

**REPORT TO THE ADVISORY BOARD ON RADIATION AND  
WORKER HEALTH**

*National Institute of Occupational Safety and Health*

**Audit of Case #PIID\* from the Blockson Chemical Company**

**Contract No. 200-2004-03805 Task Order No. 4**

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Prepared by

John Mauro, PhD CHP S. Cohen & Associates 6858 Old Dominion Road, Suite 301  
McLean, Virginia 22101

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**S. Cohen & Associates:**

***Technical Support for the Advisory Board on Radiation  
& Worker Health Review of NIOSH Dose  
Reconstruction Program***

**AUDIT OF CASE #PIID\* FROM THE  
BLOCKSON CHEMICAL COMPANY**

Task Manager: \_\_\_\_\_ Date: 02/04/05  
Hans Behling, PhD, MPH

Project Manager: \_\_\_\_\_ Date: 02/04/05  
John Mauro, PhD, CHP

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## SUMMARY

This report presents the results of an independent audit of a dose reconstruction performed by the National Institute of Occupational Safety and Health (NIOSH) for an energy employee that worked at the Blockson Chemical Company for **PIID\*** years, from **PIID\***, through **PIID\***. This time period included the time period when Blockson separated natural uranium from phosphate rock under contract to the U.S. Atomic Energy Commission (i.e., March 1952 to March 1962).<sup>1</sup>

As a result of his employment, the worker likely experienced internal exposures due to the inhalation of airborne particles of uranium oxide and external exposure from working in the vicinity of the separated uranium. In addition, the worker is believed to have had routine x-rays as part of Blockson's medical surveillance program. The worker was not provided with film badge or thermoluminescent dosimeters (TLDs) to measure external exposures, nor were bioassays performed to estimate internal exposures. As a result, exposures experienced by the worker were estimated using the exposure matrix provided in the site profile or Technical Background Document (TBD) prepared by NIOSH for the Blockson Chemical plant (ORAUT-TKBS-0002, June 29, 2004).

In **PIID\***, following his employment, the energy employee was diagnosed with cancer of the prostate gland. Table 1 summarizes the results of NIOSH's reconstruction of the doses to the energy employee's prostate gland for the purpose of deriving the probability of causation (POC) using IREP. Because ICRP 66 and IMBA do not provide the means to derive the doses to the prostate gland, NIOSH used the testes as a claimant-favorable surrogate for the prostate for both internal and external exposures. Table 1 also presents the results of the audit.

The results of the audit are expressed in terms of whether the exposures were derived in a scientifically valid manner and whether the doses were derived in a claimant-favorable manner. <sup>1</sup> The uranium content of phosphate rock is cited in the TBD as 0.014% U3O4 (Stolz, Jr. 1958).

**Table 1. Summary of Internal and External Exposure to the Testes (as a surrogate for the prostate gland) as Estimated by NIOSH, Along with the Audit Results**

Exposure Scenario	NIOSH Derived Annual Doses (rem)	Scientifically Valid?	Claimant Favorable?
Internal exposure from inhalation (alpha) during separations	~5E-4	Yes	We have some concerns
Internal exposures from ingestion (alpha) during separations	Negligible	We have some concerns	We have some concerns
Internal exposure from inhalation (alpha) of residual resuspended particles following the conclusion of separation operations	Negligible	We have some concerns	We have some concerns
External exposures during separation operations <b>(PIID*)</b>			
Ground surface contamination (chronic)	4E-2	We have some concerns	We have some concerns
Drum of aged yellowcake (chronic)	1E-1	We have some concerns *	We have some concerns
Submersion in airborne plume (chronic)	Negligible	Yes	NA**
Diagnostic x-rays (acute)	5E-3	Yes	We have some concerns
Chronic external exposure to residual contamination following the conclusion of separation operations in <b>PIID*</b>	4E-2	We have some concerns	We have some concerns

\* Our independent MicroShield and MCNP analyses revealed that the TBD underestimated the dose rates adjacent to the filled drum by about a factor of 5.

\*\* NA refers to not applicable.

## 1.0 INTRODUCTION

This report presents the results of an independent audit of a dose reconstruction performed by the National Institute of Occupational Safety and Health (NIOSH) for an energy employee that worked at the Blockson Chemical Company. This audit is one of several dose reconstruction audits being performed by S. Cohen & Associates (SC&A, Inc.) on behalf of the Advisory Board on Radiation and Worker Health.

Part one of this audit report presents a summary of our understanding of the doses derived by NIOSH, along with a brief description of the basic approach and assumptions employed by NIOSH to derive the doses. This material is extracted directly from the final dose reconstruction report published by NIOSH for this case, along with supporting documentation, including the Technical Basis Document for Blockson Chemical Company (ORAUT-TKBS-0002, June 29, 2004) and the discussions held with NIOSH during the factual accuracy review process. This section of the report summarizes our understanding of the methods used by NIOSH to reconstruct the doses to workers, and also serves as a baseline for the discussion and audit provided in Section 3 of this report.

Part two of the audit process (provided in Section 3 of this report) consists of an attempt to independently duplicate doses derived by NIOSH and a discussion of the validity of the methods employed. The doses selected for duplication are based on the judgment of the auditors as to the importance of the particular doses to the totality of the doses experienced by the energy employee. The reason for this step in the audit process is to provide the author, NIOSH, and the Advisory Board with a level of assurance that the auditors understand how NIOSH went about deriving the doses provided in their dose reconstruction report. In the process of duplicating the NIOSH derived doses, we also provide a critical review of fundamental data, information, models, and assumptions used by NIOSH to perform the dose reconstruction. This part of the audit explores the degree to which the data are adequate to support the dose reconstruction, and whether the models and assumptions adopted by NIOSH to perform the dose reconstruction are scientifically sound and claimant favorable. Areas where the methods are found to meet these criteria, or are deemed to be inadequate with regard to these criteria, are identified and discussed. The report is not exhaustive in the review of these matters, but is limited to those areas of inquiry that are judged by the auditors to be significant with respect to the dose reconstruction and the derivation of the probability of causation (POC).

Methods employed by NIOSH which are found to be either scientifically inappropriate or not necessarily claimant favorable are identified, but no attempt is made to correct these deficiencies and redo the dose calculations. It is assumed that NIOSH and the Advisory Board will have an opportunity to consider the results of this audit and determine whether a revision of the dose reconstruction is needed, and if so, how to go about making the necessary revisions.

## 2.0 SUMMARY OF DOSES

The energy employee worked at Blockson Chemical Company for **PIID\*** years, from **PIID\***, through **PIID\***. This time period included the time period when Blockson separated natural uranium from phosphate rock under contract to the U.S. Atomic Energy Commission (i.e. March 1952 to March 1962).<sup>2</sup> In **PIID\***, following his employment, the energy employee was diagnosed with cancer of the prostate gland. Table 2 presents the results of NIOSH's reconstruction of the doses to the energy employee's prostate gland for the purpose of deriving the probability of causation (POC) using IREP. Because ICRP 66 and IMBA do not provide the means to derive the doses to the prostate gland, NIOSH used the testes as a claimant-favorable surrogate for the prostate for both internal and external exposures.

The notations used in Table 2 to present the doses include the year in which the dose was received by the organ of interest, the statistical distribution that was used, and the key parameters for the distribution. For example, for exposure period number 1 in Table 2 (**PIID\***), a lognormal distribution was employed with a geometric mean of 1.89E-5 rem and a geometric standard deviation of 1.6 as the estimate of the internal alpha dose to the testes due to chronic inhalation of airborne uranium oxide (U<sub>3</sub>O<sub>8</sub>). A discussion of various types of statistical distributions and other parameters used as input to NIOSH-IREP is provided in NIOSH (2002). The external and internal doses to the organ of interest were determined by NIOSH to be 3.06 and 0.021 rem, respectively. The POC was determined by NIOSH to be 7.82% at the 99% confidence interval. On this basis, the claim was denied.

