

European Institute
for
Transuranium Elements

Interner Bericht Nr. 194

The Handling of Transplutonium Elements
at the European Institute for Transuranium Elements

K.BUIJS and W.MÜLLER

To be presented at the:
14th Meeting of the Hot Laboratories Committee
Petten (Netherlands), June 8-9th, 1972

C o n t e n t s

	Page
1. INTRODUCTION	1
2. NUCLEAR PROPERTIES OF TRANSPLUTONIUM ELEMENTS	1
2.1 Radioactive decay	1
2.2 Criticality	1
2.3 Shielding problems	2
2.4 Additional problems	2
3. EXAMPLES FOR THE HANDLING OF THE TRANSPLUTONIUM NUCLIDES	3
3.1 Purification of ^{241}Am	3
3.1.1 Properties of ^{241}Am	3
3.1.2 Aim of the operation, description of the equipment	4
3.1.3 Handling problems and observations	5
3.2 Processing of irradiated ^{241}Am	6
3.2.1 Properties of irradiated ^{241}Am targets	6
3.2.2 Aim and description of the operation	7
3.2.3 Handling problems and observations	7
3.3 Separation of ^{243}Am and ^{244}Cm on a multi- gram scale	9
3.3.1 Properties of ^{243}Am and ^{244}Cm	9

Contents continued	Page
3.3.2 Aim of the operation, description of the equipment	9
3.3.3 Handling problems and observations	11
3.4 Demonstration of a Cf-source	12
3.4.1 Properties of ^{252}Cf	12
3.4.2 Aim of the operation, description of the equipment	12
3.4.3 Problems and observations	13
4. PLANS OF A SHIELDED BOX LINE FOR THE PREPARATION, HANDLING AND INVESTIGATION OF CURIUM SAMPLES ON THE GRAM SCALE	14
4.1 Aim of the operation	14
4.2 Experimental basis of the concept	14
4.3 Plans	15
5. SUMMARY	16
6. TABLES	17-21

1. INTRODUCTION

Transplutonium elements are produced in increasing amounts; moreover, they accumulate in the waste of irradiated nuclear fuel. These elements are the objects of fundamental studies, they serve as starting material for the synthesis of higher transuranium elements; but above all there is a potential for the practical application of some of the transplutonium nuclides.

Transplutonium nuclides to be handled in quantity in research facilities or processing plants are:

^{241}Am , ^{243}Am , ^{242}Cm , ^{244}Cm and ^{252}Cf .

The problems associated with their handling depend less on the chemical or physical properties of the elements or compounds than on the nuclear properties of their atoms; more general, on the instability of their nuclei against neutron-induced fission and on their radioactivity.

2. NUCLEAR PROPERTIES OF TRANSPLUTONIUM ELEMENTS

2.1 Radioactive decay

Transplutonium nuclei decay predominantly by alpha-decay or spontaneous fission (Table 1)

2.2 Criticality

The most fissile transplutonium nuclides are hardly available in quantities attaining their minimum critical masses (Table 2). Even for

the more abundant nuclides which are less fisible, the production up to now does not yet exceed the critical masses (Table 3). From these data we can conclude that for the amounts of transplutonium nuclides available criticality may be neglected.

2.3 Shielding problems

Handling problems are determined by alpha and neutron radiation which are accompanied by gamma and X-ray emission.

The shielding requirements for transplutonium nuclides are intermediate between those for plutonium and those for irradiated nuclear fuel: the transplutonics have in general higher specific alpha and neutron activities than plutonium; the average energy of their gamma radiation is smaller than in the case of irradiated fuel.

2.4 Additional problems

Especially, the high specific alpha radioactivity causes problems due to the interaction of alpha particles with matter. Such problems are: decay heat, radiolysis of solutions, radiation damage of solids, increased chemical reactivity, formation of ozone in the presence of oxygen, erosion of container material by recoil of decaying nuclei. In addition, neutrons are generated in (α, n) reactions with light elements.

3. EXAMPLES FOR THE HANDLING OF TRANSPLUTONIUM NUCLIDES

Examples of the experience gained at the Transuranium Institute involve

- the handling of Ci and kCi amounts of ^{241}Am , ^{243}Am , ^{242}Cm and ^{244}Cm ,
- the handling of a 1 mg ^{252}Cf neutron source.

