloy stack would receive an exposure to the hands of 168 mR/week and exposure to the body of 72 mR/week, based on a 40-hr week (Silverstein 1956c). These external exposures were less than the existing NCRP recommendation of 500 mR/week to the hands and 100 mR/week to the body (Dow 1957). Dow noted that similar results were obtained on a stack of HK31 sheets, and observed that the beta component of the radiation was eliminated after crating (Silverstein 1957a). Details are provided in Attachment F, Section F.12

Thorium pellets measured 35 mR/hr on contact and Dow estimated that, at the normal working distance from the pellets, the worker's exposure was less than 1mR/hr (Silverstein 1956a). In addition, Dow estimated that a single worker handling all the thorium pellets required for a 100,000 lb run of Alloy HK31 would receive an external exposure of 53 mR (Dow 1957, Levy 1957). Based on two to three such runs per year, the annual exposure would be 0.10–0.16 R (Levy 1957).

Sludges remaining after melting Mg-Th alloys exhibited radiation levels of about 200 mR/hr at contact, but within a month, the radiation level had dropped to 10–20 mR/hr (Silverstein 1957a). HK21 scrap showed a maximum of 5 mR/hr at contact (Silverstein 1956a). Other measurements of external exposure are included in Attachment F (Sections F.8 [sludge dump], and F.9 [Th storage area]).

The annual production of Mg-Th alloys is unknown, based on the available information. One worker involved in the manufacture of shipping crates estimated that every 2 months, Dow shipped four truckloads of metal to Rocky Flats, with each truck-load being about 36,000 lb (Affidavit No. 6).<sup>17</sup> This is equivalent to 864,000 lb of alloy annually. (It is not known what Rocky Flats did with this large amount of material. Ulsh et al. 2006 sheds no light on this question. Given the information presented in Attachment G, shipments of such a large quantity

<sup>15</sup> The DOE guidelines for total residual averages were 100 dpm/100 cm<sup>2</sup> for Ra-226, 1,000 dpm/100cm<sup>2</sup> for Th-232, and 5,000 dpm/100 cm<sup>2</sup> for U-238 (Cottrell and Williams 1990, Table 1).

<sup>16</sup> According to Silverstein 1957c, the stack weighed 3,000 lb.

<sup>17</sup> The worker who made this statement was employed at the Dow Madison plant from the early 1960s through the early 2000s.

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of Mg-Th alloys to Rocky Flats appear questionable.) Assuming that the alloys shipped included equal quantities of HK31 and HM21, then the average thorium content would be 2.5%. Thus, these alloys would contain a total of 21,600 lb of Th. Other workers indicated that thorium alloys were shipped to Oak Ridge and Los Alamos, as well as contractors in the aerospace and defense industries (Dow 2007).

An alternative guesstimate of Mg-Th production can be obtained from the August 1960 AEC Compliance Inspection Report, which indicates that as of July 1960, 80 tons of thorium had been used at the foundry (USAEC 1960). Assuming that this usage was over a 3 1/2-year period beginning in 1957, annual usage would have been about 23 tons. At an average Th content of 2.5%, annual Mg-Th alloy would be about 920 tons (1,840,000 lb). This estimate seems high, since a 1957 Dow document (Levy 1957) estimates external exposures from two to three 100,000 lb casting runs of Mg-Th alloys per year. Presumably, this represented production expectations at the time.

Based on the information presented in Attachment A, the use of Mg-Th alloys has declined significantly since 1957–1960. By 1973, only about 5,000 kg of thorium oxide (9,700 lb of Th) was used in alloys for the **entire** aerospace industry.

## 2.4 Sampling Techniques (circa 1956)

### 2.4.1 Thorium Metal

According to Silverstein (1956c), dust and fume was analyzed for thorium with a Mine Safety Appliance Company Electrostatic Sampler Model F using aluminum tubes. The thorium metal or oxide is recovered by dissolution in 75–80 ml of 10% nitric acid. The solution is evaporated to dryness and the residue is redissolved in nitric acid. A 0.03-ml aliquot of the sample is analyzed using the thorium line at 4019Å on a Baird 3-meter grating spectrograph with an Ennis-type spark source.

### 2.4.2 Alpha and Beta Activity

Silverstein (1956c) describes the sampling process for alpha and beta activity as follows:

*...air samples were collected with an electrostatic precipitator and counted for alpha and beta activity in an NMC model PC-1 proportional counter. The counter efficiency for alpha was determined by counting a calibrated alpha source. Beta efficiency was assumed to be 50%, which is too low by an unknown backscattering factor. This error is on the safe side since reported beta counts are higher than actual counts.*

Silverstein (1957a) also noted that Millipore filter paper was used to a “small extent.” The samples taken in 1959, described in Table 5 above, also used Millipore filters.

This description of the air-sampling techniques would indicate that measurements reported as short-lived alpha activity were not limited to thoron (which would not be readily collected using

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these methods, because thoron is a noble gas), but would likely be comprised of the short-lived progeny of thoron, which are of a particulate nature and collected by the cited sampling methods.

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### 3.0 INTERNAL DOSIMETRIC IMPLICATIONS OF THORIUM OPERATIONS

SC&A's review of thorium operations and associated radiological data, as described in Section 2.0, reveal that we have a fairly complete understanding of the types of operations that took place between 1957 and 1960, and presumably this understanding is also applicable to post-1960 operations. However, until we have access to records for the post-1960 time period, this presumption is unverified.

With respect to radiological data pertinent to reconstructing internal exposures, it is clear that the data are limited to a few air samples; however, these data are informative. The following sections present the ranges of airborne concentrations of Th-232 and its progeny observed during various aspects of the alloying operations, along with some discussion of the dosimetric implications of these measurements.<sup>18</sup>

#### 3.1 Mg-Th Hardener Production

The first set of data is provided in Table 2 and, as discussed in Section 2.2, these data apply to hardener production, which we believe took place at Bay City and not Dow Madison. Nevertheless, it is instructive to explore the implications of the measurements reported in Table 2.

Four air samples of 5- to 10-min duration were collected in the immediate vicinity of the pots where hardener production took place, and also in the general work area. The thorium activity (which is assumed to be 50% Th-232 and 50% Th-228) was observed in one sample to be as high as  $1.1 \times 10^{-13}$  Ci/L, and in other samples, as low as about  $1.1 \times 10^{-15}$  Ci/L. To put these concentrations into perspective, it is useful to note that the 50-year committed effective dose equivalent (CEDE) for 1 micron AMAD, Absorption Type S aerosols of Th-232 and Th-228 are  $1.1 \times 10^{-4}$  Sv/Bq and  $4 \times 10^{-5}$  Sv/Bq, respectively. Hence, the high end 50-year CEDE for Th-232 is derived as follows:

$$D = 1.1 \times 10^{-13} \text{ Ci/L} \times 1.1 \times 10^{-4} \text{ Sv/Bq} \times 3.7 \times 10^{10} \text{ Bq/Ci} \times 1.2 \text{ m}^3/\text{hr} \times \\ 1000 \text{ L/m}^3 \times 1.0 \times 10^5 \text{ mrem/Sv} \div 2 = 27 \text{ mrem/hr of exposure}$$

For air samples collected in the general work area away from the pots, the airborne levels of thorium were observed to be about 1/100<sup>th</sup> of the observed concentration near the pots. At these locations, the dose rates would be about 100-fold lower.

Because of the limited number of air samples and the fact they may not represent breathing zone samples, it is difficult to draw conclusions regarding these particular measurements. However, it would seem that the potential for internal exposures to Th-232 and Th-228 in localized areas in the immediate vicinity of the pots where hardener production took place could be high, at least

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<sup>18</sup> Only the data presented in Section 2.0 are used in this analysis. More data are contained in Attachment F. While the ranges differ slightly from the data in Section 2.0, the conclusions drawn considering these data would be the same.

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during the times when operations took place. Alternatively, it also appears that in the general working area (i.e. the work bench area), the potential for exposure to Th-232 and Th-228 was low.

Table 2 also presents concentrations of Ra-228. The dosimetric implications of these measurements are as follows. The highest concentration of Ra-228 reported in Table 2 is  $1.2 \times 10^{-13}$  Ci/L in the immediate vicinity of the pots. The 50-year CEDE for Ra-228 is  $1.7 \times 10^{-6}$  Sv/Bq. Hence, the highest dose rate associated with the values reported in Table 2 is as follows:

$$D = 1.2 \times 10^{-13} \text{ Ci/L} \times 1.7 \times 10^{-6} \text{ Sv/Bq} \times 3.7 \times 10^{10} \text{ Bq/Ci} \times 1.2 \text{ m}^3/\text{hr} \times \\ 1000 \text{ L/m}^3 \times 1.0 \times 10^5 \text{ mrem/Sv} = 0.9 \text{ mrem/hr}$$

In the general work area, the Ra-228 concentrations were somewhat lower. The implications are that exposures to Ra-228 were relatively low, as compared to Th-232 and Th-228. Table 2 also reports the presence of short-lived alpha emitters at relatively high concentrations. These radionuclides could include Ra-224<sup>19</sup> (3.66 day half-life) and its progeny, which includes thoron (55.6 sec half-life) and its short-lived progeny. In the vicinity of the pots, the highest concentration of short-lived progeny of  $3 \times 10^{-8}$  Ci/L was observed. At the work bench area, the concentrations were observed to be about 60 to 375 times lower. Assuming that the short-lived progeny includes Ra-224 and all its progeny, a bounding dose from these isotopes could be estimated by assuming that 25% of the observed alpha activity was from Ra-224, and the remainder of the activity was from thoron and its progeny. This is considered bounding because Ra-224 has a much lower potential to become airborne than thoron during hardener production, and Ra-224 has a much higher dose conversion factor than thoron and its progeny, primarily because of its much longer half-life. On this basis, the following is an estimate of the dose rate (50-year CEDE) associated with the high-end concentrations of short-lived alpha emitters observed near the pots (assuming the presence of Ra-224):

$$D = 3 \times 10^{-8} \text{ Ci/L} \times 3.4 \times 10^{-6} \text{ Sv/Bq} \times 3.7 \times 10^{10} \text{ Bq/Ci} \times 1.2 \text{ m}^3/\text{hr} \times 1000 \text{ L/m}^3 \\ \times 0.25 \times 1 \times 10^5 \text{ mrem/Sv} = 4.5 \times 10^5 \text{ mrem/hr}$$

This is an extremely high dose rate. It is important to recognize that the short-lived alpha emitters may have been entirely thoron and its progeny, which have much lower inhalation dose conversion factors. For example the 50-year CEDE for 1 micron AMAD Pb-212 and Bi-212 (the major contributors to the dose associated with inhaling thoron and its progeny) are  $1.9 \times 10^{-8}$  and  $3.1 \times 10^{-8}$  Sv/Bq. Therefore, if the short-lived progeny measurements reported in Table 2 consisted primarily of thoron and its short-lived progeny, the dose rate, as calculated above, would be lower by about 100-fold. However, even this is an extremely high dose rate.

