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Dose Reconstruction Project for NIOSH

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Subject Expert(s): James M. Mahathy

Site Expert(s): Melton H. Chew

Approval:	Signature on File James M. Mahathy, Document Owner	Approval Date:	01/20/2011
Concurrence:	Signature on File James P. Griffin, Deputy Project Director	Concurrence Date:	01/20/2011
Concurrence:	Signature on File Kate Kimpan, Project Director	Concurrence Date:	01/20/2011
Approval:	Signature on File James W. Neton, Associate Director for Science	Approval Date:	01/24/2011

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PUBLICATION RECORD

EFFECTIVE DATE	REVISION NUMBER	DESCRIPTION
01/24/2011	00	This report describes activities performed by the Savannah River Site (SRS) related to the design of processes and facilities for separating ^{210}Po from ^{210}Bi with the objective of determining the feasibility of reconstructing internal and external doses potentially received from exposures to polonium. Incorporates NIOSH formal review comments. Training required: As determined by the Objective Manager. Initiated by James M. Mahathy.

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ACRONYMS AND ABBREVIATIONS

AED	Atomic Energy Division
^{o}C	degrees Centigrade
Cu	curie
d	day
g	gram
in.	inch
kg	kilogram
MeV	mega-electron volt
mg	milligram
NASA	National Aeronautics and Space Administration
NIOSH	National Institute for Occupational Safety and Health
SRDB	Site Research Database
SRL	Savannah River Laboratory
SROO	Savannah River Operations Office
SRS	Savannah River Site
yr	year

1.0 SUMMARY

This report describes activities performed by the Savannah River Site (SRS) related to the design of processes and facilities for separating ^{210}Po from ^{210}Bi with the objective of determining the feasibility of reconstructing internal and external doses potentially received from exposures to polonium. A smaller project was performed in 1967 to test perovskite mixed oxides as host lattices for a fully oxidized ^{210}Po fuel form.

This report does not provide models for assessing external and internal exposures; those will be published in a revision to the SRS site profile document (ORAUT 2005). To perform this analysis, the National Institute for Occupational Safety and Health (NIOSH) considered documentation stored in the NIOSH Site Research Database (SRDB) and interviewed a former SRS research engineer. NIOSH has determined that work to separate ^{210}Po from irradiated bismuth was carried out only in a laboratory-scale project conducted in a glovebox. Other work in support of moving to full production was performed with the use of stand-in chemicals such as cadmium. NIOSH determined that workers involved in the laboratory-scale project were monitored for internal and external exposures (electron and photon). Therefore, NIOSH finds that worker doses from potential exposures to polonium can be reconstructed.

2.0 INTRODUCTION

SRS first discussed potential separation of ^{210}Po with the U.S. Atomic Energy Commission Savannah River Operations Office (SROO) in the early 1960s (Morris 1963). The ^{210}Po was intended for use by National Aeronautics and Space Administration (NASA) programs. SROO approved a laboratory-scale project for SRS to provide equipment and to develop a vacuum distillation separation process. While the laboratory-scale project was being ramped up, SRS engineers designed a production separation facility; the ultimate goal was to move to production separation of up to 3 g of ^{210}Po per day (10 kg/yr) (DuPont 1964). For additional verification, SRS operated a pilot plant operation with higher throughput than the laboratory-scale project.

3.0 LABORATORY-SCALE PROJECT

On May 26, 1964, SRS was authorized by SROO to prepare a laboratory for designing and testing a program for separating ^{210}Po from irradiated bismuth by vacuum distillation (DuPont 1964) for use in the space program (Mahathy 2010). The authorization called for setting up the laboratory in Building 773-A, Room B-102, with construction work to be completed by December 5, 1964. The intent of the laboratory project was to derive experimental quantities of bismuth with a ^{210}Po yield of 0.3 g per trial. The authorization stated that no further containment facilities would be required for completion of the development work (DuPont 1964); however, NIOSH has no documentation on when the work to prepare Room B-102 was completed.

Designed air-tight containment was required for the laboratory-scale work because of the extremely high alpha activity (4,500 Cu/g) and the volatility of the ^{210}Po. SRS fabricated a two-compartment glovebox of stainless steel and safety glass in Room B-102 for use in the ^{210}Po program. The glovebox was supported by an understructure. Two bag ports were added (16- and 18-in.) along with an exhaust system and services such as electricity and piping. The glovebox was installed adjacent to an existing hood that was modified for use with the two ports (DuPont 1964). After initial completion of the glovebox, a distilling device was installed using a 2-in.-diameter still with tantalum liners (Mahathy 2010; Bradley, Landon, and Gibson 1970). The glovebox contained heavy, flexible Hypalon gloves.

