RADIOLOGICAL SAFETY EXPERIENCE IN HANDLING AND FABRICATION OF PLUTONIUM FUEL ELEMENTS

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Abstract

The problems and experience gained in radiological safety during PuO_2 fuel fabrication for 'PURNIMA' reactor are discussed in this paper. Safety aspects in design, construction and commissioning of metallurgical facilities and glove boxes, measures for contamination control, air, area and criticality monitoring instrumentation and their calibration are dealt with. Radiological health data are summarised to reflect the adequacy of precautionary provisions followed in fuel fabrication work. Evolution of the methods for related safety problems like assessing neutron emission from PuO_2 fuel pins and Pu in solid wastes are outlined.

Introduction

Radiometallurgy Laboratory at the Bhabha Atomic Research Centre has been operating a plutonium fuel fabrication facility during the last two years involving handling and storage of kilogram amounts of plutonium. The facility has fabricated the complete core charge of PuO_2 pins for the zero power fast reactor 'PURNIMA' at Trombay.

The fuel pin of 'PURNIMA' has stainless steel cladding and consists of a central PuO_2 core of 180 mm length followed at either end, by a molybdenum plug of 80 mm length and stainless steel end plug to serve as axial reflectors in the reactor. The pin is of 11 mm diameter and of 495 mm overall length and has a plutonium loading of 123.5 g. The complete core loading called for production of 178 full fuel pins, 4 half and 4 quarter fuel pins. In addition, seven Pu-Be start-up source pins, identical in shape to the PuO_2 fuel pins, were also fabricated. This paper deals with the safety aspects in the design of the handling facilities and the experience gained in the radiological safety during the fabrication compaign.

Fuel Pin Fabrication

The feed powder PuO_2, after milling, granulation, etc. is loaded in a suitable die and pressed with a 10 ton hydraulic press, located in a glove box. The green pellets, thus obtained, were sintered in an argon and 8% hydrogen atmosphere in a molybdenum furnace inside a glove box. After sintering the length and the weights of the pellets were checked and the geometrical density compared with that obtained by an immersion method using dibromo ethane. Acceptable pellets were then inserted in the stainless steel clad tube, whose top end plug was already welded and radiographed. The tube was inserted into the glove box; a holding spring was pushed in, followed by a molybdenum plug, PuO_2 pellets and another molybdenum reflector. The loaded clad tube was held in a special welding chamber in a glove box and the chamber was evacuated and filled with helium. After inserting the lower end plug, welding was carried out remotely by argon arc welding. The fuel pins were then decontaminated, subjected to radiography and helium leak test. Approved fuel pins were loaded in birdcages and transferred to the Plutonium Store or reactor site.

Design Safety Features

The long biological half-life and the high energy of the emitted alpha particles together with selective localisation in bone or lung makes plutonium one of the most toxic materials when deposited inside the body. With the maximum permissible lung burden for insoluble plutonium (e.g. PuO_2) being as low as 0.016 μCi ($\approx$ 0.26 μg) the glove box design called for stringent built-in safety features with regard to containment capability. Since a mass as low as 500 g of Pu could lead to a nuclear excursion under unfavourable conditions, criticality safety was to be considered through mass control, safe separation of Pu units and safe design of birdcages for storage and transport, with administrative control at each stage of handling.

Safety Features in Laboratory Design

The fuel fabrication facility is housed mainly in two high active area halls with entry from an active corridor. The corridor has a decontamination room at one end, and the other side, opens out to a personnel corridor, leading to a change room. A plutonium store room is located in the personnel corridor. Equipments for the various metallurgical operations are housed in glove boxes, located in the high active halls. Services of a high order of integrity, required for a class A laboratory have been provided. Laboratory area and glove boxes are provided with separate air supply and exhaust system, the equipments of which, located in a filter house, discharge the effluents through a 76 m stack, after filtration through high efficiency particulate filters. Ventilation for the active halls and corridors have been designed to give respectively 10 and 7 air changes per hour and pressure differentials have been maintained between the areas to enable air-flow from low active areas to high active ones.

Glove Boxes and their Safety Features

Many design safety features have been incorporated in the fabrication of glove boxes from the point of view of containment. Glove box frame and floor are made of s.s 304 for ease of decontamination. Filter boxes of aluminium are conveniently located so as to enable replacement of the inlet and outlet filters by a single hand operation through the upper port. Absolute filters of MSA Honey-comb type with an efficiency of 99.9% for 0.3 μm particles are used. Transfer and posting-in operations are carried out through air locks with double doors and bagging-in ports. Normal atmosphere was found adequate for PuO_2 fuel work; however, for Pu metal handling, the boxes could be turned to an argon system, provided with a purifier and a recirculation unit. Operations were carried out in glove boxes under -1 in.WG.

Air enters through an isolation valve, a ball valve, rotameter, a regulator and an inlet filter; and vents via an exit filter, a ball valve and a three-way solenoid valve. A mechanical pressure controller and a bellow adjust minor pressure variations but in case of accidental overpressurisation or any rupture of glove or failure of recirculation system, the three-way solenoid valve initiates emergency control by opening the box directly to the -10 in. WG. main glove-box exhaust line. The inlet regulator closes and a pressure differential switch flashes an alarm in the form of a red light on glove-box board, warning the operating staff of an emergency situation. Neoprene gloves, 0.8 mm thick, were considered adequate against the soft radiation emitted by plutonium. Apart from installed CO_2 extinguishers in the halls, eutectic salt mixture in sealed PVC bag was kept handy in glove-boxes to smother any fire. The furnace coolant water is normally on main water supply. An emergency water tank has been provided to take care of failure of main water supply or loss of pressure. Filtered water from a pool was also connected to the line as an alternative for sustained supply.

