Design of and Equipment for Hot Laboratories

Proceedings of a Symposium

Otaniemi

2-6 August 1976

PROCEEDINGS SERIES

DESIGN OF AND EQUIPMENT FOR HOT LABORATORIES

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FOREWORD

The rapid expansion of the nuclear power industry and the widespread application of radioisotopes in scientific and technological research have increased the need for specialized laboratories, called hot laboratories, for handling radioactive substances of high activity as well as plutonium and transplutonium elements.

The handling of plutonium and transplutonium elements presents special radiation protection problems because of their high specific activity and high radiotoxicity. The considerations of safety in hot laboratories are not limited to radiological problems alone. Solutions for conventional safety problems such as chemical explosions, fire, etc., are very important. Therefore, the planning, design and construction of hot laboratories must meet the stringent requirements of containment, shielding, ventilation, fire protection, criticality control, waste management and safety in operations including transfer and transport of radioactive substances. The primary objectives of such strict requirements are to protect the workers, the public and the environment from any harmful effects.

The International Atomic Energy Agency, recognizing the significance of radiation protection problems in hot laboratories, has been active in this field for more than a decade and has already published two IAEA Safety Series manuals – one, No.30, on the Safety Aspects of Design and Equipment of Hot Laboratories and the other, No.39, on the Safe Handling of Plutonium. In addition, the Agency organized in 1969, at Saclay, a symposium on Radiation Safety in Hot Facilities.

The symposium from 2–6 August 1976 was organized in Otaniemi at the invitation of the Government of Finland. The purpose of the symposium was to review the present state of knowledge and collect information on the recent developments on various aspects of safety in hot laboratories. These developments had been stimulated by the latest concepts as set out in ICRP Report No.22 (published 1973), which recommends that not only should the individual dose limits not be exceeded, but also the collective doses from a given practice have to be kept as low as reasonably achievable, taking into consideration the economic and social factors.

The symposium was attended by 140 participants from 32 countries and three international organizations. There were 45 papers presented during eight technical sessions. The topics covered were: the safety features of the planning and design of hot laboratories, air cleaning, transfer and transport systems, criticality control, fire protection, waste management, radiological protection, administrative arrangements and operational experience. The major emphasis in the symposium was placed on the safety features of planning and design.

The book contains all the papers presented at the symposium plus the discussions in full and a concluding summary presented by Mr. G. Lefort.

The Agency gratefully acknowledges the co-operation of the Government of Finland, and particularly the Local Organizing Committee, in the organization of the symposium.

EDITORIAL NOTE

The papers and discussions have been edited by the editorial staff of the International Atomic Energy Agency to the extent considered necessary for the reader's assistance. The views expressed and the general style adopted remain, however, the responsibility of the named authors or participants. In addition, the views are not necessarily those of the governments of the nominating Member States or of the nominating organizations.

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SAFE HANDLING OF KILOGRAM AMOUNTS OF FUEL-GRADE PLUTONIUM AND OF GRAM AMOUNTS OF PLUTONIUM-238, AMERICIUM-241 AND CURIUM-244

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Abstract

SAFE HANDLING OF KILOGRAM AMOUNTS OF FUEL-GRADE PLUTONIUM AND OF GRAM AMOUNTS OF PLUTONIUM-238, AMERICIUM-241 AND CURIUM-244.

During the past 10 years about 600 glove-boxes have been installed at the Institute for Transuranium Elements at Karlsruhe. About 80% of these glove-boxes have been designed and equipped for handling 100-g to 1-kg amounts of ^{239}Pu containing 8–12% ^{240}Pu (low-exposure plutonium). A small proportion of the glove-boxes is equipped with additional shielding in the form of lead sheet or lead glass for work with recycled plutonium. In these glove-boxes gram-amounts of ^{241}Am have also been handled for preparation of Al-Am targets using tongs and additional shielding inside the glove-boxes themselves. Water- and lead-shielded glove-boxes equipped with telemanipulators have been installed for routine work with gram-amounts of ^{241}Am, ^{243}Am and ^{244}Cm. A prediction of the expected radiation dose for the personnel is difficult and only valid for a preparation procedure with well-defined preparation steps, owing to the fact that gamma dose-rates depend strongly upon proximity and source seize. Gamma radiation dose measurements during non-routine work for ^{241}Am target preparation showed that handling of gram amounts leads to a rather high irradiation dose for the personnel, despite lead or steel glove-box shielding and shielding within the glove-boxes. A direct glove-hand to americium contact must be avoided. For all glove-handling of materials with gamma radiation an irradiation control of the forearms of the personnel by, for example, thermoluminescence dosimeters is necessary. Routine handling of americium and curium should be executed with master-slave equipment behind neutron and gamma shielding.