Inspection of Table 2 reveals that the concentrations of short-lived alpha emitters were observed to be about 100 times lower in the work-bench area as compared to the measurements taken at

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<sup>19</sup> Section 2 of this report provides information indicating that Ra-224 was not present in significant quantities in the short-lived progeny analyses. However, for the sake of completeness, this section includes an evaluation of the magnitude of the exposures if a significant portion of the short-lived alpha analyses included Ra-224.

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the hardener production pots. Hence, exposure potential is further reduced by about a factor of 100 to about 10 mrem/hr. This may still be a significant dose rate.

Notwithstanding the limited amount of data and the considerable uncertainties, there appears to be a high potential for exposures associated with hardener production. However, it must be kept in mind that hardener production likely took place at the Bay City facility and not at Dow Madison.

### 3.2 Melting of Alloys Using Mg-Th Hardener

Section 2.2 also presents air-sampling data collected during the melting of alloys using Mg-Th hardener. While these data are believed to have been generated at Bay City, hardener was used at Dow Madison. A 6.5-hour air sample collected at this location observed Th-232 and Ra-228 concentrations of  $0.55 \times 10^{-14}$  Ci/L and  $2.2 \times 10^{-14}$  Ci/L, respectively (see Table 3). These concentrations correspond to a 50-year CEDE rate of about 1 mrem/hr for Th-232 and about 0.2 mrem/hr for Ra-228.

For short-lived alpha, the concentrations were on the order of  $1.8 \times 10^{-10}$  Ci/L. Assuming this is predominately thoron and its short-lived progeny, the dose rate (50-year CEDE) was on the order of the following:

$$D = 1.8 \times 10^{-10} \text{ Ci/L} \times 3.1 \times 10^{-8} \text{ Sv/Bq} \times 3.7 \times 10^{10} \text{ Bq/Ci} \times 1 \times 10^5 \text{ mrem/Sv} \times 1.2 \text{ m}^3/\text{hr} \times 1000 \text{ L/m}^3 = 25 \text{ mrem/hr}$$

The implications are that the potential for exposure to Th-232 and Th-228 appears to be quite low. However, the potential exposure rate from thoron and its progeny was not insignificant.

### 3.3 Mg-Th Alloy Fire

As discussed in Section 2.2, explosions and fires were not uncommon at magnesium alloying facilities, but Th-232 and Th-228 were not detected in the fumes. However, as indicated in Table 4, short-lived alpha emitters were observed at concentrations as high as  $1 \times 10^{-9}$  Ci/L, and Ra-228 was observed at concentrations on the order of  $4.7 \times 10^{-14}$  Ci/L. These concentrations correspond to dose rates on the order of 140 mrem/hr CEDE and <1 mrem/hr, respectively. Also, Ra-224 and Pb-212 were measured in the fume at concentrations of about  $2 \times 10^{-9}$   $\mu\text{Ci/ml}$  ( $2 \times 10^{-12}$  Ci/L). This corresponds to dose rates (50-year CEDE) on the order of 30 mrem/hr for Ra-224 and <0.17mrem/hr for Pb-212. At a distance of 15 ft from the fire, the concentrations were observed to be lower by several orders of magnitude. The implications are that the potential for significant exposures were relatively small at the time of fire and explosions at Mg-Th alloying facilities, considering the magnitude of the observed concentration of thorium and its progeny and the relatively short duration of exposures during such incidents.

### 3.4 Alloy Production

Table 5 summarizes data characterizing the airborne concentrations of short-lived progeny, Ra-228, and Th-232/Th-228 during alloy melting. In general, the breathing-zone concentrations

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of Th-232/228 were below the limits of detection, but some values as high as 0.02 mg/m<sup>3</sup> were observed. This corresponds to about 0.08 Bq/m<sup>3</sup> or about  $2 \times 10^{-15}$  Ci/L. This concentration corresponds to a dose rate (50-year CEDE) of <1 mrem/hr. Hence, it appears that the potential for exposure to Th-232 and Th-228 was extremely small during Mg-Th alloy melting and casting.

The breathing-zone concentrations of thoron and its progeny were observed to be on the order of  $1 \times 10^{-12}$  to  $1 \times 10^{-13}$  Ci/L. This corresponds to a dose rate of <1 mrem/hr. The breathing-zone concentration of Ra-228 was observed to be on the order of  $5 \times 10^{-15}$  Ci/L, but one sample was observed to be on the order of  $1 \times 10^{-13}$  Ci/L. The high-end concentration was observed for a worker cutting open a drum of alloy with an acetylene torch—an operation of 10 min duration (Silverstein 1957a). These values correspond to doses on the order of <<1 mrem/hr to about 1 mrem/hr. Notwithstanding the limited data and associated uncertainties, it appears that the potential for exposure during alloying operations was small.

Section 2 also discusses the results of the analysis of air samples collected over a pot during melting of alloy HK31 at 1400°F (see Table 4). The analyses were unable to detect the presence of long-lived alpha emitters (i.e., Th-232/Th-238); however, short-lived alpha emitters (thoron and its progeny) ranged in concentrations from  $0.5 \times 10^{-7}$  to  $18 \times 10^{-7}$  Ci/m<sup>3</sup> ( $0.5 \times 10^{-10}$  to  $18 \times 10^{-10}$  Ci/L). These are high concentrations and are associated with doses on the order of tens to hundreds of mrem/hr CEDE if the exposures were sustained for such time periods.

### 3.5 Alloy Fabrication

Alloy fabrication consists of a broad range of mechanical operations, such as Mg-Th rolling. Thorium dust loadings ranging from 0.2 to 53.4 µg/m<sup>3</sup> ( $6.9 \times 10^{-17}$  Ci/L to  $1.8 \times 10^{-14}$  Ci/L) were reported by Albert (1966). Such concentrations are associated with dose rates ranging from <<1 mrem/hr to about 5 mrem/hr. As discussed in Section 2, the measured dust loading of 53.4 µg/m<sup>3</sup> is believed to be due to the presence of large, non-respirable particles, referred to as “bricks.” However, other measurements reported in Section 2 range from 18–36 µg/m<sup>3</sup>. As such, it appears that alloy fabrication operations have the potential to result in several mrem/hr (CEDE).

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## 4.0 FOLLOWUP ON THORIUM EXPOSURES AFTER 1960

At the Advisory Board Meeting held in Denver, Colorado, on May 2, 2007, SC&A was tasked to assess thorium exposures past the time period covered by the SEC petition (post-1960). In addressing this task, SC&A considered the material it had reviewed for accomplishing the reviews of the SEC petition and the NIOSH evaluation and documents made available on the “O” drive on May 1, 2007, from Dow Chemical Company. The relevant information from these sources is presented in Sections 1.0 and 2.0.

With the understanding of the Advisory Board, SC&A did not attempt to obtain information from sources other than NIOSH or that distributed to both NIOSH and SC&A by the SEC petitioners and their representatives. As recognized by the Advisory Board, SC&A did not independently request or obtain information from any other source. Therefore, the content of this report is based on the knowledge obtained from the named sources and may not represent the full universe of information available from all relevant sources. With the exception of the materials from the workers’ meetings and affidavits (Dowpet 2006, Dow 2006a, Dow 2006b, Dow 2006c, and Dow 2007), the products on the “O” drive concentrate on the 1957 through 1960 time frame. Therefore, most of the information for this discussion of thorium beyond 1960 is extracted from the four workers’ meetings and affidavits filed with the petition. A subset of these affidavits related to thorium was provided to NIOSH by Simmons-Cooper on April 16, 2007, and is available on the “O” drive.

### 4.1 June 20, 2007, Workers Meeting

The available documentary information focuses on the time frame of 1957 through 1960. The primary available source of post-1960 information on thorium alloy production and the attendant exposures is contained in the written records from the three Dow (Madison) workers’ meetings and the affidavits submitted with the SEC petition. Therefore, SC&A believed that we should attempt to more clearly define the particulars of thorium alloy production and exposure by meeting with Dow (Madison) workers to elicit information on their experiences in greater detail.

Twenty-three former workers participated in the meeting and one offered testimony via the telephone. The workers represented a wide range of jobs in all departments of the plant, including, casting, extrusion, rolling mill, shipping, and laboratory testing.

It should be recognized that workers’ testimony has both assets and liabilities relative to written documents when establishing a record of facts for events which occurred, in some instances, over 50 years previous. On the asset side, there is some information that was never recorded or not now available that is only known to the workers. On the liability side, the recollections of workers for facts so far into the past can be flawed or altered by events or information gained since the time of the events. In addition, the workers were not privileged to some important information, particularly that pertaining to work involving radioactive materials, including thorium. That having been said, the workers knowledge of the facts relative to thorium alloy production at Dow (Madison) is a key element in understanding the exposure conditions that existed at the time of occurrence.

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With the assistance of [Name redacted] and Simmons-Cooper, LLC, Chick Phillips of SC&A, accompanied by Grady Calhoun, NIOSH, met with former Dow (Madison) workers on June 20, 2007, in Alton, Illinois. The meeting was held at the offices of Simmons-Cooper, LLC, the law firm supporting the petitioners' SEC application. Simmons-Cooper provided the services of a court reporter, and the verbatim proceedings were made available to SC&A and NIOSH (Dow 2007). Most of the information was provided by the workers, most of which was contained in the previous public meetings and the affidavits from the SEC application. However, some workers participating in the June 2007 meeting had not attended the public meetings, nor had they provided affidavits.

