MASTER

GLOVEBOX HANDLING OF GRAM QUANTITIES
OF Cm^{244} AT ROCKY FLATS

by

R. A. Kirchner
K. J. Freiberg
E. A. Putzier

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THE DOW CHEMICAL COMPANY
ROCKY FLATS DIVISION
P. O. BOX 888
GOLDEN, COLORADO 80401
U.S. ATOMIC ENERGY COMMISSION
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ABSTRACT

Approximately 3 grams of Cm^{244} were processed from oxide to metal in a standard glovebox. It was demonstrated that gram amounts of Cm^{244} can be handled safely in standard gloveboxes on a "project basis." Personnel rotation and in-box shielding were used to control exposures. The average whole body exposure for 11 persons was 298 mrem. The maximum whole body exposure was 475 mrem. Based on the exposure experience of this project, "routine" work with amounts of Cm^{244} greater than 100 milligrams would be discouraged unless permanent shielding and remote handling facilities are available.

No airborne activity was noted. No alpha contamination external to the glovebox was encountered.

The inside of the glovebox and all equipment used in the project became grossly contaminated. Removal of all curium-contaminated equipment and thorough cleaning of the glovebox was necessary to reduce the external radiation to a level permissible for further routine operations.

INTRODUCTION

This report is intended to cover the health physics considerations involved with handling gram amounts of Cm^{244} in standard gloveboxes. The entire project consisted of processing slightly less than 3 grams of Cm^{244} from oxide to metal, blending the metal with kilogram quantities of plutonium to form a "master ingot," further diluting with pure plutonium, and fabricating the metal to the final product. Since health physics problems with the curium declined in proportion to its dilution, this report is devoted primarily to the project from receipt of the curium oxide until delivery to the foundry of a small metal "button" containing approximately 2 grams of curium and 15 grams of plutonium.

Since we had no other guidelines for handling gram amounts of Cm^{244} in standard gloveboxes, operational procedures were based on theoretically derived dose rates and on local experience in working with high specific activity alpha emitters. Throughout the project, radiation readings were taken whenever practical. The number of readings actually obtained was somewhat limited because the need did not justify exposing personnel to obtain a large number of readings.

Prior to the start of the project it was decided that no personnel would be permitted to accumulate a dose greater than 1000 mrem except in case of an emergency. Personnel exposure as recorded on dosimeters was used as a running index to determine the need for personnel rotation.

PREPARATION

Prior to the final decision concerning whether or not the material could be processed safely in gloveboxes, theoretical dose rates were calculated based on published decay schemes.⁽¹⁾ The curium isotopic composition of the material assumed for the project was reported by ORNL (Table I). Besides the curium content there was approximately 1.8 weight percent of Am^{243} .

TABLE I

MASS ANALYSIS OF THE CURIUM*

Cm^{242}	≤ 0.2 weight percent	} Interference due to americium
Cm^{243}	≤ 0.15 weight percent	
Cm^{244}	96.08 weight percent	
Cm^{245}	1.02 weight percent	
Cm^{246}	2.84 weight percent	
Cm^{247}	0.045 weight percent	
Cm^{248}	0.016 weight percent	

* Analysis by Oak Ridge National Laboratory.

Calculated dose rates are shown in Table II. Since the material was handled in the form of an oxide and processed to metal through a fluoride compound, it was necessary to include dose rates due to α, η reactions in our overall dose considerations.

TABLE II

CALCULATED CURIUM DOSE RATES

(All dose rates are at 1 foot from 1 gram of curium)

43-kev gamma.....	7.5 mr/hr
100-kev gamma.....	0.6 mr/hr
150-kev gamma.....	0.9 mr/hr
Fission gamma.....	8.3 mr/hr
17-kev x rays.....	3.9×10^4 mr/hr
Neutrons (spontaneous fission) ...	100 mrem/hr
Neutrons (α, η from oxide).....	1.8 mrem/hr
Neutrons (α, η from fluoride).....	150 mrem/hr

The very high dose rate due to fluorescent radiation was not considered to be serious provided that some nominal shielding was used at all times. By using lead-impregnated gloves on the glovebox and keeping the curium in metal containers when it was outside the glovebox, this requirement was fulfilled. Omitting the fluorescence contribution shown in Table II, the gamma dose rates from pure curium are relatively low.

(1) *Radiological Health Handbook*, U.S. Department of Commerce, Office of Technical Services, 1960.