The final dose reconstruction report and the Technical Basis Document (TBD) provide detailed descriptions of the methods and assumptions used by NIOSH to derive the doses presented in Table 2. As may be noted, Table 2 presents doses in terms of annual doses due to internal alpha exposure resulting from the inhalation and ingestion of uranium, and external exposure from low energy and higher energy gamma emissions associated with working at the facility. These include external exposures due to working in the vicinity of the separated uranium and diagnostic x-rays performed as part of job-related routine medical surveillance. None of the dose estimates are based on the use of dosimeters worn by the energy employee, such as film badges or TLDs, which, if used, would have provided a generally reliable method for determining doses to the organ of concern. In addition, none of the dose estimates are based on bioassay data, such as urine or fecal analysis, or whole-body counting, which would have provided a generally reliable basis for estimating internal doses to most organs from the inhalation and/or ingestion of uranium. Instead, the doses were derived indirectly, using the generic methodologies described in the TBD. This section of this audit report briefly summarizes our understanding of how the TBD was used by NIOSH to derive the doses provided in Table 2. The reader is referred to the TBD for a more complete description of the generic methods developed by NIOSH for reconstruction of the doses to claimants who worked at Blockson Chemical Company.<sup>2</sup> The uranium content of phosphate rock is cited in the TBD as 0.014% U<sub>3</sub>O<sub>4</sub> (Stolz, Jr. 1958).

**Table 2. Dose Reconstruction as Reported by NIOSH Table 2. Dose Reconstruction as Reported by NIOSH (continued) Table 2. Dose Reconstruction as Reported by NIOSH (continued)**

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## 2.1 INTERNAL EXPOSURE TO ALPHA EMISSIONS

As indicated in Table 2, the relevant exposures are assumed to begin in **PIID\*** when the energy employee began work at Blockson, and continued until **PIID\*** when the energy employee's prostate cancer was diagnosed. According to Table 2, following a period of buildup, the concentration of uranium in the organ of concern approaches equilibrium, where the alpha dose is on the order of  $5E-4$  rem per year and remains at approximately this level almost until **PIID\***, when the cancer was diagnosed. Several alternative strategies and sources of information were considered by NIOSH as the bases for deriving these doses. One source of information was aerosol measurements performed in the packaging operations area of a uranium mill. The median aerosol concentration was observed to range from 40 to  $340 \mu\text{g U/m}^3$ , depending on the work area. Though the data were taken from studies performed at a uranium mill (see Eidson and Damon 1984), NIOSH believes that these measurements could serve as surrogates for the packaging operations at Blockson if uranium production rates are taken into consideration. Specifically, the uranium oxide production rate at uranium mills was about 4,000 lbs per day, while that at Blockson was about 137 lbs per day. On this basis, NIOSH estimated an airborne uranium dust loading of 1.6 to  $14 \mu\text{g U/m}^3$  at Blockson.

Independent of these measurements, NIOSH also estimated the potential inhalation exposures by developing models specifically applicable to Blockson operations. The starting point for the analysis was the assumption that, based on operational records, Blockson produced 50,000 pounds per year of 60%  $\text{U}_3\text{O}_8$  when operating at 365 days per year, or 137 lbs per day. The model further assumed that about  $1E-6$  of the daily production became aerosolized, citing a U.S. NRC report (2002) as the basis for this partition factor. Hence, NIOSH assumed that  $1.32E-4$  lbs/day of 60%  $\text{U}_3\text{O}_8$  became aerosolized. The model further assumed that the uranium that was aerosolized in a given day was uniformly mixed in the entire free volume of Building 55 and remained at that concentration in a steady state condition. Building 55 was built specifically to house the process used to separate uranium from the phosphate solution and had a free volume of about  $13,400 \text{ m}^3$ . This approach resulted in a chronic airborne concentration of natural uranium of  $3.8 \mu\text{g/m}^3$  or  $2.6 \text{ pCi/m}^3$  in Building 55, which is consistent with the estimate based on the above described uranium mill data after adjustments are made for uranium oxide throughput at Blockson versus a typical uranium mill. Using a breathing rate of  $1.2 \text{ m}^3/\text{hr}$ , the average inhalation rate of natural uranium by workers in **PIID\*** was estimated by NIOSH to be 25 pCi/day during separation operations.

A completely independent method was employed to estimate the quantity of uranium

inhaled by Blockson workers in **PIID\*** using urinalysis data collected for 25 workers over the time period between April 20, 1954, and February 2, 1958. Based on the assumption that the inhaled uranium was Absorption Type M (ICRP 1995) and using ICRP 66 default parameters, the chronic intakes for the workers for which bioassay data were available were found to have a mean of 24 pCi/day and a geometric standard deviation of 1.6. Given the agreement between the different methods for estimating uranium intake, this value was used by NIOSH as input to IMBA for reconstructing the inhalation doses to all atomic workers at Blockson during separation operations.

The TBD assumed uranium separation operations extended from the startup of research on the separation process at the plant on March 1, 1951, through March 31, 1962. For the energy employee, the period of exposure extended from the start of employment in **PIID\*** through **PIID\*** when uranium separation operations ceased, and then continued from **PIID\*** to **PIID\*** when the worker was diagnosed with prostate cancer. Following the completion of separation operations in March 1962, chronic inhalation exposures were assumed to continue because **PIID\*** was used for processing phosphate ore and was found to have residual uranium surface contamination of 640 dpm/100 cm<sup>2</sup>. Using a resuspension factor of 1E-6/m (NRC 2002), NIOSH estimated a residual air concentration of 0.03 pCi/m<sup>3</sup>. Assuming a breathing rate of 1.2 m<sup>3</sup>/hr for 2,000 hrs/y, the inhalation rate was estimated to be 71 pCi/y for the years following termination of uranium separation operations. This intake was considered negligible, and was not explicitly included in the dose calculations.

In addition to inhalation, NIOSH assumed that workers also inadvertently ingested 0.49 pCi/day of uranium using the generic methods described in NIOSH 2004. The doses to the most exposed organ (in this case bone) resulting from ingestion were determined by NIOSH to be less than 1 mrem/y. Because of this extremely small value, ingestion doses are not explicitly included as input into IREP.

Given the uranium inhalation rates described above, the internal alpha doses to the testes (which was used as a surrogate for the prostate gland, because the prostate is not included in current ICRP models) were derived using IMBA, which implements ICRP 66-recommended methodologies for deriving inhalation doses.

Exposures to radionuclides other than uranium and its short-lived progeny are not included in the dose reconstruction, because they were not part of the uranium chemical separation process. Though other radionuclides in the uranium series, such as Ra-226 and radon, were contained in the ore processed at Blockson, the TBD explains that these radionuclides followed the phosphogypsum separation step, which was not part of the uranium separation process. As such, exposures of Blockson workers to these other radionuclides would have occurred whether or not the uranium separation step was incorporated into the process.

## 2.2 EXTERNAL EXPOSURES

The separated uranium contains U-238, U-234, and U-235,<sup>3</sup> along with their short-lived progeny, which grew in after the uranium was separated from the ore. The short-lived progeny include Th-234 ( $T_{1/2} = 24$  days) and Pa-234m ( $T_{1/2} = 1.17$  minutes). These radionuclides emit mostly weak gammas, x-rays, and beta particles, with their associated bremsstrahlung radiation. In addition, Pa-234m decays to Pa-234, which emits a strong gamma with a branching fraction of 0.16%.

The TBD evaluated four sources of potential exposure to external radiation, including (1) submersion in air containing yellowcake dust, (2) exposure to surfaces contaminated with yellowcake dust, (3) exposure to contaminated skin, and (4) exposure to drums of yellowcake.

<sup>3</sup> The mass distribution of these radionuclides in naturally occurring uranium is 99.2475%, 0.0055%, and 0.72%, respectively.