This meeting was conducted with the following objectives:

- Gaining a better understanding of the Dow Chemical operations relative to thorium and thorium alloy production, and the exposure conditions related to them
- Identifying any thorium materials that may have been provided to DOE/AWE sites outside the 1957 through 1960 time frame
- Identifying any radioactive materials, including thorium, that may have been processed at the Dow (Madison) site
- Allow workers to present any information relevant to the SEC petition

The format of the June 2007 workers' meeting was intended to be topical, in order to address the objectives above. Each worker described their knowledge of thorium-related activities, including those related to the shipping of thorium to other DOE/AWE sites. They responded to questions from the SC&A representative, [Name redacted], Mr. Calhoun, and each other. The participating workers were encouraged to present any information that they believed to be relevant. The transcript from the meeting was reviewed upon its receipt on July 9, 2007. The meeting was very useful in clarifying previously available information related to the production and distribution of thorium alloys, which had been provided by workers in prior workers' meeting and affidavits. However, no significant new facts regarding thorium alloy production, worker exposure conditions, processing of radioactive materials supplied by others, or distribution of thorium alloy products were offered. Points of emphasis for the workers included the following:

- The absence of radiation monitoring or protective measures during the Dow era
- The lack of knowledge imparted to the workers on the nature of thorium (or other radioactive) materials
- The distribution of thorium alloys to other DOE/AWE sites (Mallinckrodt, Rocky Flats, Oak Ridge, and Los Alamos – with particular emphasis on Rocky Flats)
- The provision of thorium alloys to aerospace and defense contractors

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- The processing of presumably radioactive products from outside entities (DOE/AWE sites and aerospace/defense contractors)
- The amount of airborne material present in casting and extrusion buildings during thorium alloy production
- The large amount of overtime worked
- The degree to which workers moved around the plant in different jobs

#### 4.2 Thorium Alloy Production and Distribution to DOE/AWE Sites

Thorium alloys, namely HK31 and HM21, were produced by Dow Chemical Company and its successor companies at the Madison, Illinois, plant for a number of years. The length of time over which they were produced is not entirely clear. The site companies have had radioactive materials licenses allowing the possession of thorium from 1956 to the present. Documents of affidavits filed with the SEC application and from workers' meetings indicate a varying estimate, which is understandable, since the terms of their employment vary (Affidavits 2006, Dow 2006a, Dow 2006b, Dow 2006c, Dow 2007):

- "I was employed at the Dow Chemical plant from 1962 to 1996. .... Thorium was run periodically at the Madison plant throughout the years I worked there."
- "Thorium was run at the plant from 1957 to 1999."
- "I was employed at the Dow Chemical plant from 1960 to 1987 .... Thorium metal was being cast, extruded and rolled at the Madison Plant during my entire period of employment."
- "I was employed at the Dow Chemical plant from 1964 to 2002. ... I know that Thorium metal was being processed in (redacted) throughout the years I was employed at the Madison plant."
- "I was employed at the Dow Chemical plant from 1953 to 1995. Thorium ran periodically through the (redacted) at the Madison plant during this entire period."
- "I worked at Dow Chemical from 1968 to 2002. We worked with thorium as long as I worked in the (redacted)."

The best estimate, according to the testimonies of the Dow (Madison) workers, is that thorium alloys were being produced at the site from about 1956 (Affidavits, 2006) to the late 1990s, and possibly as late as 2002.

Throughout the documents representing testimony presented at workers' meetings and the affidavits associated with the SEC petition, workers indicate that they have knowledge that

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thorium alloys HK31 and HM21 were supplied to other DOE sites, including Mallinckrodt, Rocky Flats, Oak Ridge, and Los Alamos, with a heavy emphasis on shipments to Rocky Flats. Excerpts from affidavits filed with the SEC application and from workers' meetings include the following (Affidavits 2006, Dow 2006a, Dow 2006b, Dow 2006c, Dow 2007) (see Attachment G for additional information):

- “I know that some of this thorium metal was shipped to Rocky Flats, Colorado.”
- “One of my foreman told me that the thorium metal was to be shipped to Rocky Flats.”
- “I was told that some of the thorium that we processed during this period was for the government and was being shipped to Rocky Flats.”
- “I saw sales orders showing that the metal was being shipped to Rocky Flats.”
- “This metal (thorium) was then packed to ship to Rocky Flats, Colorado. The scrap metal that the company there did not use was shipped back to our company to be melted down, and then the above procedure was started all over again.”
- “We rolled and shipped thorium to Rocky Flats, Colorado, and other plants in the United States. We received thorium scrap material back from Rocky Flats, Colorado, and other plants.”
- “And I also sent materials to Rocky Flats, Colorado.”
- “We'd also send it (thorium alloy) to Los Alamos.”

It should be noted that when the workers mention thorium metal, most often they mean thorium alloys HK31 and HM21. In an earlier workers' meeting, there was some indication that thorium metal had been shipped to Rocky Flats. However, in the June 2007 meeting, there seemed to be a consensus that any thorium sent to Rocky Flats was in the form of the commercial alloys HK31 and HM21. There is no firm evidence that thorium materials other than these alloys went to any DOE/AWE site, including Rocky Flats.

Earlier in this report there is a discussion of the quantity of Mg-Th alloys received at Rocky Flats from Dow (Madison) (Section 2.3). Appendix G contains statements from workers at Rocky Flats relative to the use of Mg-Th alloys. While some workers at Dow (Madison) recollect that a vast amount of thorium-bearing alloys were provided to Rocky Flats, Rocky Flats workers who were in a position to know do not confirm such receipts and usage at Rocky Flats.

### **4.3 Thorium or Other Radioactive Materials Processed at Dow Madison**

In addition to the uranium extrusions and rod straightening for Mallinckrodt, there are incidents as reported by the Dow (Madison) workers (Affidavits 2006, Dow 2006a, Dow 2006b, Dow 2006c, Dow 2007) of special processing of radioactive materials at the Madison site. There are

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several possible incidents of special (possibly radioactive?) materials receipt from Rocky Flats as reported by Dow (Madison) workers (Affidavits 2006, Dow 2006a, Dow 2007).

The first incident is reported by a worker as taking place in 1955, when material was received accompanied by armed guards from Rocky Flats (Affidavits 2006, Dow 2006a, Dow 2007). The material was received to be processed in the rolling mill. The material was first believed to be plutonium and later platinum by the worker (Dow 2007). It is unlikely, because of security, criticality, and health and safety concerns, that plutonium would have been involved in such a shaping process. It is not known if Rocky Flats had a need for the rolling of materials such as platinum. The worker who related the incident indicated that a Geiger counter was used to facilitate cleanup following the rolling process. This would lead one to believe that the material was radioactive, and it is unlikely that Rocky Flats had radioactive platinum.

A second event for special materials processing at Dow (Madison) reported by a worker is believed to have taken place in the 1955 time frame (Dow 2007). As in the first case, the material was accompanied by armed guards from Rocky Flats and was to be cut in a shear. The worker could not identify the material, except to recall it was too hard to cut in the shear. The worker remembered being told by the guard that the material was radioactive. The residue from the attempted shear was collected and returned with the material.

At the August 22, 2006 meeting (Dow 2006c), a third worker stated that, "At some time, they ran plutonium through the plant, too, through the homogenating ovens." The worker could not identify the time this occurred, and offered no further explanation of the origin or fate of this material.

It is difficult when reviewing the entire set of records of workers' meetings and the affidavits provided by them to discern the exact number of times the workers believe that special materials were run in the Dow (Madison) plant. It is not always clear if two workers are referring to one incident or more when they are relating the particulars of an incident. Therefore, there could be more than three incidents reported by Dow workers, but the above three seem to be the ones most consistently referred to by workers.

There are several places in the workers' meetings and the SEC affidavits where workers related the processing of special materials in support of aerospace and defense contractors. These are not considered relevant to the SEC petition process.

#### **4.4 Thorium Exposure Conditions beyond 1960**

In the documents provided to NIOSH on May 1, 2007, there were additional air-sampling data. These have been presented above in Section 2.2 and in Attachment F. No external monitoring data or bioassay data have been found for Dow (Madison) workers, and the workers have indicated that none were performed. Workers have stated that some film badge monitoring was done during the Spectrulite ownership, however, no attempt has been made to obtain these results. Just as is the case for the time period of 1957 through 1960, it would likely not be possible to reconstruct the doses resulting from radiothorium for Dow (Madison) employees beyond the 1960 time frame, for the same reasons.

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SC&A can re-examine the Dow (Madison) worker exposure to thorium beyond the 1960 time frame should more information become available from additional sources. With no additional materials to illuminate the activities and exposure conditions involving thorium after 1960, there will continue to be a large gap in our knowledge.

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## ATTACHMENT A: MAGNESIUM-THORIUM ALLOY DESIGNATIONS

(<http://www.ornl.gov/ptp/collection/consumer%20products/magthor.htm>)

### Magnesium-Thorium Alloy (ca. 1980s)

Mag-Thor is a common name for magnesium alloys containing thorium. There are three typical compositions for such alloys:

magnesium-thorium-zirconium

magnesium-thorium-zinc-zirconium

magnesium-silver-thorium-rare earth metal-zirconium

Notable properties of magnesium-thorium alloys include high strength, creep resistance at high temperatures, and light weight.

For example, the alloy designated HK31A, typically formed into sheets or plates, contains 3% thorium and 0.7% zirconium. The "H" in the designation refers to the presence of thorium and the "K" refers to the presence of zirconium. It is described as having good castability, being pressure tight, and possessing creep resistance up to 350 degrees centigrade (a reasonably high value).



Mag-thor has had a variety of applications (e.g., in missiles, spacecraft, tanks), but its primary use is in the manufacture of aircraft parts, especially engines. According to NUREG-1717, the average concentration of thorium in these alloys was about 1.7%.