The laboratory-scale project was started in 1965 under the guidance of a research engineer working with two laboratory technicians. The vacuum distillation process was first documented in 1966

(Bradley, Landon, and Gibson 1970). The process was tested using chemical stand-ins for polonium, including cadmium and magnesium (Bradley, Landon, and Gibson 1970). The research engineer was brought into the laboratory-scale project in June 1966. Three SRS managers/supervisors were involved in the project but did not routinely enter Room B-102. The research engineer occupied an office (Room B-104) next door to Room B-102. At least one of the workers, the research engineer, visited Mound Laboratory for about a week in 1966 to observe operations involving ^{210}Po (Mahathy 2010).

After irradiation of bismuth metal, molten bismuth was transferred for chemical decanting. First, the molten metal flowed through an underflow weir to trap slag; then it flowed through a valve that could be closed by freezing the metal. The molten bismuth was held for several hours in a tantalum getter bed at 750^{o}C, which removed entrained bismuth oxide and dissolved oxygen. All distilling work performed with ^{210}Po in the laboratory-scale project was done in the containment of the glovebox, piping, and exhaust venting in Room B-102 (Mahathy 2010). One source stated that up to four trials of the dissolution process were conducted in 1967 (Mahathy 2010), which is consistent with the number of irradiated bismuth events conducted from February 1, 1967, through May 2, 1967, as part of the Curium-II process (Gray 1967). A former research engineer said one trial with ^{210}Po was conducted in 1966 with others being done in 1967. It did not begin until 1967 (Mahathy 2010). One trial was documented by the Savannah River Laboratory (SRL) to have taken place in June 1968 (Copley and Moyer 1968a); SRS Health Physics monitored B-102 for exposure and contamination (Mahathy 2010). Personnel wore laboratory coats when working in Room B-102. They wore rubber gloves taped to the sleeves of their laboratory coats and company-provided shoes or shoe covers that were not worn outside Building 773-A. Laboratory personnel performed frequent contamination monitoring of their garments, hands, and bodies. SRS Health Physics performed routine surveys of Room B-102 (Mahathy 2010). Air sampling was performed in Room B-102 (Mahathy 2010). Laboratory personnel did not use respiratory protection, although the research engineer wore such protection while visiting the Mound Laboratory to work on its polonium process. It was implied that the risk of airborne release of polonium was much less with the SRS laboratory-scale process than with the Mound process. However, due to the high (5-MeV) alpha energy, polonium could diffuse through the Hypalon gloves. To prevent escape of ^{210}Po through the gloves, the laboratory technicians often measured inside the Hypalon gloves; the gloves were exchanged in a routine process. Polonium-210 was not removed from the glovebox in Room B-102 until May 1969, when an estimated 1,400 Cu were removed by the laboratory technicians and SRS Health Physics, transferred to a concrete waste cask without spread of contamination, and shipped to the SRS burial ground (Mahathy 2010; Copley and Moyer 1969a).

4.0 PILOT PLANT AND PRODUCTION INITIATIVES

Simultaneous with the performance of the laboratory-scale program, SRS planned and prepared to build the full production plant that would separate up to 3 g/d of ^{210}Po (10 kg/yr).

In 1965, SRS published building designs and cost estimates for a new building in the 200-F Area north of Building 232-F to house polonium-processing facilities (Daking and Lanci 1965). Equipment for the processing of irradiated bismuth metal and the distilling of polonium was to be based on the design of, and testing results of, the laboratory-scale equipment. The E. I. du Pont de Nemours and Company (the Management and Operations Contractor) Atomic Energy Division (AED) requested that an alternative be considered, so in 1966 SRS published a plan for interim polonium-processing facilities in the Cold Feed Preparation Area of Building 221-F (Daking and Lanci 1966a,b). By June 1966, AED had canceled planning for a new building to house polonium processing facilities but was still directing SRS to prepare budget estimates for the modification of Building 221-F (Daking and Lanci 1966c). NIOSH has no documentation to show that modifications were done.

In 1967, SRS constructed a pilot-scale facility known as SemiWorks in the TNX building. The TNX building was used to test processes using reduced-scale models of production designs and no radioactive materials (i.e., surrogate materials were used). A distillation device based on the laboratory-scale distillation equipment, but with 8-in.-diameter stills, was built in SemiWorks (Mahathy 2010; Bradley, Landon, and Gibson 1970). All testing performed in the pilot-scale program used cadmium as a surrogate for ^{210}Po (Mahathy 2010). No work with ^{210}Po was performed in the pilot-scale program.

By 1970, AED cancelled plans for the production separation of ^{210}Po from irradiated bismuth due to NASA concerns. Rather, heat sources using ^{238}Pu were pursued. A final report of the SRS polonium distillation program was published in 1970 (Mahathy 2010).

5.0 OTHER LABORATORY WORK

The SRS analytical laboratory performed radiochemical analysis of ^{210}Po as early as 1965 in Building 773-A. A technician was involved in a bench spill of a polonium sample in May 1965. SRS Health Physics monitored the spill and performed bioassay on the technician. Additional laboratory analyses are documented in 1966 and 1967 (Gray 1967).