The TBD concluded that submersion exposures were negligible, but the other pathways require explicit consideration. For the claimant, however, beta doses can be ignored, because the betas do not have sufficient penetrating power to expose the prostate gland.

Table 3 presents the external exposures experienced by the organ of interest (i.e., the testes as a surrogate for the prostate gland) as reported in the TBD. External exposures were assumed to continue beyond the conclusion of separation operations in March 1962 due to residual radioactive material in the workplace.

**Table 3. External Exposure to the Testes (as a surrogate for the prostate gland) Due to 365 Days of Exposure**

External Exposure Scenario	Annual Dose to Testes (rem)	
	E= 30-250 keV	E>250 keV
During Separation Operations (PIID*)		
Ground surface contamination (chronic)	1.9E-2	2.08E-2
Drum of aged yellowcake (chronic)	5.23E-2	4.68E-2
Diagnostic x-rays (acute)	5.0E-3	
Chronic residual exposures following the conclusion of separation operations in PIID*	1.9E-2	2.08E-2

External exposures due to uranium deposited on surfaces were estimated by using external gamma dose rate measurements taken at the facility in 1978. According to the survey, the median external exposure rate was 0.03 mR/hr, with a maximum of 0.3 mR/hr. The external dose to the organ of concern was derived using the measured median external exposure rate multiplied by the Exposure (R) to Organ Dose (rem) photon dose conversion factors from Appendix B of the NIOSH External Dose Reconstruction Implementation Guideline (NIOSH 2002a). The exposure geometry was assumed to be isotropic, and the exposure rate was divided evenly between the conversion factors for photons with energy between 30 and 250 keV and photons with

energy greater than 250 keV.

Direct external exposure to the filled drums of yellowcake was estimated by NIOSH using several methods, including the use of the computer codes MicroShield and MCNP, and direct measurements of the radiation field in the vicinity of similar drums containing UF<sub>4</sub>. The three methods produced similar results, except the direct measurements were slightly higher than the results obtained using MicroShield and MCNP (by about a factor of two). In order to be claimant favorable, the direct measurements were used by NIOSH to derive external doses from working in the vicinity of the containers. The results of the direct measurements were 1.3 mrem/hr on contact with the side of the drum, and 0.24 mrem/hr at 30 cm from the side of the drum. The median doses to the testes were derived assuming the worker spent 8 hours per day, 1 day a week, 50 weeks per year 1 foot (about 30 cm) from the container. Uncertainty in exposure was characterized by a lognormal distribution where the 95<sup>th</sup> percentile value was the dose associated with spending 40 hours per week, 50 weeks per year at 1 foot from the container. To convert the measured dose rate to the dose rate to the organ of interest, the Ambient Dose Equivalent (H\*(10)) to Organ Dose (H<sub>T</sub>) photon dose conversion factors provided in Appendix B of *NIOSH External Dose Reconstruction Implementation Guideline* (NIOSH 2002) were used. The exposure geometry was assumed to be anterior-posterior, and the dose rate was divided equally between photons with E=30-250 keV and photons with E>250 keV.

NIOSH also assumed that the workers received an annual x-ray as a part of routine medical surveillance of workers using the methods described by Kathren et al. 2003. Unlike the other exposures, these exposures were assumed to be acute, and the photon energies were assumed to be E=30-250 keV. The uncertainty in the x-ray exposures was assumed to be normally distributed, with a standard deviation of 30%.

NIOSH assumed that exposure of workers continued after the termination of uranium separation operation in March 1962 because of the presence of residual contamination. Based on a survey performed in 1978, the median external dose was found to be 0.03 mR/hr at 1 meter, with a maximum reading of 0.3 mR/hr. On this basis, the dose rate was assumed to have a lognormal distribution with a median of 0.03 mR/hr and a geometric standard deviation of 4.0. The dose rate to the organ of interest was derived by using the conversion factors in Appendix B of NIOSH 2002a. The exposure geometry was assumed to be isotropic and the dose rate was divided equally between photons with E=30-250 keV and photons with E>250 keV.

### **3.0 INDEPENDENT DUPLICATION AND REVIEW OF SELECTED NIOSH DERIVED DOSES**

This section presents a series of hand and computer calculations that attempt to duplicate selected doses derived by NIOSH. In so doing, we will have confirmed that we understand how NIOSH performed the dose reconstruction and that the calculations are correct, given the models and assumptions employed by NIOSH. In the process of attempting to duplicate the doses, we also discuss and critically review the data, models, and assumptions employed by NIOSH to reconstruct the doses.

#### **3.1 INTERNAL DOSE FROM INHALATION**

As indicated in Table 2, the annual alpha doses to the testes were determined by NIOSH to be about  $5E-4$  rem/y as the geometric mean of a lognormal distribution with a geometric standard deviation of 1.6. In this section, this dose is checked by reviewing the source documents, and by performing hand and IMBA calculations.

The starting point for this calculation is the cited uranium production rate of 50,000 lbs/y, or 137 lbs per day, 365 days per year. This value is based on the production limit established in the contract between the AEC and Blockson Chemical Company. DOE 1983 and 1985 were cited as the source of this information. We were unable to obtain copies of these documents and accepted this information on face value. Our first observation is that other credible estimates of the production rate of uranium oxide provided in the TBD include 400 and 600 pounds per day, but these values were not considered in the dose reconstruction. This decision does not appear to be claimant favorable. However, as discussed below, the uranium production rate was not a critical parameter in the derivation of uranium inhalation rates at Blockson.

NIOSH assumed that  $9.6E-7$  of the amount of uranium oxide that was produced each day became airborne and was uniformly mixed in the entire free volume of the building, which was cited as  $13,400 \text{ m}^3$ . This resulted in a steady state uniform airborne concentration of  $3.8 \text{ } \mu\text{g}/\text{m}^3$  throughout the building. This concentration corresponds to a total uranium inhalation rate of  $25 \text{ pCi}/\text{day}$ , based on an ICRP 66-recommended inhalation rate corresponding to 1/3 sitting and 2/3 light exercise, which corresponds to  $1.2 \text{ m}^3/\text{hr}$ . Given these assumptions, our independent calculations match the values estimated in the TBD. However, upon more detailed inspection of these assumptions, we have determined that there are fundamental flaws with this approach to deriving the uranium intake rate, as described below.

We confirmed that the resuspension factor of  $9.6E-7/\text{m}$  came from NRC 2002 and is recommended by the NRC for deriving upper-end indoor airborne concentrations of resuspended dust in work areas where the levels of surface contamination are known. Specifically, the product of the known surface contamination (expressed in units of

$\text{pCi/m}^2$ ), with the resuspension factor (expressed in units of  $1/\text{m}$ ), yields an estimate of the airborne radionuclide concentration due to resuspension processes, expressed in units of  $\text{pCi/m}^3$ . The TBD doesn't use the resuspension factor in this manner. It applies the resuspension factor to the quantity of uranium oxide produced per day. Mechanically, this bears no relationship to the purpose for which the resuspension factor was derived. The product of a resuspension factor, expressed in units of  $1/\text{m}$ , with the production rate of uranium oxide, in units of pounds/day, as used in the TBD, results in a meaningless value and is scientifically incorrect.

The appropriate approach would have been to derive a resuspension rate expressed in terms of the fraction of the uranium oxide production rate that becomes airborne per day. However, even assuming a valid resuspension rate could be developed for the uranium oxide production process employed at Blockson, it is inappropriate to assume that the amount of uranium oxide that becomes airborne each day can be simply divided by the free volume of the building to determine the steady state uranium oxide concentration in the building. In theory, if the rate at which uranium became airborne was known, along with an estimate of the rate at which the airborne uranium was removed by settling and building ventilation, the steady state average airborne concentration in the building could be estimated. However, even if this information were available, the derived airborne concentration would apply to the average concentration in the building and not to the areas where workers handled the drums. One would expect the airborne concentrations at these locations to be much higher than the average concentration in the overall building. We believe that the resuspension factor approach used in the TBD to derive airborne radionuclide concentrations in **PIID\*** should be deleted from the report.