1. ALPHA-CONTAINMENT

The transuranium elements, especially plutonium, are known to be highly radiotoxic, owing to the damaging effects of alpha-emitting nuclides when incorporated into the human body. The greatest risk originates from inhalation. Protection of people working with transuranium nuclides must be based on adequate control and containment of these materials. Although plutonium generally can be handled at short distances in glove-boxes without additional shielding, certain isotopes such as plutonium-238, 240, 241, 242 and the transplutonium elements Am, Cm, Cf require shielding against gamma and/or neutron radiation (see Table I).

The efficient protection of personnel and of the general public against release of and radiation from transuranium nuclides are ensured in the Transuranium Institute at Karlsruhe by suitable installations and equipment; appropriate working rules and handling techniques; and the radiological protection service.

Installation and equipment

The buildings of the Institute have been constructed such that the exchange of air between outside and inside occurs at only a few places and in predetermined quantities. A system of stepwise increasing underpressure forces air entering the buildings to flow in the direction of greater contamination risks, i.e. to the hot laboratories. The air leaving the buildings passes through a series of absolute filters and is continuously monitored. In the past five years the amount of

TABLE I. RADIATION CHARACTERISTICS AND SHIELDING REQUIREMENTS FOR MOST COMMON TRANSURANIUM NUCLIDES [1]

Group [a]	Nuclide	Dose-rate at 50 cm from 1 g of nuclide (mrem/h)	Main gamma energy (MeV)	Half-value layer
I	^{238}Pu	80 (γ)	0.017	0.6 cm H_2O or Plexiglass 0.015 mm Pb
	^{239}Pu	0.07 (γ)	0.014 0.017	
	^{240}Pu	0.8 (γ)	0.017	
	^{244}Cm	240 (γ)	0.018	
II	^{241}Pu [b]	10 (γ)	0.060	0.13 mm Pb
	^{241}Am	150 (γ)	0.060	
III	^{237}Np [c]	0.2 (γ)	0.31	1.5 mm Pb
	^{238}Pu	0.1 (n:α, n)		5 cm H_2O
	^{240}Pu	3.5 (n:SF)		5 cm H_2O
	^{241}Pu [b]	250 (γ)	0.21	0.6 mm Pb
	^{241}Am	0.08 (γ)	0.3–0.4	2 mm Pb
	^{243}Am	40 (γ)	0.28	1.2 mm Pb
	^{242}Cm	13 (γ) 120 (n:SF+α, n)	0.58	5 mm Pb [d] 5 cm H_2O
	^{244}Cm	3 (γ) 35 (n:SF)	0.5–3.0	8 mm Pb [d] 5 cm H_2O
	^{252}Cf	6×10^5 (γ) 8×10^6 (n:SF)	0.5–3.0	8 mm Pb [d] 5 cm H_2O

[a] Radiation types are divided into 3 groups based on shielding requirements:
Group I: Very light shielding. Main source of irradiation: contaminated surfaces.
Group II: Light shielding of glove-box and equipment required.
Group III: Medium to heavy shielding required.
[b] In equilibrium with 6.7 d ^{237}U (daughters from 0.002% α-decay).
[c] In equilibrium with 27 d ^{233}Pa.
[d] Shielding required to reduce the dose-rate due to capture gamma radiation is not considered.