SRS performed an additional activity to assist the Mound Laboratory in testing ^{210}Po as a candidate for fuel capsules for use in aerospace applications. One of the design objectives was to formulate and test oxidation-resistant fuel forms of ^{210}Po. The work at SRS included design of containment facilities and equipment needed for small-scale research of perovskite, spinel, garnet, fluorite, and brookite mixed oxides to investigate systems that would reduce the decomposition pressure and rate of loss of polonium that would meet aerospace safety criteria (Hilborn 1967a,b). Mound shipped 20 mg of ^{210}Po to SRS to use for this particular fuel capsule testing; the quantity is about one-tenth the mass quantity of ^{210}Po extracted in a trial mentioned in Section 1. Equipment, including a fume hood, was used for the work, which took place in Building 773-A. Nonradioactive tellurium was used as a stand-in for ^{210}Po (Hilborn 1967b). Active research with this effort appears to have ceased by the end of 1967 because the activity is not mentioned in SRL Isotopic Power and Heat Sources quarterly progress reports issued after 1967; however, external exposure rates from ^{210}Po were still being measured in December 1968 (Copley and Moyer 1969b).

SRS Health Physics monitored Building 773-A for ^{210}Po contamination and for beta/gamma and neutron exposure rates through May 1969 when polonium was removed from the glovebox in Room B-102 and buried.

6.0 FEASIBILITY OF DOSE RECONSTRUCTION

A summary of tasks performed at SRS to plan and prepare facilities for separation of polonium and actual separation work with polonium is presented in Table 3-1.

As shown in Table 3-1, actual separation of polonium from irradiated bismuth metal was done in 1967 in a laboratory-scale effort with up to four trials and with one documented trial in 1968. That work was performed in a glovebox environment along with routine workplace monitoring (Mahathy 2010). Work was performed in a glovebox with a small quantity of ^{210}Po in 1967 to research oxidation-resistant fuel forms of ^{210}Po (Hillborn 1967a).

Personnel who entered Room B-102 were monitored for external radiation electron and photon exposures by film badge (Mahathy 2010; SRP 1967a,b,c,d). The research engineer and two laboratory technicians were sampled twice in 1967 by *in vitro* bioassay for exposure to ^{210}Po. They were first sampled in February 1967 (Author unknown 1967a); which coincides with the first bismuth irradiation event of 1967 performed a few days before (Gray 1967). Each of the three was also

Table 3-1. Summary of activities, polonium separation program.

Task	circa	Reference
Discussion of separation of Po-210 with SROO for NASA use	1963	Morris 1963
Design and construction of laboratory for laboratory-scale separation	1964	DuPont 1964
Vacuum distillation process developed using surrogate chemicals	1965–1966	Bradley, Landon, and Gibson 1970
Design of production separation facilities	1965–1966	Daking and Lanci 1965
Bismuth metal irradiated in Curium-II campaign	1966–1967	Gray 1967
Laboratory-scale separation of Po-210 for irradiated bismuth	1967–1968	Mahathy 2010
Test oxidation-resistant fuel forms for Po fuel capsules	1967	Hilborn 1967a,c
Construction of pilot plant at TNX	1967–1968	Mahathy 2010
Pilot plant separation of cadmium (surrogate) at TNX	1968–1969	Mahathy 2010
Polonium separation program terminated	1969	Mahathy 2010
Final report of polonium separation program published	1970	Bradley, Landon, and Gibson 1970

sampled in August 1967 (Author unknown 1967b). Some additional results are available for September and November 1967. Results of all polonium bioassay results were reported as less than detectable.

NIOSH has shown that three people involved in routine daily operations in the laboratory-scale separation activities in Room B-102 were sampled for intake of polonium (Mahathy 2010; Author unknown 1967a,b). NIOSH has obtained polonium bioassay results for additional SRS workers, all of whom worked in Building 773-A. NIOSH concludes that SRS workers with the potential for internal intake of polonium were monitored by bioassay. SRS Health Physics monitored locations where polonium was used (Copley and Moyer 1968b,c,d; 1969c,d,e) with no events of contamination highlighted in monthly reports. Therefore, bioassay data for a claimant can be modeled for intake to reconstruct potential doses from polonium. If a claimant does not have polonium bioassay data, NIOSH concludes that claimant was not exposed to polonium.

NIOSH has documented that staff members who entered the laboratory-scale laboratory (Room B-102) were monitored for external electron and photon exposures. As a consequence, film badge data can be used to reconstruct external exposures from polonium (Mahathy 2010).

It is uncertain to NIOSH if all laboratory-scale staff members were monitored for exposure to neutrons. Some neutrons could have been produced from concentrated polonium deposited on tantalum due to the [α,n] reaction of oxygen impurities in the tantalum liners (Bradley, Landon, and Gibson 1970). Neutron shielding was used on the laboratory-scale glovebox. Neutron dose rates taken at 3 in. from the glovebox in Room B-102 showed no measureable neutron exposure (Copley and Moyer 1968b,c,d; 1969c,d,e).

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