The TBD uses other methods for deriving the airborne uranium oxide concentration and associated uranium inhalation rate that have more technical validity than the resuspension factor approach described above. Specifically, NIOSH refers to a study by Eidson and Damon (1984) of uranium oxide dust loadings in a uranium mill. We reviewed the cited document and confirmed that various areas in the mill observed median airborne dust loadings ranging from 40 to 340  $\mu\text{g U/m}^3$ . Since uranium oxide production rates in uranium mills is on the order of 4,000 lbs/day, as compared to 137 lbs per day at Blockson, one could argue that the airborne dust loading at Blockson could be estimated by prorating based on uranium oxide production rates. Using this method, one could derive a dust loading of about 1.6 to 14  $\mu\text{g U/m}^3$ , as cited in Table 3 of the TBD. It is probably a coincidence that this range is consistent with the values derived by NIOSH using the resuspension factor approach. However, we do not agree that the dust loading at a uranium handling facility is proportional to the production rate of uranium oxide at the facility. Air handling systems are designed to achieve a certain air turnover rate per hour. As a result, the ventilation rate at a larger facility, such as at a uranium mill, will likely be greater than at a smaller facility. As a result, the proportionality concept employed by NIOSH may not be scientifically correct. We suggest that NIOSH revisit this approach to deriving airborne dust loadings.

We acknowledge that NIOSH did not actually use the resuspension model or the uranium mill data surrogate model for deriving uranium inhalation rates at Blockson,

but instead relied on inhalation rates based on urinalysis data, discussed below. However, we believe that it is appropriate to point out the problems associated with the resuspension factor approach and uranium mill data surrogate approach because, if these methodologies were used at other facilities, they would be in error.

Finally, the TBD employs one additional method to estimate the inhalation rate of uranium for workers at Blockson. As described above, urinalysis data for 25 Blockson workers were found with daily uranium concentrations ranging from 0 to 0.017 mg/L. The average uranium concentrations for each worker over the time period urine samples were collected ranged from 0 to 0.006 mg/L. Using these data, the TBD estimates that the average chronic intake rate of total uranium was 24 pCi/day, with a geometric standard deviation of 1.6, assuming Absorption Type M.

Following our review meeting with NIOSH, NIOSH provided SC&A with the urinalysis data for the 25 workers. Among the 25 workers, NIOSH reports that the average daily uranium intakes range from 0 to 99 pCi/day assuming Absorption Type M, and 0 to 1680 pCi/day assuming Absorption Type S. NIOSH elected to employ 24 pCi/g, Absorption Type M, as representative of the inhalation rate of uranium for the workers at the facility. NIOSH cites this value as the average inhalation rate for the 25 workers.

Given a chronic intake of Absorption Type M uranium of 24 pCi/day, we used IMBA to determine the anticipated concentration of uranium in urine. Assuming a chronic average inhalation rate of 24 pCi/day of Absorption Type M uranium, and ICRP default assumptions, we determined, using IMBA, that the chronic excretion rate of uranium in urine would be 1.6 pCi/day. This translates to a chronic uranium excretion rate of about 0.0017 mg/day, or about one-third the highest average uranium concentration observed among the 25 workers. The implication is that, for the purpose of estimating the uranium inhalation rate, NIOSH assumed that the uranium inhalation rate was about one-third that associated with the maximum uranium inhalation rate among the 25 workers over the time period that urine samples were collected. A more claimant-favorable assumption would have been to assume that the uranium inhalation rate was closer to a value that is associated with the higher uranium excretion rates. For example, assuming the intakes are lognormally distributed, the upper 95% inhalation rate among the 25 workers over the time period that urine samples were collected is 53 pCi/day. A counter argument to this position could be that the urinalyses were performed on workers who were expected to have high-end exposures, and thus represented the critical population group. During our meeting with NIOSH, NIOSH explained that, of the 25 people sampled, 21 appear on more than one urinalysis report. This implies that the same small group of people were the only ones monitored, and that these people were selected because they were workers who had jobs with a high potential for exposure. This interpretation of the data seems reasonable.

An additional concern we have regarding the approach employed by NIOSH is the assumption that the chemical form of the inhaled uranium is Absorption Type M. On first inspection, it would appear that such an assumption is claimant favorable because the inhalation dose conversion factor for internal organs, other than the lung, is much

greater for Type M than for Type S uranium. However, for this case, the results of urinalysis were used as the basis for deriving uranium inhalation rates. This implies it would require a much larger quantity of Type S inhaled uranium to give the same concentration of uranium in urine as compared to the inhalation of Type M uranium. It is not immediately apparent whether Type S or Type M uranium is the more claimant-favorable assumption for reconstructing the doses for this employee. Some discussion of this matter is required. This matter was discussed with NIOSH during the January 12<sup>th</sup> and 13<sup>th</sup> review meeting, and NIOSH explained that there is little doubt that the uranium was Absorption Type M.

Assuming an inhalation rate of 25 pCi/day of Type M natural uranium and default ICRP modeling assumptions, NIOSH reports that an internal alpha dose to the organ of concern (i.e., the testes as a surrogate for the prostate) is about 5E-4 rem/yr. We confirmed this dose rate using IMBA.

### **3.2 INTERNAL DOSES DUE TO INGESTION**

Through screening calculations, the TBD (ORAUT-TKBS-0002) concluded that the doses to workers due to inadvertent ingestion of uranium oxide were negligible. The TBD estimated an ingestion rate of 0.49 pCi/day or (0.72 µg of natural uranium per day), which corresponds to an annual dose to the most exposed internal organ (bone surfaces) of less than 1 mrem. On this basis, the TBD eliminated ingestion from further consideration. We concur that a chronic ingestion rate of 0.49 pCi/day of uranium oxide would result in a dose to the gonads that is a small fraction of 1 mrem/y, and that the dose to bone surfaces is less than 1 mrem/y.

However, we are concerned with this analysis because it is based on a very small ingestion rate of uranium oxide; i.e., 0.72 µg/day. A review of the literature on this subject, as provided on page 4-17 of EPA 1977, reveals that the inadvertent ingestion of soot by adults is as low as 0.56 mg/day in typical living space (i.e., almost 1,000 times higher than the values used in the TBD for the ingestion of uranium oxide), to 110 mg/day while working in a dusty attic, to 480 mg/day during outdoor work in a garden. Assuming that the working environment at Blockson was not unlike a dusty attic, the ingestion rate of soot, including natural uranium, could have been as high as 93.5 mg/day, which corresponds to a dose of about 147 mrem/y to the testes at equilibrium, if the soot is assumed to be entirely uranium oxide. Inspection of Table 2 indicates that this dose would by far dominate all other sources of exposure and should have been explicitly considered in the dose assessment. We recognize that it would be unrealistic to assume that all the soot inadvertently ingested by workers would be uranium oxide. However, this calculation reveals that inadvertent ingestion of uranium oxide could have been an important contributor to the internal dose, as compared to the inhalation dose, and should have been explicitly included in the analysis. We suspect that an inadvertent ingestion rate 0.72 µg/day of natural uranium employed by NIOSH is unrealistically low.

During our review meeting, NIOSH explained that, since the intake rates for uranium are based on urinalysis data, which integrate across all pathways, such large ingestions of uranium would have been detected in the urinalysis data. Hence, such large intakes are not likely given the results of the urinalysis. We concur in this observation. However, NIOSH did indicate that, though not an issue in this case, NIOSH will give explicit consideration of the inadvertent ingestion rates, as cited by EPA in the Exposure Factors handbook, in future dose reconstructions.