There is a general trend away from the use of this material. Its radioactive content can be high enough that special procedures are required when handling it and disposal can be expensive. From 1973 to 1983, approximately 4,000 to 5,000 kg of thorium oxide was used per year in alloys for the aerospace industry. In 1991, only 500 kg per year was being used in all metallurgical applications, not just the aerospace industry, and by 1993 that number had dropped to 100 kg per year. At the time that NUREG-1717 was written (2001), there were only two U.S. manufacturers of magnesium thorium alloys (Wellman Dynamics Corp. and Hitchcock Industries) and they had indicated that they might be ceasing production.

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## Dose Estimates

### Routine Use

NUREG-1717 estimated a potential exposure of 50 mrem per year to an aircraft engine maintenance worker. The assumption was that the thorium content of the parts was 1.7%, that the thorium was ten years old, that there was a single large (75 kg) casting and ten smaller castings (1.6 kg each), that the worker was an average of 0.5 meters away from these parts, and that this exposure was for 40 hours per year with an additional exposure of 1000 hours per year to the ten small castings only.

### Distribution and Transport

NUREG-1717 calculated a potential dose of 4 mrem per year to the driver of a truck transporting parts made of a magnesium-thorium alloy. The exposure rate at the drivers location was estimated to be 0.04 mrem per hour.

The assumptions behind the calculations were that the parts contained 1.7% thorium, that each shipment consisted of 100 cartons with two 1.6 kg castings each, that the first row of cartons was 2 meters away from the driver, and that the exposure was for 100 hours per year.

## Pertinent Regulations

10 CFR 40.13 Unimportant quantities of source material (2003)

(c) Any person is exempt from the regulation in this part and from the requirements for a license set forth in section 62 of the Act to the extent that such person receives, possesses, uses, or transfers: . . .

(4) Any finished product or part fabricated of, or containing tungsten or magnesium-thorium alloys, provided that the thorium content of the alloy does not exceed 4 percent by weight and that the exemption contained in this subparagraph shall not be deemed to authorize the chemical, physical or metallurgical treatment or processing of any such product or part;

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**ATTACHMENT B: EXCERPT FROM FUSRAP CLOSE-OUT REPORT  
FOR DOW MADISON SITE, SEPTEMBER 2001**  
(SRDB Ref ID 3084.pdf)

**SITE DESCRIPTION AND HISTORY**

The Madison Site consists of a large, multi-sectional complex of 10 interconnecting buildings with a total under-roof area of about 130,000 square meters (m<sup>2</sup>) [1.4 million square feet (ft<sup>2</sup>)]. Building 6 is about 83-meters (m) [270-feet (ft)] wide and 303-m (1,000-ft) long. The main bay ceiling is approximately 14-m (46-ft) high, 18-m (60-ft) at the highest point along the building centerline. The building structure consists of steel columns on approximately 7.6-m (25-ft) centers, connected by trusses and multiple smaller vertical and horizontal cross members. Walls are concrete block with some brick veneer. Floors are concrete; with rough and pitted surfaces. Much of the floor in the vicinity of the extrusion press is covered with a thin layer of oily dirt and fine metal debris.

The Madison Site located in Madison, Illinois, (see Figures 1 and 2) was used to perform extrusions of uranium metal and straightening of extruded uranium rods for the AEC, the predecessor agency of the Department of Energy (DOE), during the late 1950s and early 1960s. The Dow Metal Products Division of Dow Chemical Company (Dow) conducted this work under subcontract to the Uranium Division of the Mallinckrodt Chemical Works (Mallinckrodt). The work was conducted in Building 6, a large multi-story metal building with a concrete floor. The adjoining Building 4 was used for material transfers. The AEC-funded research was conducted at the plant to determine what factors in the extrusion of uranium metal affected the selection of tools and auxiliary supplies to be used at a planned extrusion press to be located at another AEC production facility. The work included researching properties of various die metals, the contour of the die cavity, the nature of the lubricant to apply to the uranium metal, the composition of the "follower block" (the material placed between the uranium metal and the ram press), and the speed at which the metal could be extruded. At least two rod-straightening campaigns occurred at the Madison Site. Records suggest a small quantity of uranium was involved in these operations. Mallinckrodt retained accountability for the uranium throughout the operations and was responsible for removing unused uranium and for cleanup of facilities following operations. The AEC-funded operations resulted in residual radiological contamination in dust on overhead steel beams in the plant.

In the Designation Summary for the Former Dow Chemical Company Site in Madison, Illinois (ORNL, 1990), the Department of Energy indicated that Dow also supplied materials (chemicals, induction equipment, and magnesium metal products) and services under purchase orders issued by Mallinckrodt. In March 1960, the Uranium Division of Mallinckrodt Chemical Works issued a purchase order for Dow to straighten Mallinckrodt-supplied uranium rods. Two rod-straightening campaigns were identified in the purchase order. One was to be completed on December 21, 1959, the second on January 25, 1960. Each campaign also included a cost for the cleanup of the area after each campaign. The actual periods of performance for this work and the actual quantity of uranium that was processed are unknown. However, the total value of the purchase order and the unit cost identified with the "lot size" indicates that the quantity of metal involved was most likely small. DOE indicated that no other operation or period of involvement

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with the processing or handling of FUSRAP related material at the Madison Site has been discovered.

Dow leased the Madison facility to Phelps Dodge Aluminum Corporation in 1969. Consolidated Aluminum Corporation assumed the lease in 1973 and exercised an option to buy the plant in 1973. Consolidated Aluminum Corporation processed magnesium thorium alloys at the Madison Site. Consolidated Aluminum Corporation sold the Madison plant to Barnes Acquisition, Inc. [which appears to have been a subsidiary of the Spectrulite Consortium, Inc. (Spectrulite)] in September 1986. Although Spectrulite (the current operator of the facility) has also processed magnesium-thorium alloys at the site, these operations are beyond the scope of FUSRAP remedial actions and this document.

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**ATTACHMENT C: SECTIONS 14, 15, AND 16 EXCERPTED FROM  
USAEC INSPECTION REPORT, AUGUST 4, 1960**  
(compliance report\_0001.PDF)

*Note: The original document was not always completely legible, so the transcription presented here may have some minor errors.*

14. Program and Scope of Work

The Dow Chemical Company is licensed to receive possession of and title to thorium metal and/or thorium compounds, both domestically and through import from Canada, for the use in the preparation of magnesium alloys at Madison, Illinois.

Dow Chemical Company is further licensed to transfer and deliver possession of and title to refined source material to any person licensed by the Atomic Energy Commission, within "the limits of his license."

The largest supply of thorium comes from Canada in the form of pellets. As of July 1, 1960, a total of eighty tons of thorium had been used at the magnesium foundry at Madison, Illinois. A breakdown on the amount and type of thorium used at the foundry is as follows:

- (1) 64 tons in the form of pellets were obtained from Canada
- (2) 23 tons in the form of scrap were obtained from Consolidated Edison Company
- (3) 3 tons of master alloy ranging from 25 to 40 percent thorium in the form of notched ingots were obtained from Canada

Thorium is stored inside a fenced off area in an isolated section of the Melt Room Building. The fenced off storage area and the Melt Room are posted with signs comprising the conventional radiation symbol in color and the words "Caution – Radioactive Material, Unauthorized Persons Stay Away."

In the melting area, 3% thorium is added to the magnesium melt. The magnesium-thorium alloy is poured in the adjacent Pour-Off Area. The slag and flux and are [sludged] off and are remelted. The metal is poured. Then the remaining sludge is poured off. It contains up to 50% thorium. The sludge is broken up, and the metal is recovered and remelted.

The sludge containing from 4% to 8% thorium is taken to the Waste Area, where it is kept segregated for possible future recovery of the contained thorium. Fines resulting from grinding the alloy are [burned] at the Waste Area. The Waste Area is fenced off from the rest of the plant and is under constant guard.

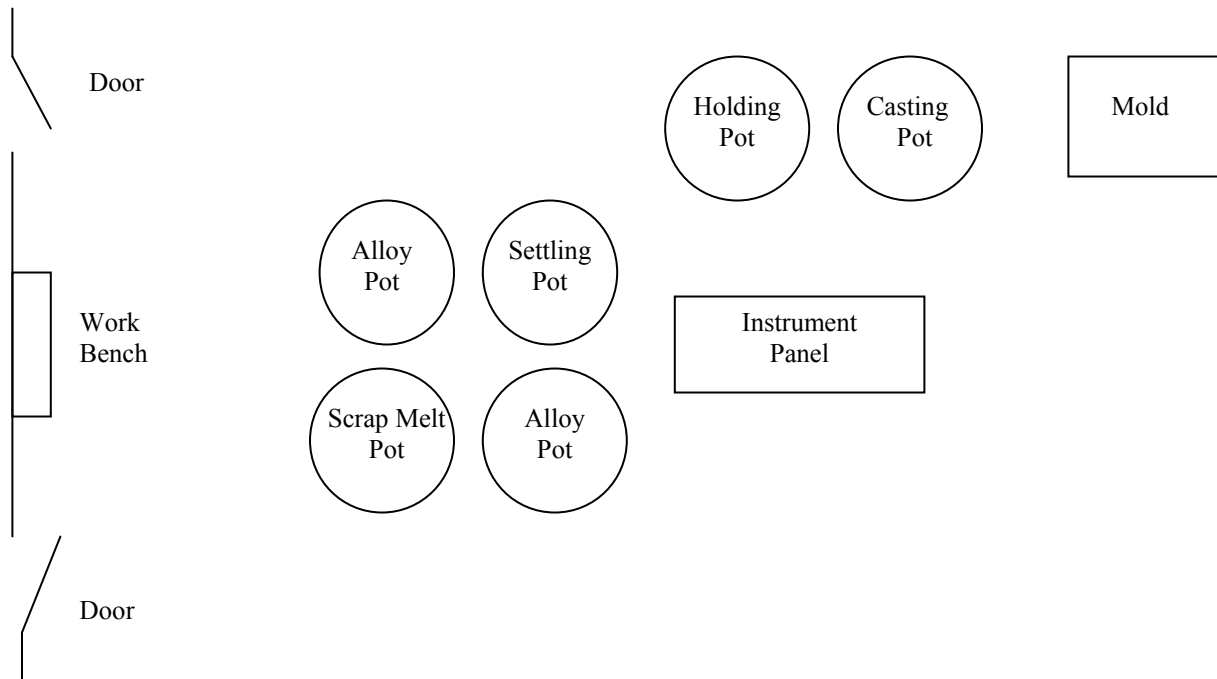
In the foundry, scrap from shipping, sawing, and milling magnesium-thorium alloy is kept segregated all through the plant for remelting.