The solution of handling problems is closely related to the chemical treatment, physical form or the practical application of the nuclides. Therefore, the discussions of handling problems will be introduced by short descriptions of the related operations:

- the purification of ^{241}Am
- the processing of irradiated ^{241}Am targets
- the separation of ^{243}Am and ^{244}Cm , and
- the application of a 1 mg Cf neutron source to activation analysis.

3.1 Purification of ^{241}Am

(Low energy gamma radiation, alpha radiolysis, attack of gloves).

3.1.1 Properties of ^{241}Am

34 % of the α -decays of ^{241}Am are accompanied by the emission of a 60 KeV gamma radiation. The $1/2$ layer is 0,13 mm for Pb, 0,8 mm for Fe, 3,5 cm for H_2O .

3.1.2 Aim of the operation, description of the equipment

In order to obtain the starting material for the preparation of high purity metal and compounds, ^{241}Am has been purified at the milligram level.

5g batches of AmO_2 (~ 20 Ci) were purified in standard glove boxes shielded by 2 mm Pb equivalent lead glass. Dissolver vessel and chromatography columns were shielded by PVC-Pb-PVC sandwich layers (2 mm Pb equivalent).

At first, the purification procedure was based on ion exchange in hydrochloric acid and thiocyanate medium. Normal neoprene gloves without lead were used.

The procedure was later replaced by a combination of extraction chromatography in nitrate medium and the precipitation of $\text{K}_3\text{AmO}_2(\text{CO}_3)_2$ from K_2CO_3 solutions after oxidation of the americium with ozone (see section 3.3.2). Here, hypalon-covered gloves were used.

Dose rates were surveyed by body monitors, and radiation doses were determined with finger luminescence dosimeters and film badges.

3.1.3 Handling problems and observations

Work in chloride and thiocyanate medium was hampered by formation of highly corrosive Cl_2 and precipitation of sulphur. In addition, the neoprene gloves were attacked by oxide powder. Purification campaigns carried out in nitrate media were free from corrosion problems nor were the hypalon-covered gloves affected.

The radiation doses received by all technicians involved in typical purification runs (1-2 months) are added in the following list:

10 - 16 Rem on the hands
0,1 Rem on the bodies of the
 technicians.

As a conclusion, for the purification of 5g batches of Am on a routine basis, the protection offered by lead glass and lead foils in a standard glove box does not prevent the finger doses from reaching the tolerance level (16 Rem/3 months).

In order to purify larger quantities of ^{241}Am , we decided, therefore, to use telemanipulators and a 25cm water shield available at the Institute. By these means up to 30 g of ^{241}Am were safely and successfully purified by extraction chromatography. On the basis of this experience, telemanipulators and wa-

ter-shielded boxes are now routinely used for recycling and/or recovery of multigram quantities of ^{241}Am .

3.2 Processing of irradiated ^{241}Am

(Isolation of ^{242}Cm in a high energy and high intensity gamma field, attack of manipulator bootings, decay heat and neutron emission of ^{242}Cm)

3.2.1 Properties of irradiated ^{241}Am targets

Neutron capture by ^{241}Am leads to $^{242\text{m}}\text{Am}$ which decays to ^{242}Cm (beside decay to ^{242}Pu and fission).

Gradually, also heavier transplutonium nuclides, such as ^{243}Am and ^{244}Cm , are formed. Although being a minor constituent (by mass) of the heavy element fraction, ^{242}Cm gives rise to the greater part of the alpha activity in irradiated ^{241}Am targets. Its characteristics are:

6,11 MeV alpha radiation, $T_{1/2} = 163$ d;

specific alpha activity: 3,3 kCi/g

specific power: 120 W/g

specific neutron activity: 2×10^7 n/sec.g

Table 4 presents data characterizing 2 sets of targets ($^{241}\text{AmO}_2$ in Al cermet and cladding) irradiated in BR2 /Mol.

3.2.2. Aim and description of the operation

The aim of the processing of targets (a) and (b) was the isolation of the transuranium elements. Part of the ^{242}Cm - ^{241}Am fraction (a) was to serve as fuel for the heat source powering a thermoelectric generator.

The processing involved the removal of cladding and structure material, the removal of fission products, the separation of plutonium from the transplutonium elements.