### **3.3 EXTERNAL ANNUAL DOSES TO TESTES DUE TO SUBMERSION IN AIR CONTAINING YELLOWCAKE DUST**

The TBD used the airborne concentration of natural uranium derived from the bioassay results to calculate the external dose to the testes due to submersion in air containing yellowcake dust. NIOSH concluded that this dose was negligible (below 1 mrem/year), and our analysis revealed the same result. We calculated the dose to the testes due to submersion in air to be  $5E-5$  mrem/year, far below 1 mrem/year. We concur with NIOSH's conclusion that external exposures to the organ of concern from airborne dust were negligible.

### **3.4 EXTERNAL ANNUAL DOSES TO TESTES DUE TO PHOTONS EMANATING FROM DRUMS OF YELLOWCAKE**

The TBD assumed that 50 drums of yellowcake were loaded and packed each year, and that each drum contained 1,000 lbs of uranium oxide. This assumption is consistent with the reported contractual production rate of uranium oxide. In addition, our independent analysis revealed that a single 55-gallon drum could hold about 1,000 lbs of uranium oxide if filled completely and slightly compacted. Based on an interview with a claimant, NIOSH also determined that a typical worker spent 8 hours per day, 1 or 2 days per week loading drums onto trucks and boxcars. Given this information, NIOSH assumed that the worker spent 8 hours per day, 1 day per week (i.e., 400 hours per year) at a distance of 30 cm from the drums, and that uncertainty in the duration of exposure was lognormally distributed with a 95% confidence level based on an exposure duration of 40 hours per week, 50 weeks per year (i.e., full time at 2,000 hours per year). The implication is that some workers may have spent full time in the vicinity of the drums involved in various activities associated with the loading, sampling, sealing, and unsealing the drums, and the general handling of the drums.

The derivation of the external exposures to the drums also required a determination of the radiation field in the vicinity of the drums, and converting the external exposure to the dose to the organ of interest. The external exposures were determined by NIOSH based on the application of models and also actual measurements performed on drums filled with UF<sub>4</sub>. The UF<sub>4</sub> measurements revealed an external dose rate of 0.24 mrem/hr at 30 cm, which is about a factor of 2 higher than the exposure rates obtained using models. On this basis, NIOSH estimated a median external dose of 0.096 rem/y. Using the dose

conversion factors in Appendix B of NIOSH 2002, the dose to the organ of interest is estimated in the TBD to be 0.0991 rem per year. Using these same assumptions, we obtained comparable results; i.e., 0.095 rem/y. In addition, in order to verify the external dose rates obtained by NIOSH using models and the UF<sub>4</sub> drum measurements, we performed an MCNP and MicroShield simulation for 1,000 lbs of pure U<sub>3</sub>O<sub>8</sub> in a 55-gallon drum. Appendix A presents the results of our independent calculations. Appendix B presents the photon energy spectrum used as the basis for our calculations. The results of independent analysis yields dose rates that are several times higher than those reported in the TBD. Specifically, Table 7 of the TBD estimates an air dose rate of 8.4E-2 mR/hr at 30 cm from the surface of a drum containing 1,000 lbs of uranium oxide. Our MicroShield calculations yield an air exposure rate of 0.36 mR/hr. Similarly, the TBD estimates an H(10) dose of 0.13 mrem/hr at 30 cm from the surface of the drum, while we obtained a dose rate of 0.66 mrem/hr.

Based on our calculations, we cannot verify that the external dose rate from drum handling reported in the TBD are scientifically correct and claimant favorable. In fact, based on the investigations provided in Appendix A, we believe an error was made in NIOSH calculations, which has resulted in an underestimate of the external doses. During our meeting, NIOSH stated that it is investigating this discrepancy between our MCNP and NIOSH MCNP calculations.

NIOSH also explained that, since the reconstructed external doses from the drums was actually based on empirical data from measurements made on drums containing UF<sub>4</sub>, they would sooner rely on these measurements than the MCNP calculations. We do not concur with this position because, given the defined exposure conditions, it is more likely that the MCNP values are more reliable than measurements that either may be in error or may not be representative of the defined exposure conditions.

### **3.5 EXTERNAL ANNUAL DOSES TO TESTES DUE TO PHOTONS FROM CONTAMINATED SURFACES**

As described above, the TBD estimated the external doses to the testes from contaminated surfaces to be 0.0398 rem/y, based on 1978 radiation survey data that revealed a median external exposure rate of 0.03 mR/hr with a maximum of 0.3 mR/y. These values were converted to doses to the testes using the dose conversion factors in Appendix B of NIOSH 2002, and assuming that 50% of the exposure is due to E=30-250 keV photons and 50% is due to E>250 keV photons. Using these, we obtain the same dose, as follows:

$$[(0.03 \text{ mR/hr} \times 0.5 \times 0.632) + (0.03 \text{ mR/hr} \times 0.5 \times 0.693)] (2,000 \text{ hrs/y}) = 0.04 \text{ rem/y}$$

We have two concerns with this method for deriving external doses from contaminated surfaces. First, the measurements are based on 1978 data, i.e., 16 years after the termination of separation operations at the facility. Clearly, residual contamination remaining on surfaces between 1962, when uranium separations ceased, and 1978, when the surveys were performed, would have declined due to general housekeeping and other natural attenuation processes. In fact, ORAUT-OTIB-0004 refers to 1% per day as the

natural removal rate for residual contamination of surfaces. Clearly, it would be inappropriate to use the 1% per day depletion rate to back-calculate the residual surface contamination levels that would have been present on surfaces prior to the time when the surveys were performed (i.e., 1978) because of the enormously unrealistic contamination levels that would be derived. However, it would appear that the approach used by NIOSH to derive external doses from residual radioactivity is not claimant favorable, because it did not take into consideration the natural depletion of residual radioactivity.

The second concern we have with the approach used by NIOSH to derive external doses from residual contamination is that dose estimates were based on the median value of the measurements made in 1978. It would seem to be more reasonable and claimant-friendly to employ a high-end value of the measurements, perhaps the 90<sup>th</sup> percentile level, as opposed to the median value.

In its response to this concern, and at the review meeting, NIOSH indicated that it is evaluating these concerns.

### **3.6 EXTERNAL DOSES TO TESTES DUE TO ROUTINE, WORK-RELATED MEDICAL X-RAY EXAMINATIONS**

The 5 mrem/y dose to the testes from medical examinations of workers at Blockson, as estimated in the TBD, is based on the assumption that workers received a single conventional posterior-to-anterior x-ray annually. This estimate of frequency of exposure and dose to the testes is based on guidance provided in Table 4.0-1 of ORAUT-OTIB-0006 for pre-1970 exposures. In accordance with the guidance, and lacking information to the contrary, it would have been more claimant favorable to assume that at least some examinations included photofluorography, because such examinations were a matter of standard practice prior to 1960. Table 3.3-1 of the guideline recommends a default entrance kerma dose for a PA chest x-ray pre-1970 of 200 mrad, and a corresponding gonadal dose of 5 mrem per x-ray. However, the guideline also recommends a default pre-1970 photofluorographic entrance kerma dose of 3,000 mrem, with a corresponding gonadal dose of 75 mrem per photofluorograph. If the worker had received some photofluoroscopic examinations, the annual doses to the organ of interest would have been substantially greater than the values employed in the dose reconstruction.

In its response to this concern, and at the review meeting, NIOSH indicated that it is evaluating these concerns.