At the time of the inspection there was approximately 7200 pounds of contained thorium and notched ingots of master alloy scrap stored inside the fenced off posted area inside the Melt Room Building.

#### 15. Facilities and Equipment

The production area where two and three percent alloys are made in 20,000 or 30,000 pound lots, consist of a workbench, an instrument panel, a mold, and six large pots, all loaded in the Melt Room Building. (See Exhibit A.)

**Exhibit A  
Production Area**



Located sixty-five feet above the production area is a twenty-foot exhaust fan. Other 20-foot exhaust fans are located on each side of the production area.

Adjacent to the Melt Building are other buildings and areas where a variety of mechanical operations are carried out. These operations are surface and open wheel grinding, rotary filing, buffing, sawing, drumming of powder and mill slab heat treatment and rolling. In each building and area where fumes are a problem, there are exhaust fans available to adequately remove the fumes.

#### 16. Radiation Measurements

Radiation surveys along with a material inventory report are made each month and sent to the home office in Midland, Michigan. Radiation Evaluations are made of the waste storage area

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where sludge containing up to 8% thorium is stored and the fenced off posted cage in the Melt Room Building where the virgin thorium is stored. Thorium is stored in the cage in such a manner as to prevent any large dose rate from emitting from the cage. The latest survey results which were made on August 4, 1960, are as follows:

<b>Waste Sludge Storage Area</b>			
	<b>Location</b>	<b>Open Window (beta)</b>	<b>Closed Window (gamma)</b>
(1)	On secondary road 50 feet from sludge	.08 (mrem/hr)	.06 (mr/hr)
(2)	At Radiation Sign about 35 feet from sludge	.13 (mrem/hr)	.10 (mr/hr)
(3)	At fence – 80 feet north of sludge pile	.06 (mrem/hr)	.04 (mr/hr)
(4)	Surface reading of sludge piles	10.0 (mrem/hr)	8.0 (mr/hr)

<b>Virgin Thorium Storage Area</b>			
	<b>Location</b>	<b>Open Window (beta)</b>	<b>Closed Window (gamma)</b>
(1)	At door of cage	1.0 (mrem/hr)	.8 (mr/hr)
(2)	12 inches above 7000 pounds of master alloy scrap	7.0 (mrem/hr)	6.0 (mr/hr)
(3)	Center of cage	2.5 (mrem/hr)	3.5 (mr/hr)

(Dose rate measurements were made with a Precision Radiation Instruments, Geiger Counter Model 121 with Model-B Beta-gamma probe.)

Mr. L.G. Silverstein, the Radiological Safety Officer located at Dow Chemical Company's Midland, Michigan offices, makes periodic visits to the Madison, Illinois plant to made dose rate evaluations of the two storage areas (waste thorium and virgin thorium areas) and to make toxicity and contamination evaluations of the breathing zone air inside the production buildings. He also makes recommendations to the Safety Department and to the Works Manager on radiological problems whenever they arise.

Air samples taken recently by Mr. Silverstein were collected with an electrostatic precipitator and counted for alpha and beta activity in a NMC Model PC-1 proportional counter. The counter efficiency for alpha and beta were determined by counting a calibrated alpha and beta sources.

Mr. Saunders stated that with the exception of welding, the common operations of fabricating magnesium do not contaminate workroom air sufficiently to be of concern. The safe practices normally followed to collect grinding dust, the standard storage methods employed for raw materials and finished products and the proper ventilation used in pickling operations all adequately control air contamination.

Mr. Eugene Burnett, the plant's safety advisor, stated that the Industrial Hygiene Standard for thorium is 76 micrograms per cubic meter of air. This standard is used when determining air

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concentrations. (See Thorium Content, in Micrograms/Cubic Meter in Breathing Zone Air during several Mechanical Operations as Exhibit B.<sup>20</sup>)

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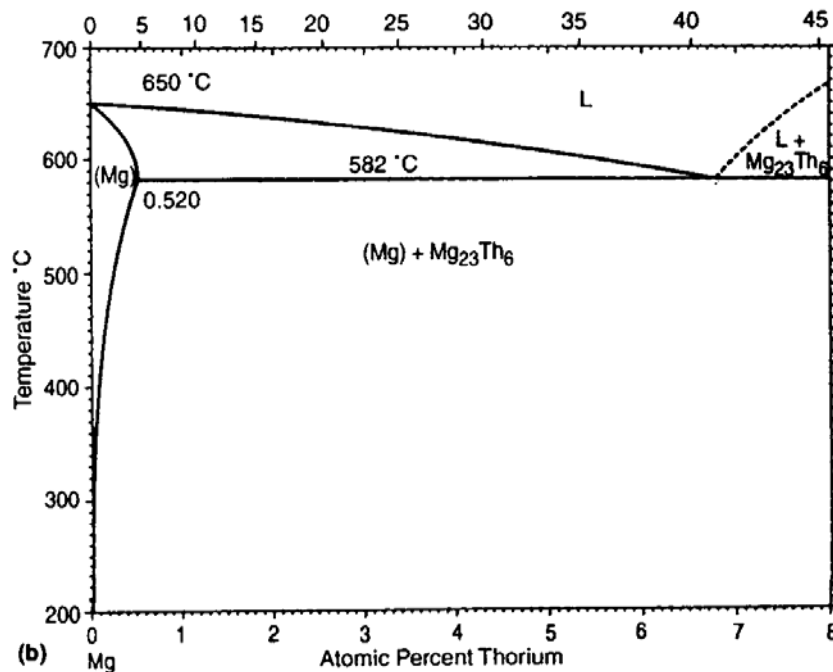
<sup>20</sup> Exhibit B is not reproduced here but it contains the same information as included in Figure 3 of this report.

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## ATTACHMENT D: MG-TH SYSTEM (ASM 1999)

The binary phase diagram for the portion of the Mg-Th system up to about 45 wt% Th is shown in Figure D-1. Over this composition range, a simple eutectic system exists with a eutectic temperature of 582°C and a eutectic composition of about 41 wt% Th. The eutectic consists of Mg with Th in solid solution and the intermetallic phase Mg<sub>23</sub>Th<sub>6</sub>. At the eutectic temperature, the maximum amount of Th in solid solution in Mg is 4.75 wt% (0.52 at %). Under equilibrium conditions, as the temperature decreases further, Mg<sub>23</sub>Th<sub>6</sub> will precipitate from the solid solution.



**Figure D-1. Magnesium-rich portion of Mg-Th binary phase diagram**  
(This is copyright-protected material)

The diagram in Figure D-1 describes the situation when a metal solidifies under equilibrium conditions. In actual practice the situation may be different. As described in ASM 1999 (p. 28):

***Thorium.** At the eutectic temperature of 589°C (1092°F), 4.5% Th is soluble in magnesium; however, because of alloy segregation, magnesium alloys containing as little as 2% Th often contain a divorced eutectic and show massive Mg<sub>4</sub>Th compound at the grain boundaries. At temperatures below the eutectic, this compound is also precipitated from solid solution. In castings, the precipitate forms within grains and is seldom visible. In worked structures, the precipitate is often clearly visible at grain boundaries.*

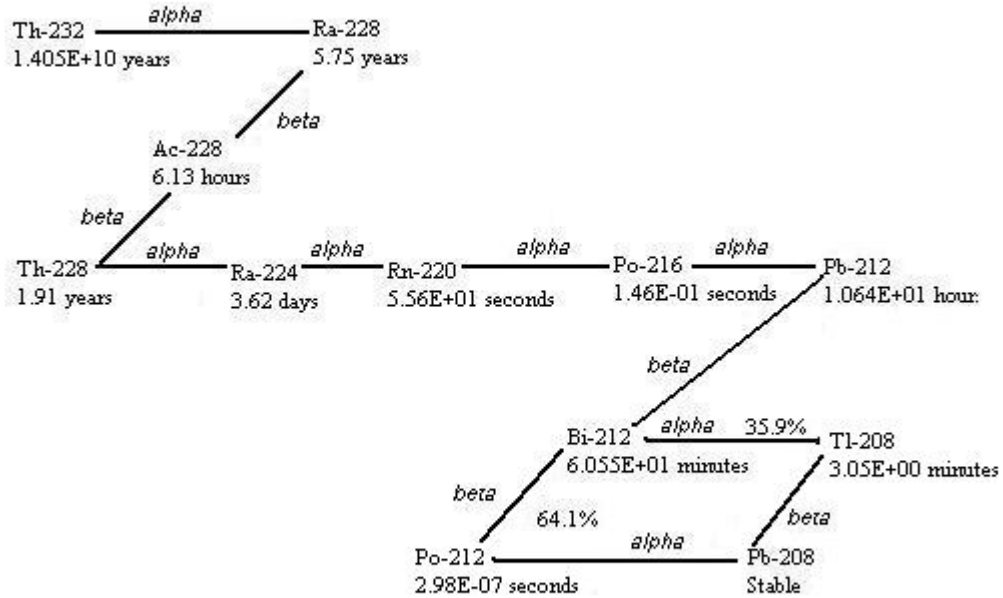
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*The addition of thorium to magnesium-zinc alloys changes the degenerate eutectic, which contains magnesium-zinc compound, to a lamellar eutectic, which contains a magnesium-thorium-zinc compound.*

It is thus likely that any metallic particles generated during the processing of commercial Mg-Th alloys will be either Mg with a Mg<sub>23</sub>Th<sub>6</sub> (or Mg<sub>4</sub>Th) precipitate within the particles or, for very fine particles, possibly only Mg<sub>23</sub>Th<sub>6</sub>.

ASM 1999. *Magnesium and Magnesium Alloys – ASM Specialty Handbook*. ASM International, Metals Park, Ohio.