The same processing scheme, based on ion exchange, extraction chromatography and oxalate precipitation was applied to both campaigns.

Because of the fission product radiation, dissolution of the targets and removal of the fission products had to be carried out in an \bar{A} -tight concrete shielded cell. On the basis of calculations, 3 boxes were equipped with telemanipulators and shielded with a 25cm water wall for the handling and encapsulating of the ^{242}Cm - ^{241}Am fraction of campaign a.

3.2.3 Handling problems and observations

The most serious problems arose during the processing of the targets from campaign a, when initially more than 0,5 g of ^{242}Cm (1,7 kCi) were handled:

- Radiolysis of acid chloride solutions interrupted the ion exchange separation of Pu and some fission products (Zr),

- during an interruption of the nitrogen ventilation system in the hot cell, the latex manipulator bootings were destroyed within minutes (probably by ozone formed from oxygen in the presence of alpha radiation). The latex parts had to be covered by a hypalon layer or replaced by PVC.

Oxalate precipitates of the ^{241}Am - ^{242}Cm fraction were calcined by their own α -decay heat: the oxide powder formed became red hot despite external cooling.

After removal of the fission products, the attenuation of the neutron and gamma dose rates of a mixture of 0,1 g ^{242}Cm and 1 g ^{241}Am were determined:

Thickness of water layer (cm)	5	10	15	20	25
Neutron dose rate (mrem/h) at 57 cm distance	10	3.8	1.3	0.7	0.4
Gamma dose rate (mrem/h) at 57 cm distance	100	50	35	25	20

Despite the high gamma dose rate, the 25cm water shield proved sufficient for the safe handling and encapsulating of a mixture of approximately 0,5 g of ^{242}Cm and 5 g of ^{241}Am .

3.3 Separation of ^{243}Am and ^{244}Cm on a multigram scale

(Shielding of the gamma radiation of ^{243}Am (^{239}Np); shielding of the spontaneous fission neutron radiation of ^{244}Cm ; ozone oxidation of Am, behavior of extraction chromatography and ion exchange columns at high power density of the sorbed ^{244}Cm).

3.3.1 Properties of ^{243}Am and ^{244}Cm

^{243}Am $T_{1/2} = 7,3 \times 10^3 \text{ a}$ 5,5 MeV α

$^{243}\text{Am}/^{239}\text{Np}$ (equilibrium): 3×10^{11} photons/min.g
(50% > 0,2 MeV)

1/2 layer of gamma radiation: 0,2 cm Pb

^{244}Cm $T_{1/2} = 18 \text{ a}$ 5,8 MeV α 2,9 W/g
($1,7 \times 10^{14}$ R/min.g.)

7×10^8 n/min.g. 5×10^{10} photons/min.g.
(≤ 1 MeV)

1/2 layer for neutron radiation: 5,2 cm H₂O

3.3.2 Aim of the operation, description of the equipment

An oxide mixture containing 5,9 g of ^{244}Cm and 5,1 g of ^{243}Am formed the starting material for the preparation of high purity Cm oxide. After a preliminary Am-Cm separation by precipitation of $\text{K}_3\text{AmO}_2(\text{CO}_3)_2$ (oxidation by ozone) the Cm fraction containing 2,7 % of ^{243}Am was purified further by extraction chromatography and cation exchange in nitrate medium.

CmO_2 was prepared by calcination of curium oxalate precipitated from slightly acid solution. Filtration of the oxalate had to be carried out in platinum filter crucibles, in order to avoid the introduction of impurities due to alpha erosion.

Preliminary measurements showed that the whole operation could be carried out in the (25cm) water-shielded boxes equipped with telemanipulators (M 11) mentioned before.

The shielded boxes were freely accessible at the back side. Here, blocks of paraffine were stacked inside the boxes to provide protection during short time manipulations with gloves. Also, by this arrangement, semi-quantitative analyses could be carried out by gamma and neutron dose rate measurements. Temporary storage of active materials was possible in storage pits, welded to the bottoms of the boxes and surrounded by layers of lead (3 cm) and water (25 cm). The boxes were ventilated with air.

Great care was taken to prevent the escape of ozone into the box atmosphere during ozonisation procedures: Ozone was generated outside the box and was passed to the inside in a 3 % O_3 - O_2 stream through PVC tubing and a porous stainless steel filter. Excess ozone was passed through drying towers filled with finely divided MnO_2 which catalytically

destroys ozone. No decrease of the efficiency of ozone decomposition could be detected after several days' operation.