Measurements by ORNL on the curium sample to be used in this project showed significant amounts of fission product contamination. Because of this contamination, tentatively identified by ORNL as rare earth fission products,⁽²⁾ the penetrating gamma dose rate was much higher than that calculated. Figure 1 shows a gamma spectrum (filtered by 1/8-inch lead) performed by ORNL on a sample of the curium to be used at Rocky Flats. The measured gamma dose rate at the surface of a small vial containing the curium (approximately 2.8 grams) was 100 R/hr. The neutron dose rates measured at ORNL were the same as the calculated values.

Since most of the gamma radiation is relatively low in energy, it seemed logical to concentrate our efforts on eliminating most of the gamma radiation with in-box shielding and use the neutron dose rates as the controlling factor for personnel exposures. Gamma shielding consisted of standard lead bricks. The bricks were drilled to accept the curium carriers and arranged within the box in a manner facilitating maximum shielding benefit. Two movable blocks of Plexiglas 2 inches in thickness were used between the curium and the operators whenever practical. A thin cadmium sheet was taped to the operator side of the blocks to act as a thermal neutron shield.

All personnel who worked with the curium were indoctrinated on the nuclear properties of the material. The importance of using the in-box shielding and minimizing physical contact was stressed. The expected dose rates at the surface of the box and personnel stay-times were discussed. All personnel were instructed to maintain as great a distance as possible between themselves and the curium at all times. This included the use of tongs for handling whenever practical.

All personnel directly involved in the curium project were given special gamma and neutron film badges for whole body and wrist dose evaluations. In addition all personnel wore self-reading gamma dosimeters to provide continuing information on accumulated dose. Two locally developed, pocket-size neutron counters were also available. Using the curium as a source, these counters were calibrated for dose by comparing with a commercial neutron dose meter incorporating a 10-inch polyethylene sphere. The neutron counters were worn by selected personnel to assist in evaluating accumulated neutron exposures.

Because of the very high alpha particle activity, special precautions were taken to ensure containment of the alpha contamination. All glove openings were fitted with heavy gage (30-mil) leaded gloves. Air locks and sample takeout ports were closed and taped. All material was inserted

into or removed from the line through heavy plastic bags. Special air sampling stations were set up and used as a continuing check for airborne contamination.

To prevent a loss of equipment due to curium contamination, all equipment not necessary for the actual processing was removed from the glovebox. It was decided that decontamination of curium-contaminated equipment would not be feasible and therefore contaminated equipment would either be discarded or packaged for use in future curium projects.

CHRONOLOGY OF OPERATIONS AND RADIATION DOSE RATES

The curium was received at Rocky Flats in two separate containers, each with just under 2 grams of the oxide. The combined curium weight was approximately 2.8 grams. Each of the curium shipments was doubly enclosed and then additionally enclosed in a large shielded shipping container. The large containers were opened outside the glovebox and the inner containers, containing the curium, removed. The inner containers were checked for alpha contamination (none was found), inserted into the glovebox, and placed behind the in-box shielding. The maximum gamma reading from the inner containers was 50 R/hr. With both containers in the glovebox and behind the shielding of Plexiglas and lead, the average dose rate at the surface of the glovebox was 5 mr/hr of gamma and 30 mrem/hr of neutron. All handling of the inner containers was with tongs.

Because of possible pressurization due to radiation heating and a decrease in atmospheric pressure, the innermost containers were cooled with argon prior to opening. During decanning with one package behind shielding and the other not shielded, the average dose rate at the surface of the glovebox was 20 mr/hr gamma and 85 mrem/hr of neutron radiation.

After unpackaging, the curium oxide was put into a crucible and placed in a small furnace for drying and fluorination. After the crucible was loaded into the furnace, but prior to fluorination, the average dose rates at the surface of the box were 60 mr/hr gamma and 95 mrem/hr neutron. After fluorination the average dose rates were 65 mr/hr gamma and 220 mrem/hr neutrons.

After cooling, the curium fluoride was mixed with approximately 15 grams of plutonium fluoride. Iodine and calcium were added and the mixture was returned to the furnace for reduction to a metal "button." After reduction the average dose rates at the surface of the box were 75 mr/hr of gamma and 90 mrem/hr of neutron radiation. After cooling the button was removed from the reduction vessel, placed in a small steel container, and removed from the glovebox

(2) Letter to E. Vejvoda, The Dow Chemical Company, Rocky Flats Division, R&D Chemistry; from R. D. Baybarz, Oak Ridge National Laboratory, Chemical Technology Division; dated April 6, 1966.