^{239}Pu released into the atmosphere did not reach 0.3% of the amount permitted by the local authorities. Within the buildings four zones with respect to possible irradiation and contamination of personnel are established. Zone 1 (offices, clean side of change rooms, main entrance) is always a clean area. Zone 2 (corridors for personnel, control rooms, work shops, offices) is a normally clean area, which is surveyed with respect to alpha and gamma radiation. Zone 3 (laboratory rooms, storage areas and transport corridors for radioactive material) is an alpha-beta-gamma radiation control zone, where contamination may be encountered. Zone 4 (glove-boxes, hot cells, decontamination areas) is a contaminated area. Zones 1, 2 and 3 are separated by six locks and transition zones. Direct access to zones 2, 3 and 4 is normally not possible. The unirradiated transuranium elements are handled in the hot laboratories in glove-boxes, which are at constant underpressure with respect to their surroundings. The glove-boxes have their own separate ventilation system (Fig. 1). Each glove-box is equipped with 2 inlet and 2 outlet absolute filters and a pneumatic

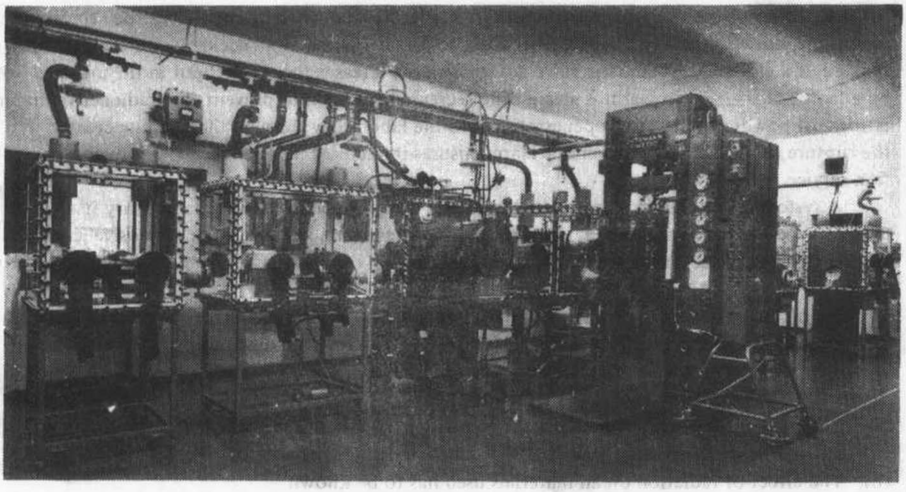

FIG.1. Laboratory for (U, Pu)O_2 fuel preparation.

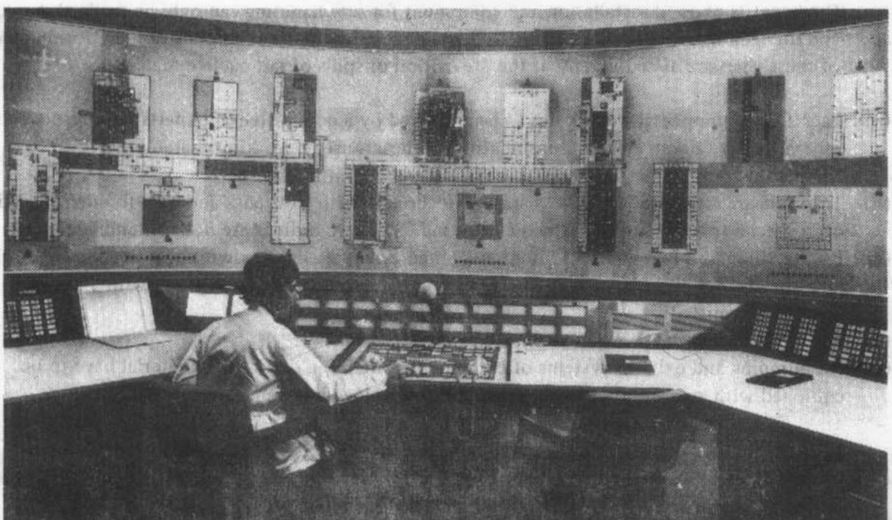

FIG.2. Control room.

valve which keeps the underpressure constant. Under normal working conditions the atmosphere within the glove-boxes is exchanged 5–8 times an hour. In case of an accident, for example the rupture of a glove, the pneumatic valve automatically increases the throughput in the affected box by a factor of six, and an alarm is given. The increased throughput prevents the radioactive material in form of powder or dust from penetrating into the laboratory through the opening created by the rupture of a glove. The automatic alarm ensures immediate help from the radiological protection service.