## ATTACHMENT E: TH-232 DECAY CHAIN



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## ATTACHMENT F: SUMMARY OF MONITORING RESULTS AT DOW MADISON

This attachment summarizes the radiation monitoring results reported by Dow in various documents. Many of these results are also discussed in the main body of the report but are also included here for completeness. Dow frequently refers to radiation levels relative to “tolerance” in citing measurement results. “Tolerance” refers to an exposure level acceptable to Dow rather than a regulatory mandate. Tolerances used by Dow in 1957 (Silverstein 1957) were:

- Thorium: 100 micrograms of Th per cubic meter of air<sup>21</sup> or  $1.1 \times 10^{-11}$  microcuries per milliliter
- Mesothorium (Ra-228):  $10^{-11}$  microcuries per milliliter ( $1.2 \times 10^{-11}$   $\mu\text{Ci/mL}$  in Silverstein 1956b)
- Short-lived alpha emitters:  $10^{-7}$  microcuries per milliliter ( $10^{-8}$   $\mu\text{Ci/mL}$  in Silverstein 1956b)

Monitoring results are presented in the following sections.

### F.1

Date: July 22-23, 1956

Activity: Melting of 35,000 lb of Alloy HM21 in Pot Room (see Section 2.2 above for additional details)

Reference: Silverstein 1956, Silverstein 1957

Results: Air Monitoring Data are included in Table F-1

**Table F-1. Air Sampling Measurements Taken During Melting and Casting of Alloy HM21 at Dow Madison (July 22-23, 1956)**

Sample Number	Time	Description	Short-lived Radioactivity (Thoron & daughters) (Ci/L)	Radium-228 (Ci/L)	Thorium (mg/m <sup>3</sup> )
M-1	1701–1715 7-22-56 1.2 m <sup>3</sup>	Breathing zone (BZ) of man cutting open 10 drums of HM21 heels with acetylene torch. HM21 was cast into drums on 2-11-56.	$6 \times 10^{-13}$	$12 \times 10^{-14}$ (10 × tolerance)	0
M-2	1800–1820 1.7 m <sup>3</sup>	Between pots 36 & 37, 8 ft. away from either pot. HM21 scrap (last melted on 2-11-56) was being melted, some burning of metal was observed. Workers occupied this area once every 5 or 10 min. for less than one minute each time.	$4 \times 10^{-13}$	$3.5 \times 10^{-14}$ (3 × tolerance)	0

<sup>21</sup> In a 1959 document the maximum allowable concentration was quoted as 76  $\mu\text{g/m}^3$  (Levy 1959).

**Table F-1. Air Sampling Measurements Taken During Melting and Casting of Alloy HM21 at Dow Madison (July 22-23, 1956)**

Sample Number	Time	Description	Short-lived Radioactivity (Thoron & daughters) (Ci/L)	Radium-228 (Ci/L)	Thorium (mg/m <sup>3</sup> )
M-3	1842–1904 1.87 m <sup>3</sup>	At workbench across aisle from pots 36 & 37. Workers spent most of their time here or outside door during melting down of HM21 scrap.	$5 \times 10^{-13}$	0	0
M-4	2027–2140 6.2 m <sup>3</sup>	50 ft. south of operation near another group of settings on which men were working.	$5 \times 10^{-13}$	0	0.004
M-5	2214–2222 0.68 m <sup>3</sup>	BZ of worker alloying 70# of thorium pellets into 5000# of HM21 + cell Mg in pot 32. Pellets burned before being immersed.	$6 \times 10^{-13}$	$0.27 \times 10^{-14}$	0
M-6	0036–0056 7-23-56 1.7 m <sup>3</sup>	In front of instrument panel. Pot 32 was recharged with HM21 scrap, melt was pumped from pot 37 to pot 32, and from 33 to pot 31, during sampling period. Some burning occurred during pumping and also in pot 32 when scrap was added.	$8.7 \times 10^{-13}$	$0.27 \times 10^{-14}$	0.02
M-7	0220–0250 0.425 m <sup>3</sup>	Near washroom, about 100 ft. north of the group of settings in use.	$7 \times 10^{-13}$	$0.3 \times 10^{-14}$	0
M-8	0315–0320 0.935 m <sup>3</sup>	BZ of worker alloying 50# of thorium pellets in pot 32. No burning of the pellets was observed.	$6.2 \times 10^{-13}$	0	0
M-9	0624–0635 1.87 m <sup>3</sup>	BZ of man alloying 100# of hardener into pots 31 & 32, 50# had been alloyed into pot 30 just before sampling began. Hardener was last melted on 2-7-56.	$6.2 \times 10^{-13}$	$0.44 \times 10^{-14}$	0.01
M-10	0907–0929 1.78 m <sup>3</sup>	BZ of worker attending mold during casting of HM21.	$8 \times 10^{-13}$	$0.17 \times 10^{-14}$	0.011
M-11	1007–1028 1.78 m <sup>3</sup>	By billet caster 50 ft. south of operation. 70# of pellets were alloyed in pot 32, pot 32 was pumped into pot 31, pot 36 was mechanically sludged and pot 33 was hand-sludged during sampling period.	$6.0 \times 10^{-13}$	0	0
M-12	1032–1053 1.78 m <sup>3</sup>	By workbench across aisle from pot 37. Pot 33 was pumped over to pot 31, sludge was pumped from pot 32 into cake molds.	$7.5 \times 10^{-13}$	0	0

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**Table F-1. Air Sampling Measurements Taken During Melting and Casting of Alloy HM21 at Dow Madison (July 22-23, 1956)**

Sample Number	Time	Description	Short-lived Radioactivity (Thoron & daughters) (Ci/L)	Radium-228 (Ci/L)	Thorium (mg/m <sup>3</sup> )
M-13	1107–1140 2.7 m <sup>3</sup>	By instrument panel. Sludge and metal were pumped out of pots 30 and 31 into cake molds.	$3.8 \times 10^{-13}$	0	0.006
M-14	1243–1310 2.3 m <sup>3</sup>	By instrument panel. Sludge and metal were pumped out of pots 30 and 31 into cake molds.	$3.8 \times 10^{-13}$	0	0.006

## F.2

Date: Unknown but between July 23 1956 and June 19, 1957

Activity: Production of Alloy HK31 and HM21

Reference: Silverstein 1957

Results: No-long-lived alpha or beta on any sample. Short-lived alpha in the range of  $10^{-10}$   $\mu\text{Ci/mL}$ .<sup>22</sup>

## F.3

Date: May 21, 1959

Activity: Annual Industrial Hygiene Summary

Reference: Levy 1959

Results: A cumulative summary of all Madison survey to that date is included in Table F-2.

Correspondence attached to Levy 1959 (Levy letter to H.R. Hoyle, July 27, 1959) suggested that it would be timely to evaluate thorium and daughter products evolved during sludge centrifuging since the first unit had been used in production for some time and second unit was being considered. According to worker interviews, the centrifuge(s) was only used for about two years. Because of an industrial accident, their use was discontinued (REFERENCE).

<sup>22</sup> Silverstein (1957) is not specific in his definitions of short and long-lived alpha and long-lived beta. He states on p. 6 that short-lived alpha samples in Table F-1 displayed the characteristic half-life of radon daughter products. This suggests that Ra-224 was either not considered to be one of the short-lived alpha products or it was not released in measurable quantities. It can be inferred from Silverstein's paper that Th-232 and Th-228 are the principle sources of long-lived alpha. It can also be inferred that Ra-228 is the primary source of long-lived beta. See Attachment E for details on the Th-232 decay chain.

**Table F-2. Cumulative Summary: Madison Division Industrial Hygiene Surveys (May 21, 1959)**

Substance	Maximum Allowable Concentration	Concentration Found	Location	Date	Comments
Ra-228	$1.2 \times 10^{-14}$ Ci/l	0 - $12 \times 10^{-14}$ Ci/L	Pot room, alloying and casting HM21	7/56	Present operation no problem
Thorium	76 $\mu\text{g}/\text{m}^3$ *	0 - 3	Pot Room	9/55	
		0.2	By #1 Slab Oven	10/55	Sl. Dust on HK31 Slab
		1	#7 Mill	10/55	Rolling HK31
		<.35	Sheet wire brush machine	2/56	HK31
		<10	Slab Scalper	7/56	HM21
		Nil - 20	R.M. hand sanding HK31	3/57	
		18 - 36	R.M. Salvage	12/57	Vibrator sanders on HK31
		52 - 94	Tech. Dept.	3/58	Rotary sanding HK31
		<9 - <15	#2 Mill	3/59	Rolling steam annealed HK31
Thoron, etc. (short lived)	$10^{-11}$ Ci/l	$3.8 - 8.7 \times 10^{-14}$ Ci/l	Pot room, alloying and casting HM21	7.6	Max. is only 9% of tolerance

\* 1957 Dow documents used 100  $\mu\text{g}/\text{m}^3$  as the Th tolerance limit.