Figure 1. GAMMA SPECTRUM ON A Cm^{244} SAMPLE

through a plastic bag. The button was then transferred in a shielded container to the foundry for blending with additional amounts of plutonium metal. After the button was removed from the box, but before the slag from the reduction was removed, the average dose rates at the surface of the box were 20 mr/hr gamma and 35 mrem/hr neutron.

The duration of the curium project described above was approximately 16 working hours. Close proximity of personnel to the surface of the box was required for less than half of this time. The rest of the time included chemical reaction times and cooling and heating cycles.

Operations with the curium after this initial chemistry operation did not result in serious radiation exposure problems. It was noted that as the curium was diluted with additional plutonium the dose rates decreased in a predictable manner. Because self-absorption of neutrons within the metal mixture is negligible, the neutron dose rates were especially easy to predict. Estimation of gamma dose rates required that compensation be made for self-absorption (especially for the x rays and low-energy gamma). Even with consideration given to self-absorption, gamma dose rates accurate enough for health physics control could easily be estimated.

Special health physics coverage was continued until the final product was shipped. The results obtained during

the later operations merely confirmed the predictability of dose estimates.

RESULTS

Table III shows the personnel dosimetry results. The highest whole body penetrating exposure (as measured by film) was a combined gamma and neutron dose of 475 mrem. The highest wrist exposure was 920 mrem. All dosimetry results indicated that personnel exposures were well within an acceptable level for a one-time project. The results also showed that the average neutron exposure was almost three times the average gamma exposure. With the operational procedures and shielding arrangements used, a relatively high neutron:gamma ratio was expected.

No alpha contamination external to the glovebox was detected. Air sample results confirmed the absence of airborne contamination.

After the button was transferred, all the material in the glovebox was removed and packaged. Some of the equipment will be discarded and some will be saved for future curium work. Radiation readings external to the empty glovebox confirmed the expected curium contamination of the box. The inside of the glovebox was thoroughly cleaned and all gloves and filters were changed. After cleaning the glovebox and making the glove and filter

TABLE III

EXPOSURE RESULTS FROM Cm²⁴⁴ PROJECT

Operation		Whole Body Exposures (mrem)			Wrist Exposures (mrem)		
		Gamma Film	Gamma Dosimeter	Neutron Film	Gamma Film	Gamma Dosimeter	Neutron Film
Unpackaging	a	115	65	360	130	80	420
	b	115	60	300	120	70	440
Chemistry	a	50	70	280	130	125	390
	b	60	55	90	100	85	420
	c	120	70	300	140	90	780
	d	50	30	70	50	35	240
Health Physics	a	35	27	220	50	35	200
	b	60	50	340	100	90	480
	c	150	110	50	130	100	80
	d	70	40	260	60	40	300
Foundry	a	35	30	150	70	60	260
Average Exposures		78	55	220	98	74	365

changes, the external radiation coming from the glovebox was essentially at the background level.

SUMMARY AND RECOMMENDATIONS

The results of this project show that Cm^{244} in gram amounts can be handled safely in standard gloveboxes on a "project basis." The average external whole body exposure during the 16-hour unpackaging to reduction cycle was just under 300 mrem. While this type of exposure can be permitted on a project basis, it is considerably above the level that can be permitted for routine operations. Based on this exposure experience, routine work with amounts of Cm^{244} greater than 100 milligrams would be discouraged unless permanent shielding and remote handling facilities were provided. Since most of the exposure dose came from neutrons, purification of the curium to remove the rare earth fission products would not reduce the dose rate sufficiently to warrant exceeding the 100-milligram limitation.

The lack of alpha contamination problems during this project should not be construed to mean that the alpha activity from the Cm^{244} is not a potentially serious hazard. The

vast experience of the personnel involved in working with high alpha activity can be credited with much of the success. Another contributing factor was that the curium was in the glovebox for less than a week (from unpackaging to final cleanup). If it had been permitted to remain in the box for long periods of time it is possible that the material would have started to leak through the gloves, filters, and/or gaskets of the box. Before routine use of even milligram amounts of Cm^{244} in standard gloveboxes, the existence and/or extent of this leaking should be thoroughly investigated.

The project showed that neutron dose rates from Cm^{244} and its compounds can be accurately predicted, even when mixed with other materials. It also demonstrated that small amounts of fission product contamination can add significantly to the total gamma dose rate. However, if the gamma spectrum is known, the gamma dose rate can be predicted with a reasonable degree of accuracy.

Cm^{244} is radiologically very active. When handled in any amount, extreme care should be taken. Personnel experienced in radiological safety should be consulted before the start of any new project involving Cm^{244} .