### 3.7 CONCLUSIONS

We have found that several aspects of the dose reconstruction for this claimant were either scientifically inappropriate and/or not claimant favorable, as follows:

1. 1. The worker may have experienced inhalation doses that were higher than the average values experienced by workers at the facility. It is possible that the responsibilities of the worker placed him at locations in the facility where the airborne dust loadings were substantially higher than the average values at the facility. The Computer Assisted Telephone Interview (CATI) report summarizing the results of interviews with the survivor of the worker indicates that the worker was an **PIID\***. As a **PIID\***, we assume that he helped to fill the containers with uranium oxide, which placed him at locations where his potential for inhalation exposure was higher than the average. However, given that the 25 workers for which urinalysis data were collected represented the critical group, as suggested by NIOSH, then the reconstructed inhalation doses for this worker would not be unreasonable.
2. 2. An independent calculation of the radiation field in the vicinity of the 1,000-lb drums of uranium oxide appears to indicate that the TBD underestimated the radiation field in the vicinity of the drums by about a factor of 5.
3. 3. An inhalation rate of 1.2 m<sup>3</sup> per hour might underestimate the actual breathing rate if the nature of the work involved strenuous exercise. For example, sealing and unsealing the containers and loading the containers on trucks may have required more strenuous activities than that corresponding to 1/3 sitting and 2/3 light exercise.
3. 4. The quantity of uranium oxide that NIOSH assumed to be inadvertently ingested appears to be extremely small compared to the default soot ingestion rates recommended by EPA for use in risk assessment calculations.
4. 5. The exposures associated with routine medical x-rays may have been significantly underestimated because they did not take into consideration the possibility that the worker received periodic fluoroscopic examinations.

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## **APPENDIX A: EXTERNAL EXPOSURE TO DRUM OF YELLOWCAKE**

The dose rate due to external exposure to a drum of yellowcake is calculated in order to

estimate the dose to the prostate of a claimant. Since the prostate is not one of the organs for which tissue weighting factors were recommended by the ICRP (1991), the testes are used as a surrogate. This substitution is client-favorable because the testes are superficial to the body, while the prostate is situated deeper within the body. The overlying tissue offers some radiation shielding to the prostate, which is not afforded to the testes.

Two methods are employed for these calculations. The first utilizes the Los Alamos Monte Carlo code MCNP5 (LANL 2003). The second uses MicroShield Version 4.21 (Grove Engineering 1995). We will first describe the exposure geometry for the MCNP calculations.

The radiation source is yellowcake, assumed to be pure  $U_3O_8$ , in a 55-gal drum (see illustration on the right). According to a specification of the Oak Ridge National Laboratory for carbon steel open-head drums (ORNL No. 100-1A2-0006), 55-gal drums have a nominal thickness of 1.5189 mm. According to the Steel Shipping Container Institute (1997), 55-gal drums have an inside diameter of 22½ in. The height of nominal 55-gal drums varies among different manufacturers. A nominal overall height of 34 in was adopted for the present analysis. The resulting volume is slightly more than 55 gallons. The yellowcake is assumed to completely fill the actual volume of this Drum of Yellowcake container. The density is calculated to be 2.055 g/cm<sup>3</sup>.



The drum is constructed of ASTM A 366 steel, as cited by ORNL No. 100-1A2-0006. The steel is assigned a nominal density of 7.86 g/cm<sup>3</sup>. The average elemental composition of the alloy is listed in Table A-1.

**Table A-1. Elemental Composition of Steel Drum**

Element
C
P
S
Mn
Fe
Total

**Mass fraction**

8.5E-4

1.5E-4

1.75E-4

0.003

0.996

1.000

The drum is located in the center of a stylized cylindrical room, 3 m high, with a radius of 5 m, which is filled with moist air. The walls, floor, and ceiling are concrete, which is optically thick. The dose rates are calculated at two locations, 30 cm and 100 cm from the exterior of the drum, at a height of 77.9 cm above the floor. This height is the position of the testes in the BodyBuilder, a commercial MIRD phantom computer program from White Rock Science, which is one of the phantoms used by the Los Alamos National Laboratory for MCNP analyses. (The phantom is not used in the present analysis).

The yellowcake is assumed to have the isotopic ratio of natural uranium, listed in Table A-2. The specific activities of the three uranium isotopes are calculated from the isotopic composition and the uranium fraction of  $U_3O_8$ . Uranium-238 is assumed to be in equilibrium with its short-lived progeny; Th-234, Pa-234m, and Pa-234. The 6.7-hour Pa-234 is the product of the isomeric transition of Pa-234, which occurs in 0.16% of the disintegrations. Uranium-235 is assumed to be in secular equilibrium with Th-231.

<b>Table A-2. Specific Activity of Yellowcake Nuclide</b>	<b>Mass Fraction</b>	<b>Specific Activity (Bq/g)</b>
U-234	0.000054	1.04E+04
U-235	0.007204	4.82E+02
Th-231	—	4.82E+02
U-238	0.992742	1.05E+04
Th-234	—	1.05E+04
Pa-234m	—	1.05E+04
Pa-234	—	1.68E+01

**MCNP Analysis**

The MCNP calculations are performed in stages. In the first stage, the photon fluence at the dose point is calculated for each of 11 monoenergetic photons, ranging in energy

from 15 keV to 4 MeV. The photon fluence is converted to air kerma using the conversion coefficients listed in Table A.1 of ICRP 1996. The dose to the testes is calculated from the air kerma using the data for the anteroposterior (AP) exposure geometry in Table A.8 of ICRP 1996. The doses from the uranium isotopes and their short-lived progeny are calculated by cubic spline interpolation, as described by Eckerman and Ryman (1993). The dose from external bremsstrahlung from  $\beta$  rays and other electrons are calculated in the manner described by Eckerman and Ryman (1993, Appendix C). The radionuclide data are taken from the compilation by the Tokai Research Establishment, JAERI (2001).

The results of the MCNP analysis are listed in Table A-3.

**Table A-3. MCNP Dose Calculations**

Dose rate (mrad/h)	Distance from edge (cm)	
	30	100
Air kerma	0.55	0.15
Testes	0.66	0.18

### MicroShield Analysis

The analysis was repeated using MicroShield Version 4.21. MicroShield is a point-kernel code that is quick and easy to execute. It uses line-of-sight attenuation of the intervening material and empirical buildup factors. It includes a built-in library of radionuclides; the compilation date of this library is not specified, but it most likely predates the initial release of MicroShield 4.0 ca 1993. Consequently, there may be differences in the  $\gamma$ -ray spectra used in the MCNP and the MicroShield analyses.

MicroShield has 12 built-in materials. The program allows the user to create custom materials by specifying the elemental composition. The attenuation can be correctly calculated for such materials; however, buildup data exists only for the built-in materials and has not been experimentally verified for user-specified materials. The present MicroShield analysis is therefore restricted to the built-in materials, which include uranium and iron.

The MicroShield analysis uses a simplified version of the model used in the MCNP calculations. The yellowcake is modeled as pure uranium, one of the 12 built-in materials; however, the specific activities are those calculated for  $U_3O_8$ . The density is the same as used in the MCNP analysis. The steel drum is modeled as an open cylindrical shell of pure iron. Since MicroShield does not include scatter from objects not in the line-of-sight path from the source to the receptor, the top and bottom of the drum are omitted, as is the concrete room. The analysis is performed only at one distance—30 cm from the outside of the drum. The vertical position of the dose point is the same as in the MCNP run.

The air kerma at 30 cm is 0.32 mrad/h—somewhat lower than the corresponding

MCNP result. This is not unexpected for several reasons:

- MicroShield does not calculate bremsstrahlung from  $\beta$  rays and other electrons.
- Pure uranium has somewhat higher self-shielding than  $U_3O_8$ .
- Air kerma was calculated for the MCNP results using the conversion coefficients presented in ICRP 1996, while MicroShield uses the earlier (1987) coefficients. The later coefficients are slightly higher at high energies.

### Comparison with ORAU Technical Basis Document

Table A-4 compares the present results with those listed in the Technical Basis Document for the Blockson Chemical Company.

<b>Table A-4. Comparison of SC&amp;A and ORAU Exposure and Dose Calculations</b>	<b>Calculation</b>	<b>Distance from edge (cm)</b>	
		<b>30</b>	<b>100</b>
MicroShield exposure (mR/h)	SC&A <sup>a</sup>	0.360	—
	ORAU <sup>b</sup>	0.084	0.022
Dose to testes (rad/h)	SC&A <sup>a</sup>	0.672	0.185
Dose (mrem/h)	ORAU <sup>b</sup>	0.130	0.036

a

Present analysis b Technical Basis Document for the Blockson Chemical Company, Table 7.