3.3.3 Handling problems and observations

Total neutron and gamma dose rates of the starting material at a distance of 50 cm were 250 and 200 mrem/h, respectively. At the rear side of the boxes, behind paraffine blocks, radiation dose rates of 16 mrem/h (n) and 55 mrem/h (γ) were measured when the curium fraction was sorbed on the chromatography column. These values justify the concept of short time manual intervention. At the front side of the water shield, dose rates were 3 mrem/h (n) and 10 mrem/h (γ), respectively.

Corrosion of the glove box was low due to the absence of hydrochloric acid during the whole operation. During sorption and elution of the curium fraction, the extraction chromatography columns were not affected by the high power density (55 mW/ml) in the Cm band; on the other hand, the cation exchange column was disturbed by large gas bubbles above the curium containing zone which, however, did not reduce the efficiency of the purification.

3.4 Demonstration of a Cf-source

3.4.1 Properties of ^{252}Cf

$T_{1/2}$: 2,6 a 6,11 MeV 40 W/g
 3×10^{12} n/sec.g 1-2 MeV

Results of neutron and gamma dose rate measurements of 1 mg ^{252}Cf encapsulated in stainless steel are summarized in the following table (source and detector at 1,40 m from the floor):

Distance between source and detector (m)	Neutron dose rate rem/h	Gamma dose rate $\approx 0,1$ Neutron dose rate For neutrons 1/2 layer: 5cm H ₂ O Neutron dose rate at 1 m distance, 40 cm of H ₂ O): 10mRem/h
0,50	6,80	
1,00	2,40	
2,00	0,67	
3,00	0,33	
4,00	0,20	
5,00	0,14	

Therefore, shipping and storage of Cf require bulky shielding, the weight of which reaches several tons for mg-quantities of the nuclide.

3.4.2 Aim of the operation, description of the equipment

In order to demonstrate the application potential of (1 mg) Cf neutron sources, e.g. for the production of radio-nuclides or activation analysis, a series of equipment and handling devices were conceived:

- a mechanical handling device capable of gripping and releasing the (shipping) capsule, moving it vertically and along a circle of 2,5 m radius;
- a series of containers placed along a semicircle within reach of the handling device, including
 - a MnSO_4 bath for calibration of the neutron sources,
 - a $4,5 \text{ m}^3$ water filled basin accomodating various irradiation set-ups with different geometries,
 - a storage container (same as shipping container)
 - a 50 cm i.d. container shielded by an annular 50 cm water layer for experiments with high energy neutrons and for dose rate attenuation measurements.

Free floor space is available for various experiments involving free air irradiations. These experiments included irradiations in a simulated neutron shielded glove-box, as well as irradiations of body phantoms aimed at development of finger and body neutron dosimeters.

3.4.3 Problems and observations

When handling the 1 mg source with the device at a constant distance of 5 m, the operator is exposed to a (direct) neutron dose rate of 150 mRem/h. Neutron scattering from walls.

floor or ceiling may considerably increase the neutron dose rate in comparison to the value estimated from shielding calculations. Estimation of a fast neutron dose rate from a measured gamma dose rate using a fixed ratio between these two, may be considerably in error because of preferential attenuation of either radiation and generation of capture gammas.

4. PLANS OF A SHIELDED BOX LINE FOR THE PREPARATION, HANDLING AND INVESTIGATION OF CURIUM SAMPLES ON THE GRAM SCALE

(Neutron shield, double glove box system)

4.1 Aim of the operation

According to our Institute's research program we have the intention

- to prepare Cm metal, alloys and compounds of high purity;
- to prepare samples for the determination of physical and physicochemical properties by compacting, sintering and polishing of pellets or by evaporation;
- to handle and store such neutron emitting material in inert, dry atmospheres.

4.2 Experimental basis of the concept

The general outline of the shielded box system is based on experience with

- the purification of Cm and the preparation of the oxide in water shielded telemanipulator boxes (3.3), and
- the preparation of americium metal in gram amounts via the vapour phase in a double box system and the handling and storage of the metal in argon: an inner stainless steel box is filled with circulating argon; the vacuum equipment and the induction furnace are contained in an outer nitrogen-filled glove box; since the nitrogen pressure is lower than those of the argon and laboratory atmospheres, diffusion of water vapour through the ozone resistant gloves is reduced, and water and oxygen content in the argon can be kept below 5 and 1 ppm, respectively.