A system of automatic alarm, installed within the glove-boxes and the laboratory itself, monitors under- and overpressure, temperature of atmosphere, water leakage, explosive gases such as hydrogen, cooling-water temperature and flow and the purity of protective atmospheres used in special cases.

The alarm system connects the various working places with the control room (see Fig.2), which is manned round the clock. The glove-boxes themselves and all equipment used in these glove-boxes have been carefully selected and checked for all kinds of safety risks.

(a) The fully equipped glove-boxes are tested with a 10-mm water gauge for both over- and underpressure (permitted leak-rate < 5-mm water gauge pressure/m^3 h)
(b) The effect of radiation on all materials used has to be known
(c) Heating systems, for example furnaces etc., have to be shut off automatically if a failure in the cooling system occurs
(d) The water-cooling circuit for all equipment used in handling radioactive materials is separated by a heat exchanger from the normal outside water supply.

Working rules and handling techniques

The benefits of good installation and equipment for safe handling can only be realized if rules and handling techniques specific to each radionuclide and its chemical composition are formulated. Most of these rules are aimed mainly at the prevention of spills of radioactive material.

(a) Each fully equipped glove-box has to be approved by a committee of experts with respect to general safety (alarm, electrical installation, materials used), working characteristics of equipment and manner of operation before it can be put into service;
(b) Transfers are only allowed by a "bagging" technique with PVC bags and high-frequency sealing or in special transfer systems which ensure that the surroundings are not contaminated;
(c) For handling uranium and plutonium, neoprene gloves are used, whereas for nuclides with high specific activity (Ac, ^{238}Pu, Am, Cm, Cf) only hypalon gloves or gloves with a hypalon layer are permitted;
(d) All gloves have to be changed at predetermined intervals;
(e) All pumping and exhaust systems of equipment containing radioactive material have to be equipped with an absolute filter, connected to the glove-box ventilation system.

In addition, by frequent checking of hands, feet, clothing and equipment for contamination each worker contributes to the early detection of uncontained radioactivity.

Radiological protection service

The radiological protection service, which is fully independent of the technical and scientific groups:

(a) Assists in the planning of experiments by advising on safety matters;
(b) Supervises the execution of agreed safety measures;

(c) Provides and maintains safety equipment and instrumentation for checking for contamination and radiation;
(d) Carries out a great variety of contamination checks in the laboratories and on equipment;
(e) Monitors radiation doses;
(f) Is able to organize and supply relief in emergency situations.

Continuous air monitors rapidly detect quantities of radioactivity which exceed the maximum permissible values. Much more sensitive, but with a delay of eight days, a discontinuous measurement for ^{239}Pu has a limit of detection of 6×10^{-11} mg/m^3.

The air monitoring is complemented by routine contamination checks of clothing, shoes, floors, furniture, equipment, filters and gloves.

2. SPECIAL ARRANGEMENTS

2.1. Precautions against criticality

To avoid criticality accidents special arrangements are necessary for handling fissile materials such as ^{239}Pu and ^{235}U. The working areas for these materials are divided in two types of working zones with limited amounts of fissile material. One for "wet" work (mainly chemical laboratories) with a maximum of 225 g fissile material and one for "dry" work with a maximum of 2.5 kg.

Certain equipment such as water-cooled furnaces, centreless grinding machines and hydraulic presses are only permitted in areas where up to 2.5 kg fissile material can be handled, if it can be ensured that hydrogen-containing liquids, such as water and oil, cannot create a criticality accident. At the Transuranium Institute this is achieved by limiting the quantity of the hydrogen-containing liquids which, in case of an accident (leak, rupture of a tube etc.), will be in contact with fissile material, to the amount that ensures that a safe geometry is maintained (columns of 3 in. diameter or volumes with a height of 1 in.). In some laboratories criticality alarms are installed.