The SC&A results are approximately 5 times as high as the ORAU values. The ORAU analysis used an exposure geometry similar to that in the present analysis. The thickness of the steel drum was almost identical (1.519 mm vs. 1.52 mm in the present analysis). The inner radius of the drum was 28.106 cm vs. 28.575 in the present case, while the height was 84.15 vs. 86.36 cm (Hertel 2004). These dimensions are quite similar and should not lead to any significant discrepancies. We do not know the height above ground of the dose point in the ORAU analysis.

There are several reasons for the discrepancy between our results and the ORAU analysis. One difference is that, in the absence of data on the composition of the yellowcake, we made the claimant-favorable assumption that it was pure  $U_3O_8$ , whereas ORAU assumed that it was 50%  $U_3O_8$  and 50%  $UO_2PO_4 \cdot 4H_2O$ . The specific activity of the uranium isotopes in such a mixture is about 82% of that in pure  $U_3O_8$ , leading to a proportionately lower dose rate. A countervailing effect would be a somewhat smaller self-absorption of the emitted photons due to the lower effective atomic number of the mixture assumed by ORAU.

The most significant difference is the a priori assumption made in the ORAU analysis that the only significant radiation would be bremsstrahlung generated by the  $\beta$  decay of the short-lived daughter products of U-238. Our analysis, based on pure  $U_3O_8$ , indicates that bremsstrahlung contributes less than 20% of the total external dose rate. Consequently, by assuming that only bremsstrahlung significantly contributed to the external dose, the ORAU analysis overlooked over 80% of the dose that is due to the x-

ray and  $\gamma$ -ray photons. The complex decay scheme of U-238 and its short-lived progeny is discussed in Appendix B of this report.

Another, albeit minor, discrepancy is due to ORAU's use of the ANSI/ANS-6.1.1-1977 fluence-to-dose rate conversion factors, rather than the organ-specific coefficients of ICRP 1996. For most of the energy range of interest, the ANSI/ANS factors lead to a slightly higher dose rate.

We conclude that ORAU seriously underestimated the external dose rate from the drum of yellowcake. Our calculated rate is not only higher than that calculated by ORAU, it is also significantly higher than the rate of 0.24 mrem/h measured at a distance of 30 cm from a drum of UF<sub>4</sub>.

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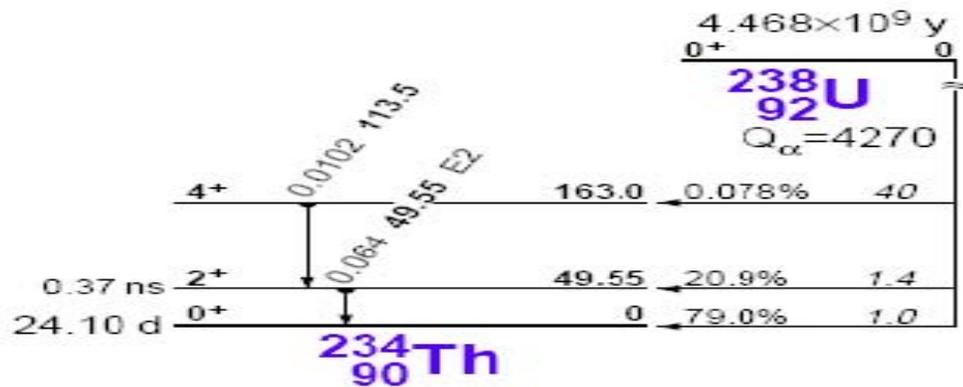
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### **B-1 Decay of U-238 to Th-234**

Uranium-238 decays by  $\alpha$  emission to Th-234, which has a half-life of 24.1 d, 100% of

the time. Alpha particles of three discrete energies are emitted. Seventy-nine percent of the transitions are to the ground state of Th-234. The remaining 21% are to one of two excited states. In the latter case, the decay is accompanied by  $\gamma$  emissions with very low intensities: a 113.5 keV  $\gamma$  ray is emitted with an intensity of  $1.02 \times 10^{-4}$  (i.e., ~ 1 in 10,000 decays), while a 49.55 keV  $\gamma$  has an intensity of  $6.4 \times 10^{-4}$ . **Figure 0. Decay of  $^{238}\text{U}$**

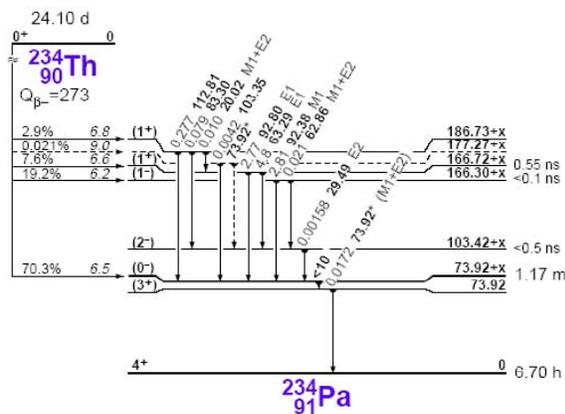


(Firestone et al. 1999)

(The intensities shown in Figure 1 are in percent).

### B-2 Decay of Th-234 to Pa-234m

Thorium-234 decays by  $\beta$  emission to Pa-234m, which has a half-life of 1.17 m, 100% of the time. The decay is to one of four (possibly five) excited states of Pa-234. (These transitions are illustrated in the upper part of Figure 2). In the case of the three (or four) higher level states, the decay is accompanied by the emission of  $\gamma$  rays ranging in energy from 112.81 to 20.02 keV. The lowest-lying state is the metastable Pa-234m. **Figure 0. Decay of  $^{234}\text{Th}$**



(Firestone et

al. 1999) B-3 Decay of Pa-234m

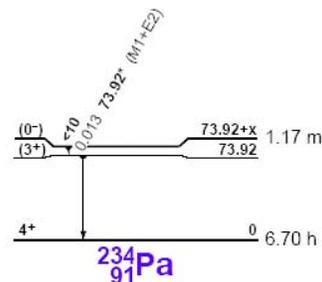
Protactinium-234m has two distinct decay modes. In 99.84% of the transitions, it decays to U-234. In the remaining 0.16% of the decays, the transition is to the ground state of Pa-234. The latter transition is illustrated in the lower part of Figure 2, and again in Figure 3. This transition is accompanied by the emission of a 73.92 keV  $\gamma$  ray about 10% of the time.

In 98.2% of the Pa-234m decays, the transition is to the ground state of U-234, as shown in Figure 4. This transition is accompanied by the emission of  $\beta$  rays with energies up to

Figure 0. Decay of  $^{234m}\text{Pa}$

about 2.2 MeV. The only  $\gamma$  emission in such cases is from the buildup (Firestone et al. 1999)

of Ra-226 and its progeny, which begins immediately but proceeds very slowly, due to the long half-lives of the intermediate members of the decay chain. In the remaining 0.64% of the decays, the transition is to one of over 20 excited states of U-234. These transitions result in the complex  $\gamma$ -ray spectra illustrated in Figures 4 and 5. The  $\gamma$  rays have energies up to almost 2 MeV. However, most have very low intensities.