#### F.4

Date: December 15-16, 1959

Activity: Melting of Alloy HK31

Reference: Dow 1959

Results: Air Monitoring Data (from Millipore filter samples) included in Table F-3

**Table F-3. Air Sampling Measurements Taken During Melting and Casting of Alloy HK31 at Dow Madison (December 15-16, 1959)a**

Sample Number	Time	Description	Short-lived Radioactivity (Thoron & daughters) ( $\mu\text{Ci}/\text{mL}$ )	Short-lived Radioactivity (Radon & daughters) ( $\mu\text{Ci}/\text{mL}$ )	Thorium ( $\mu\text{g}/\text{m}^3$ )
1	1354-1458 12-15-59 1.37 $\text{m}^3$	Work area in pot room, 6 to 8 ft from pots 37 & 38. Alloying HK31	$1.3 \times 10^{-11}$	$1 \times 10^{-10}$	<26
2	1351-1500 1.54 $\text{m}^3$	Work area next to control panel for sludge recovery unit which was not operating.	$3.1 \times 10^{-11}$	$0.73 \times 10^{-10}$	<23

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**Table F-3. Air Sampling Measurements Taken During Melting and Casting of Alloy HK31 at Dow Madison (December 15-16, 1959)a**

Sample Number	Time	Description	Short-lived Radioactivity (Thoron & daughters) ( $\mu\text{Ci/mL}$ )	Short-lived Radioactivity (Radon & daughters) ( $\mu\text{Ci/mL}$ )	Thorium ( $\mu\text{g/m}^3$ )
3	1533-1553 0.428 m <sup>3</sup>	Same as Sample 1.	$2 \times 10^{-9}$	$2.3 \times 10^{-10}$	<84
4	1533-1554 0.471 m <sup>3</sup>	Same as Sample 2.	$1 \times 10^{-10}$	$1.2 \times 10^{-10}$	<76
5	0934-1034 12-16-59 1.28 m <sup>3</sup>	Pot room air near pot 37 & 38, about 7 to 8 ft distant. Rotameter dropped to 5 between 10:13 and 10:26.	$1.2 \times 10^{-11}$	$1.7 \times 10^{-10}$	<12
6	0854-0954 1.34 m <sup>3</sup>	Sludge recovery room next to panel on operator's side. Rotameter on bottom at the end of run. .	$3.7 \times 10^{-11}$	$3.5 \times 10^{-10}$	<11
7	1038-1138 1.28 m <sup>3</sup>	Same as #5 above. Ringstand knocked over.	$2 \times 10^{-11}$	$2.2 \times 10^{-10}$	<12
8	0958-1058 1.34 m <sup>3</sup>	Same as #6. Rotameter at 55 at end of sample. Centrifuge started at 10:25	$0.5 \times 10^{-11}$	$3.1 \times 10^{-10}$	<11
9	1141-1241 1.28 m <sup>3</sup>	Sampler next to telephone and instrument panel. Air flow down to zero at 12:40	$3.4 \times 10^{-11}$	$1.1 \times 10^{-10}$	<11
10	1101-1206 1.46 m <sup>3</sup>	Sample on the centrifuge side ?? shield. Centrifuge started at 11:28 with AZ31 sludge.	$3.2 \times 10^{-11}$	$2.6 \times 10^{-10}$	<10
11	1318-1413 1.18 m <sup>3</sup>	Control panel near pots 30 and 31..	$1.6 \times 10^{-11}$	$1.9 \times 10^{-10}$	<13
12	1249-1349 1.34 m <sup>3</sup>	Same as #10. Centrifuge started with HK31 at 1:26	$2 \times 10^{-11}$	$3.6 \times 10^{-10}$	<13
13		.Blank			
14	1355-1455 1.34.3 m <sup>3</sup>	Sample on drum next to operator's desk in sludge recovery. Batch of HK31 started at 2:25..	$2.3 \times 10^{-11}$	$1.1 \times 10^{-10}$	<11

a – Long-lived beta activity above background was not detected in any samples except Sample 8. The value for this sample is illegible in the available copy but appears to be  $2 \times 10^{-10}$   $\mu\text{Ci/ml}$  or less.

## F.5

Date: Unknown but prior to June 19, 1957

Activity: Various mechanical operations

Reference: Silverstein 1957

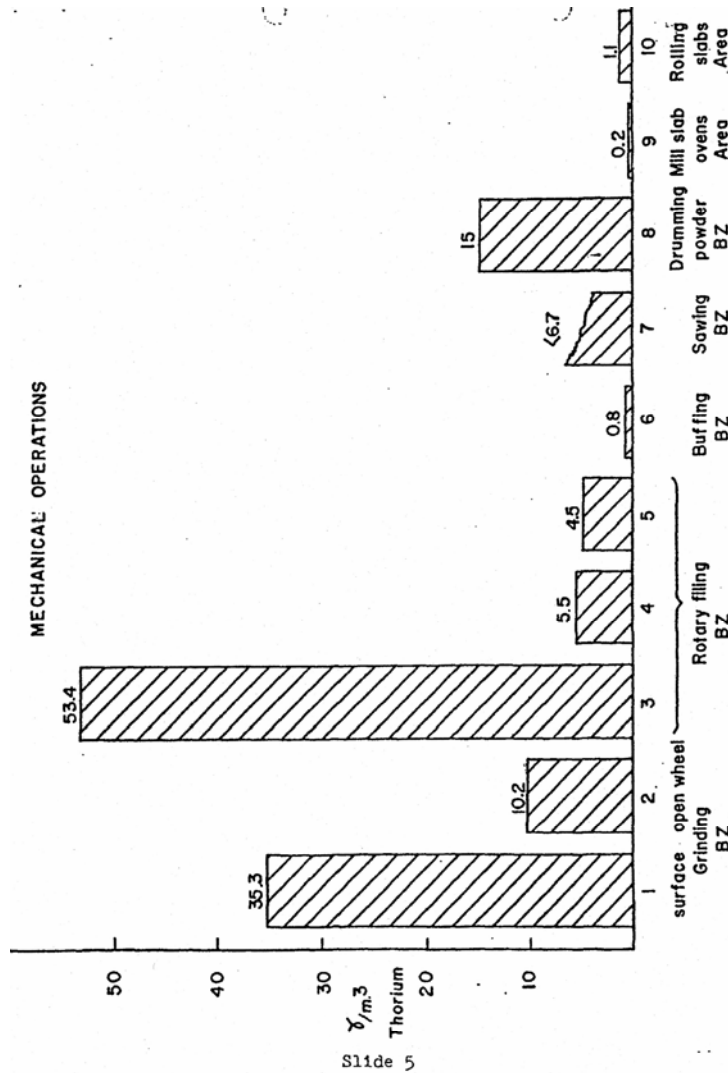
Results: Air sampling results are summarized in Figure F.5-1. (See also Section 2.3 of main report). All samples except "Mill slab ovens" and "Rolling slabs" were breathing zone samples

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taken during the rolling of HK31. The samples were taken with an electrostatic precipitator and spectrographically analyzed for Th. As described by Silverstein (1957):

*All [except as noted above] were breathing zone samples and the operation took place on equipment that is designed for safe operation with magnesium, which means local exhaust and water traps to collect the magnesium fines. The high sample in rotary filing probably resulted from a stray "brick," a large particle which is typical of the operation, but would be breathed by an operator.*



TDCC 000380

**Figure F.5-1. Thorium Dust Generated During Various Mechanical Operations on Th-Mg Alloys**

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## F.6

Date: March 24, 1958

Activity: HK31 sanding

Reference: Mitchell 1958 and Silverstein 1958

Results: Air sampling data were also obtained during sanding of HK31 with a vertical metallurgical sanding disks on 2 in by 3 in samples. These samples were taken in the Technical Department metallurgical area (location unknown). Based on the sample size and location of the work, it is presumed that was small-scale experimental work rather than production work. The samples were collected on Millipore filters using a sampler operating at 0.5 cfm in the breathing zone of the operator. There was no local exhaust on the sanding equipment. Results are summarized in Table F-4.

**Table F-4. Thorium in Dust Samples Taken During Sanding HK31**

Sample No.	Air Volume (ft <sup>3</sup> )	Sanding Procedure	Thorium Concentration (µg/m <sup>3</sup> )
1	2.7	4-grit sequence – 50, 150, 320 and 0	52.3
2	1.5	150 grit only with periodic drag on wheel flange	94
3	1.5	320 grit only with periodic drag on wheel flange	94

## F.7<sup>23</sup>

Date: Unknown but prior to June 26, 1957

Activity: Casting of HK31

Reference: Silverstein 1957b

Results: External exposures based on film badges worn by operators for 13 working days are included in Table F-5

**Table F-5. Film Badge Survey of HK31 Casting Production**

Job	Exposure (mr)	
	min	max
Pour Off	10	75
Knockout	<10	10
Sandblast	<10	10
Bandsaw	10	50
Chipper	<10	10
Machine Trim	10	10
Inspection (rough)	10	10
Pickler	<10	<10
Rotary File	10	50
Buffer	10	10

<sup>23</sup> After a detailed examination of Silverstein 1975b and other references, it is clear that these film badge results are for materials at Dow (Bay City). However, these results should be directly applicable to materials at Dow (Madison) because of their similarity.

**Table F-5. Film Badge Survey of HK31 Casting Production**

Job	Exposure (mr)	
	min	max
Heat Treat	10	10
Wheelabrator	<10	<10
Inspection (final)	10	10
Touchup	<10	<10
X-ray	<10	<10
Fixture	<10	<10
Shipping	50	50
Metal Reclamation	10	10
Control Badge	10	10

**F.8**

Date: August 4, 1960

Activity: Survey of Mg-Th sludge dump

Reference: Saunders 1960

Results: The posted sludge dump (east of a north-south road adjacent to Building 7) was surveyed with a Precision Radiation Instruments Geiger Counter, Model 121 (AEC #S6M93A) with Model B beta-gamma probe. The measurement data summarized in Table F-6 were submitted to the AEC. Based on these measurements, Dow concluded that the results indicated no problem and solicited AEC's opinions relative to the significance of the data.

**Table F-6. Beta-Gamma Activity at Mg-Th Sludge Dump**

Sample	Location	Exposure (mr/hr)	
		Cover Closed (excludes beta)	Cover Open
1	At hard road-secondary road intersection	<0.05	---
2	On secondary road- nearest to location of sludge cakes	0.06	0.08
3	At fence- south of dump	0.16	0.28
4	Two feet from a pile 25' x 10' x 5'	2	2.5
5	Surface of above pile	7-8	10
6	At fence – south of dump but 30 ft more easterly than Sample 3	0.04	0.06
7	At fence – extreme east of dump	0.04	0.04
8	Surface of another pile	6-7	---
9	At location of "HM" sign	0.10	0.13
10	Another pile	8	9
11	Intersection of secondary road and path-road	0.05	0.06

**F.9**

Date: August 4, 1960

Activity: Survey of virgin Th storage area

Reference: Saunders 1960

Results: External exposure measurements made at the posted cage where virgin Th was stored are summarized in Table F-7. At the time the measurements were made approximately 3,200 lb of contained Th was in storage.

**Table F-7. External Exposure Survey of Thorium Storage Cage**

Sample	Location	Exposure (mr/hr)	
		Cover Closed (excludes beta)	Cover Open
1	At door – near sign	0.8	0.9–1.0
2	12 in above 7,000 lb of notched ingots of master alloy scrap (5 to 19.2% Th)	5-6	6-7
3	Center of cage area	2.5-3.5	---
4	Center of a cluster of eight 50-gallon steel drums each containing about 350 lb of master alloy (27-42% Th)	>20	
5	Immediate external surface of steel drum containing 170 lb of Th pellets	>20	

### F.10

Date: September 17-18, 1956

Activity: Melting and Casting of HM21 and HK31.