2.2. Pyrophoric materials

Oxide material is handled in glove-boxes under air, whereas pyrophoric materials such as metals, carbides and nitrides are handled in glove-boxes with a protective atmosphere (closed system with purified argon or nitrogen). The transport and storage of these materials is only permitted in metal containers.

3. SHIELDING PROBLEMS

Alpha and neutron radiation, which are accompanied by gamma and X-ray emission, cause handling problems of certain plutonium and transplutonium elements.

3.1. Handling of plutonium

The isotopic composition of plutonium determines the degree of shielding necessary. As the isotopic composition of plutonium from spent reactor fuel depends both on the burnup attained and the type of reactor no generally valid data for the necessary shielding can be given. From our experience during fuel preparation [3] with kilogram amounts of low-exposure plutonium (wt% $^{238}Pu = 0.05$; $^{239}Pu = 88.8$; $^{240}Pu = 10.1$; $^{241}Pu = 0.94$; $^{242}Pu = 0.11$) and high-exposure plutonium (wt % $^{238}Pu = 0.11$; $^{239}Pu = 68.5$; $^{240}Pu = 25.88$; $^{241}Pu = 4.11$; $^{243}Pu = 1.39$) and gram-amounts of ^{238}Pu the following conclusions can be drawn:

(a) Low-exposure plutonium can be handled in normal glove-boxes without additional shielding. The gamma dose-rate of the high-exposure Pu can be reduced by lead glass (0.25 cm Pb equivalent) and lead gloves to about the same level as for low-exposure Pu;

(b) The gamma dose-rates depend strongly upon proximity and source size. The electromagnetic radiation is primarily a surface phenomenon because the relatively low-energy photons from most Pu isotopes are efficiently shielded by the plutonium and in U-Pu blends, by the heavy metals themselves. Table II gives the dose-rate measurements of 320 g of Pu high-exposure metal ingot (ϕ = 5 cm × 1 cm) and 140 g of the oxide from the same material measured during sieving (ϕ = 12 cm × 1 cm);

(c) The gamma dose-rate decreases strongly with uranium dilution owing to self-shielding;

(d) For high-exposure Pu direct gloved hand-to-fuel contact should be avoided as far as possible;

(e) The neutron yield, mainly produced by even-numbered Pu isotopes is essentially unaffected by self-shielding of heavy metals and, in the case of U-Pu blends, is directly proportional to the Pu-content;

(f) Predictions of expected dose-rates for the personnel are difficult and depend strongly on the fabrication steps. A strict irradiation control, for example especially of the forearms, by thermoluminescence dose meters is necessary. Radiation measurement during well-defined single fabrication steps is the best possibility of detecting and finally decreasing irradiation doses for the personnel.

Similar findings have been published recently by Smith and co-workers [2].

TABLE II. GAMMA AND NEUTRON DOSE MEASUREMENTS ON LOW- (10 wt.% ^{240}Pu, 0.9 wt.% ^{241}Pu) AND HIGH- (25 wt% ^{240}Pu, 4.1 wt% ^{241}Pu) EXPOSURE PLUTONIUM

Material	Dose-rate mrem/h distance	10 cm	30 cm	Manipulation	Finger dose (TLD) mrem/h
High-exposure Pu					
metal (318 g)	γ	38	5	Preparation	145
ϕ 5 × 1 cm	n	6	1	of turnings	(50)[a]
PuO_2 (140 g)	γ	1280	250	Sieving	530
ϕ 12 × 1 cm	n	2	0.4		(190)[a]
PuO_2 (50 g)	γ	256	52	Sieving	428
ϕ 10 cm	n	1	0.1		(260)[a]
$(U_{0.8}Pu_{0.2})O_2$	γ	120	25	Sieving	171
blend (50 g PuO_2)	n	0.9	-		(107)[a]
Low-exposure Pu					
PuO_2 (50 g)	γ	19	2.6	Sieving	53
ϕ 10 cm	n	0.3	-		(30)[a]
$(U_{0.8}Pu_{0.2})O_2$	γ	6	1	Sieving	21
blend (50 g PuO_2)	n	0.3	-		(14)[a]

[a] With 0.1 mm Pb equivalent protection.