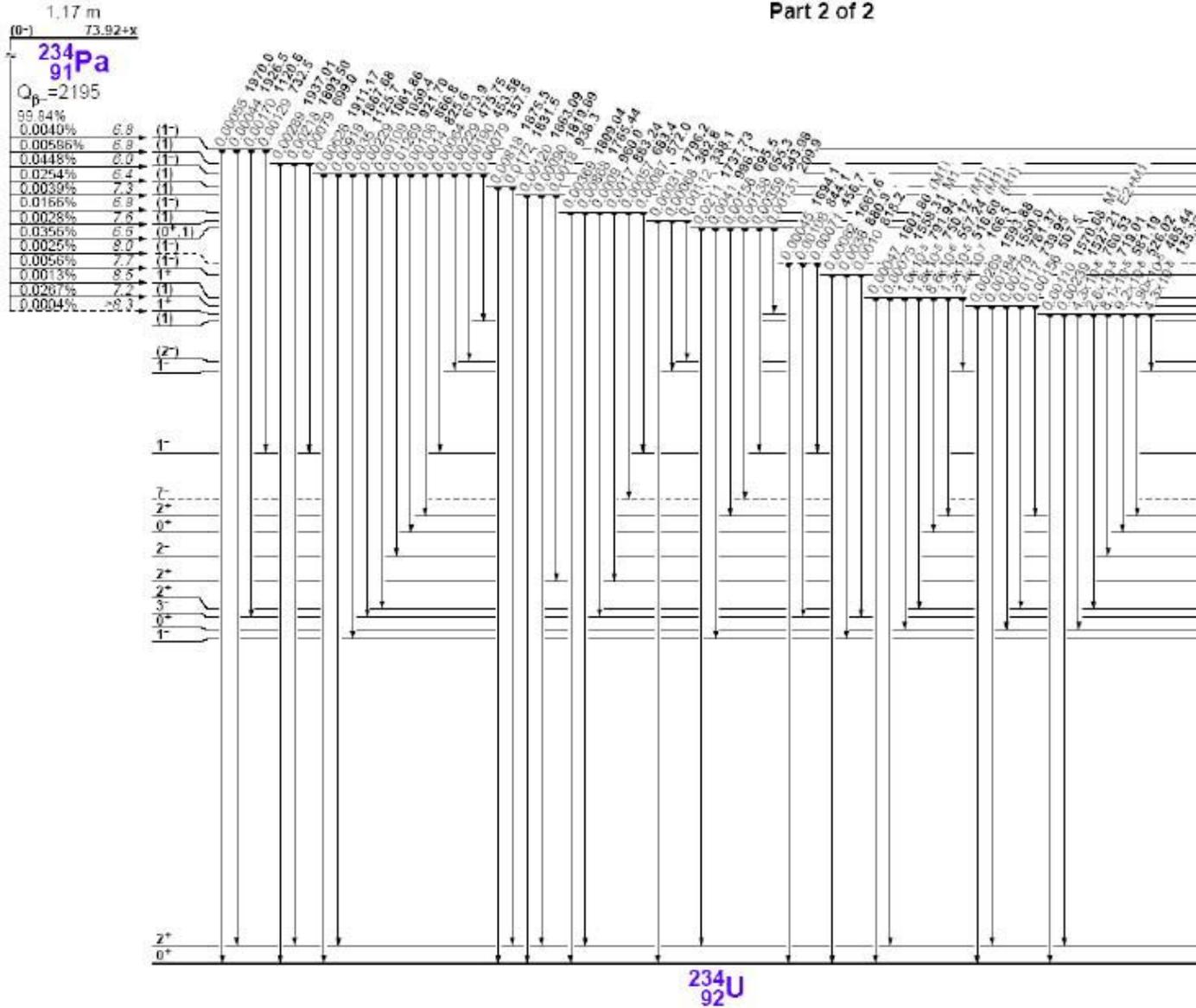
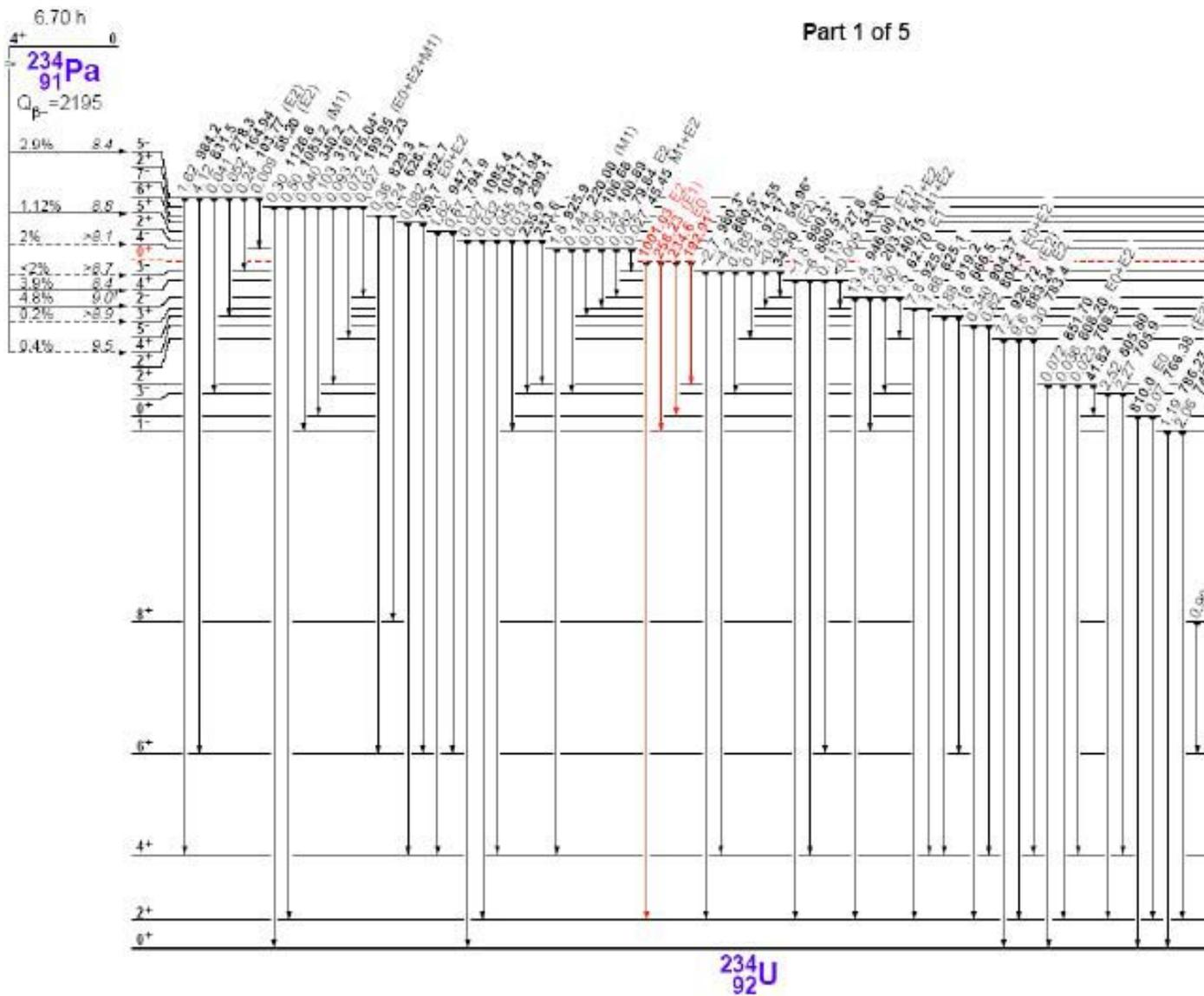


Figure 5. Decay of  $^{234m}_{91}\text{Pa}$  to  $^{234}_{92}\text{U}$ —Part 2 (Firestone et al. 1999)

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**Figure 6. Decay of  $^{234}_{91}\text{Pa}$  to  $^{234}_{92}\text{U}$  (Firestone et al. 1999)**

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### B-4 Decay of Pa-234

Protactinium-234 undergoes  $\beta$  decay to U-234. All of the transitions are to excited states of U-234. These transitions produce a very complex  $\gamma$ -ray spectrum, with energies ranging up to almost 2 MeV. Portions of these transitions are illustrated in Figure 6.

## **REFERENCE TO APPENDIX B**

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