Reference: Dow 1956 (TDCC 000014 - 000018)

Results: Results of air sampling measurements made using electrostatic precipitator during the melting and casting of HM21 and HK31 are summarized in Table G-8. Note: The copy from which this table was prepared was difficult to read and some of the interpretations and entries may not be accurate. ??? indicates illegible text.

**Table F-8. Air Sampling Measurements Taken During Melting and Casting of HM21 and HK31 Alloys at Dow Madison (September 17-18, 1956)**

Sample Number	Time	Description	Short-lived Alpha Radioactivity (Ci/m <sup>3</sup> )	Long-Lived Alpha Radioactivity (Ci/m <sup>3</sup> )	Long-Lived Beta Radioactivity (Ci/m <sup>3</sup> )
101	1352-1452 9-17-56 5.1 m <sup>3</sup>	By control panel on west wall. Sludging Pot 38, melting scrap HK31 sheet in Pot 38, alloying hardener in Pot 35.	$1 \times 10^{-10}$	none	none
102	1508-1538 2.55 m <sup>3</sup>	Front of instrument panel. Scrap burned (????) and melted in Pot 32	$1 \times 10^{-10}$	none	none
103	1545-1602 1.275 m <sup>3</sup>	Sample no good			
104	1600-1624 2.04 m <sup>3</sup>	By Pot 33. Mechanical sludging took place.	$2 \times 10^{-10}$	none	none
105	1625-1647 1.87 m <sup>3</sup>	Pot 35 melting scrap. ?????	$2.3 \times 10^{-10}$	none	none
106	1648-1652 0.34 m <sup>3</sup>	Pouring ingots.	$3.2 \times 10^{-10}$	none	none
107	1733-1802 2.96 m <sup>3</sup>	?????	$1.2 \times 10^{-10}$	none	none
108	???? 2.55 m <sup>3</sup>	?????	$1.1 \times 10^{-10}$	none	none
109	1933-1952 1.7 m <sup>3</sup>	10 ft from Pots 35 and 36. Pumping from Pot 39 to Pot 26. Mechanical sludging of Pot 39 took place.	$1.74 \times 10^{-10}$	none	none
110	0855-0928 9-18-56 1.46 m <sup>3</sup>	5 ft from Pot 35. Charged scrap sheet into Pot 38. Alloyed 150 lb of Mg-Th hardener into Pot 35.	$6.4 \times 10^{-10}$	none	none
111	1005-1020 1.275 m <sup>3</sup>	By workbench opposite Pot 35 where ??? scrap was being melted.	$7 \times 10^{-10}$	none	none
112	1015-1037 1.87 m <sup>3</sup>	By workbench opposite Pot 37	$4.7 \times 10^{-10}$	none	none
113	????	????	????	????	????
114	1249-1304 1.275 m <sup>3</sup>	By ??? billet casting panel	$2.2 \times 10^{-10}$	none	none
115	1056-1121 2.13 m <sup>3</sup>	4 ft from Pots 31 and 33. ?????	$2.1 \times 10^{-9}$	none	none

**F11**

Date: March 14, 1957

Activity: Hand Sanding HK31 Alloy (See also Table G-2 above)

Reference: Hoyle 1957

Three samples were collected in the operator's breathing zone during the hand sanding of a sheet of HK31 and analyzed spectrographically for Th. Sample No. 1, based on a 10-minute air sample at 1 ft<sup>3</sup>/min, contained less than 1 µg Th per 0.283 m<sup>3</sup>. Sample No. 2, based on a 5-minute air sample at 1 ft<sup>3</sup>/min, contained less than 1 µg Th per 0.141 m<sup>3</sup>. Based on the measurements, these samples were judged to contain essentially no Th. Sample No. 3, based on a 10-minute air sample at 1 ft<sup>3</sup>/min, contained 6 µg Th per 0.283 m<sup>3</sup> or 21 µg/m<sup>3</sup> of Th. Assuming a tolerance level of 100 µg/m<sup>3</sup> for repeated 8-hour exposures, "It can therefore be said that there is no hazard to the health of workmen who are hand sanding sheets of HK-31 alloy under the conditions represented by these samples."

## F12

Date: January 10, 1956

Activity: External Exposure to Mg-Th Alloys

Reference: Dow 1956b

External exposure measurements made with a Raychronix instrument on Mg-Th alloy sheets and slabs are summarized in Table F-9. The units of measurement are not stated in Dow 1956b but are inferred from other Dow documents to be mR/hr.

**Table F-9. External Exposure Measurements on Mg-Th Alloys**

Material	Source Thickness (in)	Distance from Source (in)	Beta/Gamma Measurement (mR)	Gamma Measurement (mR)
12-in squares HK31 sheet	0.25	0	2.8	0.5
		6	1.2	0.6
	5.7	0	2.8	1.6
	9.3	0	2.9	1.7
	12	0	2.5	1.6
140 in x 53 in sheets of HK31	14	0	4.7	2.4
		12	1.5	1.5
		48		1.5
40 in by 50 in sheets before crating	7	0	4.2	2.5
		12	2.0	1.8
		24		1.5
		36		0.5
		48		0.5
		60		0.1
40 in by 50 in sheets after crating with cardboard and tarred paper	7	0	2.5	2.5
		12		1.5
		24		1.2 - 1.3
		36		1.0
		48		0.5
		60		0.0

**Table F-9. External Exposure Measurements on Mg-Th Alloys**

Material	Source Thickness (in)	Distance from Source (in)	Beta/Gamma Measurement (mR)	Gamma Measurement (mR)
Slabs of HK31 10 in by 26 in by 76 in	108	0	4.3, 3.5, 4.0. 3.2, 5.1, 4.0. 3.5	2.2
		12	1.7	1.7
		24	1.2	1.2
		36	1.0	1.1
		48	0.5	0.7
		60	0.4	0.6
		72	0.2	0.2
		84	0.0	0.0

### Attachment F References

Dow 1956a. Industrial Hygiene Sampling Report. September 17-18, 1956. TDCC 000014-000018.

Dow 1956b. *Industrial Hygiene Sample Record – Madison*. January 10, 1956. TDCC 000001 – TDC 000004.

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Levy, D.J. 1959. Memo to J.R. Burns: *Annual Industrial Hygiene Summary*. The Dow Chemical Company. May 21, 1959.

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Silverstein, Lawrence G. 1957. *Industrial Hygiene Experience with Magnesium-Thorium Alloys*. Dow Chemical Company. Presented at the Health Physics Society Meeting, Pittsburgh, Pennsylvania, June 19, 1957. USAEC Docket 40-17.

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## ATTACHMENT G: USE OF MG-TH ALLOYS AT ROCKY FLATS

Source: *NIOSH Evaluation of SC&A's draft report on Other Radionuclides (thorium)*  
February 28, 2007.

*Telephone interviews were conducted January 9<sup>th</sup> and 10<sup>th</sup> with four former Rocky Flats personnel that started their careers at the facility in the 1950s, and each worked there for some 25 to over 35 years. The four workers interviewed were in various management positions and would have knowledge of thorium uses and sources at Rocky Flats. The comments contained in this summary are specifically related to the use of magnesium alloy at Rocky Flats, and if these employees would have knowledge of any significant quantities (more than a few kilograms) of magnesium alloy present at the facility.*

One worker stated that the conveyor system in Building 776 box line used magnesium for reduction in weight and strength stability, and that no magnesium alloy was used in any processes for development or fabrication of weapons components. The worker had no recall of significant quantities or other uses of magnesium alloy at Rocky Flats. That worker also stated that if there were significant quantities of magnesium alloy present, [the worker] would have known about it. The worker also stated that pure magnesium salts were used in some of the chemical processes, primarily for reduction of metal.

Another worker clearly remembered that only the pennants in the conveyor system were made of magnesium alloy. The conveyor was constructed of stainless steel. The worker stated that the conveyor system and pennants were fabricated and installed by an outside vendor. This worker recalled being one of the first persons back into Building 776 after the May 11, 1969, fire, and clearly remembered that only a few pennants that were no longer present. The worker estimates that each pennant weighed no more than 4 pounds, and there were about 10 to 15 pennants missing, primarily from Box B-4 in the North line; all others were present. There were about a hundred or so pennants in the box line. The entire box line was removed and sent as waste to Idaho.

This worker also stated that there was no magnesium alloy used in any processes for development or fabrication of weapons components, and if there were significant quantities present, [this worker] would have known about it.

A third worker did not remember exactly what the conveyor system was made of, but did remember that the pennants that held the parts on the conveyor were a special alloy. This worker recalled completing and filing a special record for the waste recovered after the 1969 fire to account for any fire hazard in the waste being sent to Idaho. The worker thinks it was magnesium. The most important point is that there was no magnesium alloy used in any processes for development or fabrication of weapons components. The worker also said the conveyor system and pennants would have been an outside vendor fabrication. The worker added that it would have been highly unlikely that Rocky Flats would have purchased the magnesium alloy from Dow Madison and provided it to the vendor as government furnished equipment (GFE). This worker also acknowledged the use of magnesium salts in metal reduction processes.

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This worker also stated that if there were significant quantities of magnesium present at Rocky Flats, [this worker] would have known about it. The worker emphasized that because of the high degree of concern for fire potential, any materials used at Rocky Flats were reviewed and approved by the fire department prior to use. According to comments made during this interview, this worker would have known about any use of potentially flammable structural materials.

This worker also added that the weapons component specifications at Rocky Flats were very precise and any materials used were carefully evaluated for possible effect on the neutronic properties of the weapons components.

The fourth worker interviewed did recall that the conveyor line in B776 had some magnesium alloy, and some of it did burn in the May 11, 1969, fire. When asked for an estimate of the quantity of magnesium alloy in the box line, the worker said maybe a few hundred pounds at most, because "it goes a long way." If there were significant quantities of magnesium alloy present, this worker would have known about it. This worker stated that there was no magnesium alloy used in any processes for development or fabrication of weapons components.