4.3 Plans

The shielded box line suitable for the program outlined before (4.1) will consist of 4 double boxes (grinding and pressing; microbalance; induction and resistance furnaces with vacuum systems) and 3 single boxes (analytical balance; polishing; vacuum deposition). The double boxes will be of 2 types: In the first type (e.g. microbalance) the inner box is in a fixed position with 12 cm space between the front panels to allow renewal of the gloves. This space may be put to use by the installation of a removable perspex plate for neutron shielding. The inner box of the second type (e.g. induction furnace) can be rolled back when gloves are renewed.

Up to 20 cm of neutron shielding may be rather cumbersome for work in glove boxes. It is considered safest, however, as a rule to have the neutron shielding mounted in its position, and to remove it only in special cases.

5. SUMMARY

Gram amounts of Am and ^{244}Cm are handled in (single or double) standard boxes; neutron and low energy gamma attenuation is provided by plexiglass and lead glass.

For multigram amounts of americium and curium, water shielded boxes with telemanipulators are used; manual intervention is possible from the rear through ozone resistant gloves.

Hot cells are required only in special cases involving e.g. fission product activities or mg amounts of californium.

6. TABLES

- Table 1: Decay properties of transplutonium nuclides
- Table 2: Minimum critical masses of fissile transplutonium nuclides in aqueous solution and corresponding concentrations
- Table 3: Calculated critical radii and masses of transplutonium metals in spherical geometry
- Table 4: Thermal power and gamma dose rates of irradiated ^{241}Am targets

T A B L E 1

Decay properties of transplutonium nuclides

Nuclide	Half-life	Decay			Watt/g.	Neutrons/sec.g
		α (%)	β (%)	SF (%)		
<u>241_{Am}</u>	433a	100		1.5×10^{-10}	0.11	0.7
242 _{Am}	16h		$84\beta^-; 16\epsilon$		2000	
242 ^m _{Am}	144a	0.43	I.T.	1.6×10^{-8}	3×10^{-2}	2×10^2
<u>243_{Am}</u>	7340a	100		2.3×10^{-8}	6.5×10^{-3}	6.5
<u>242_{Cm}</u>	163d	100		6×10^{-6}	122	2×10^7
243 _{Cm}	32a	99.7	0.3 ϵ		1.68	
<u>244_{Cm}</u>	18.1a	100		1.3×10^{-4}	2.83	1.2×10^7
245 _{Cm}	8532a	100			5.7×10^{-3}	
246 _{Cm}	4820a	100		0.03	9.8×10^{-3}	9.1×10^6
247 _{Cm}	1.6×10^7 a	100			2.8×10^{-6}	
248 _{Cm}	3.6×10^5 a	89		11	5.2×10^{-4}	4.0×10^7
249 _{Bk}	314d	0.0017	100 β^-		0.36	
249 _{Cf}	352a	100		6×10^{-7}	0.15	3×10^3
250 _{Cf}	13.1a	100		0.08	4.1	3.5×10^9
251 _{Cf}	900a	100			5.8×10^{-2}	
<u>252_{Cf}</u>	2.646a	96.8		3.2	39	2.4×10^{12}
254 _{Cf}	60.5d	0.2		99.8	10600	1.2×10^{15}
253 _{Es}	20.5d	100		8.6×10^{-6}	1010	3.3×10^8
257 _{Fm}	94d	99.8		0.21	~ 200	~ 1.5×10^{12}

The most frequently handled nuclides are underlined.

I.T: isomeric transition

ϵ : electron capture

T A B L E 2

Minimum critical masses of fissile transplutonium nuclides
in aqueous solution and corresponding concentrations

(Value for ^{239}Pu included for comparison)

Nuclide	Critical Mass [g]	Concentration [g/l]
^{239}Pu	430	30
$^{242\text{m}}\text{Am}$	23	5
^{243}Cm	213	40
^{245}Cm	42	15
^{247}Cm	159	60
^{249}Cf	32	20
^{251}Cf	10	6