Pre-commissioning Tests

The primary responsibility of the health physics staff at the time of commissioning of the facility was to check the adequacy of the protective features and assess operational safety.

Glove Box Containment Evaluation. The gloves and glove boxes were checked for leakage before commissioning for Pu handling. The box leakage rates were found to be less than 0.05% box volume/hour, as is prescribed for inert atmosphere boxes.

Glove Box Filter Efficiency Tests. Filter efficiency checks were carried out with uranine aerosols. Filters were approved for use only when they conformed to 99.9% efficiency for 0.3 μm particles.

Effluent Drains Checks. High and low active drains were checked with inactive cold runs using rhodamin dye to ensure proper pump connections and valve operations.

Breathing Air Line Checks. Compressor air was checked for presence of oil mist, moisture and CO to ensure that their levels were below the tolerance limits. The minimum requirement of 3 cft/m at the breathing points was checked.

Operational Safety and Hazards Control

Mass of Pu, handled was initially limited to 75 g per batch to acquire experience and later, the batch size was progressively increased to 500 g Pu, after reviewing the safety aspects. In all about 85 sintering runs and about 200 in-box welding operations, covering fuel pins and start-up source pins were carried out.

Constant health physics surveillance was provided for the operations. Access control to the fuel laboratory was enforced through change rooms. In potentially active areas like filter room, decontamination room, entry was effected under health physics supervision or after obtaining special work permits. Use of protective clothing consisting of overalls, overshoes, head caps and surgical gloves for handling pellets and pins was recommended. Further, TLD's on forehead and chest, normal beta-gamma and fast neutron film badges, criticality badge and pocket dosimeters were worn while at work with PuO_2. Air line respirators connected to 15 lbf/in^2 airline via quick connection couplings were kept readily available for emergency use.

Equipments for sintering and weighing operations were located in a train of interconnected glove boxes to preclude the necessity of intermittent bagging out operations and consequent external exposure. Different phases of work were segregated to avoid contamination spread. The welding operations, metallography work and source pin fabrication were grouped separately in another train of glove boxes to facilitate flexibility and control of radiation exposure and contamination.

As the quantity of PuO_2 handled was progressively increased, extensive radiation survey was conducted to control personnel exposure using conventional radiation monitoring instruments. The area and air monitors were strategically located in the laboratory. Provision was also made to monitor the effluent streams. The monitors along with a remote read-out on a Central Health Console give alarm at pre-set limits for initiating corrective action.

External Hazards and Control

PuO_2 powder was obtained from reprocessing nat.U fuel from Cirus reactor.

Radiation survey data of the first seven sintering runs, with 75 g Pu per batch, indicated high beta-gamma dose rates from pellets; the pellets showed a gamma dose rate of 300-450 mR/h and the beta dose rate was 1-2 R/h. The glove box panels registered a gamma dose rate of 50-75 mR/h. This also indicated a ratio of about 6 between contact and chest level dose rates. Analysis of an aliquot sample of PuO_2 by health physics staff indicated mainly $^{95}Zr-^{95}Nb$ activity and a total activity of about 5 $\mu Ci/g$ of PuO_2. Subsequently PuO_2 was therefore obtained from spent fuel rods with lesser fission product content. As a result, the gamma dose rates on the sintering glove box panel came down to 12-30 mR/h even with increased quantities of 200-550 g PuO_2.

The fabrication of fuel pins as well as start-up source pins did not call for special shielding to boxes. As a measure of radiation safety, fuel pin welding and Pu-Be source pin fabrication jobs were carried out by rotation of staff. This was necessary as the beta-gamma dose rate from a full fuel pin at 1 cm was nearly 40-50 mR/h while the neutron dose rate was 130-140 mrem/h. Fuel pins were checked individually for loose contamination by an alpha probe inside the box and also with swipe counting. After radiography and helium leak test, the weld-zones were checked for fixed contamination. Before machining of the welds for most pins, the counts varied in the range of 100-1000 dpm/cm^2, maximum being 64000 dpm/cm^2 while after machining and polishing, the levels for most pins came down to 200-400 dpm/cm^2.

For start-up source pins of 0.9 Ci strength, the method of fabrication was to mix nearly 16 g of Pu as PuO_2 with nearly equal quantity of Be followed by pressing and sintering in a high vacuum induction furnace. The neutron exposures incurred during fabrication of 2 source pins were of the order of 50 mrem on chest and 400 mrem on wrist per man. The contact gamma dose rate of the source pin was nearly 150 mR/h while the dose rates at 30 cm from the pin were 16 mR/h due to gammas and 25 mrem/h due to neutrons.

Cumulative dose(beta-gamma-neutron) received by a few members of the staff, directly involved in the fabrication work during the compaign period 18.6.70 to 21.3.72 are indicated below:

Persons	Exposure(mrem)	Person	Exposure(mrem)	Person	Exposure(mrem)
A	177	F	658	L	763
B	482	G	867	M	820
C	199	H	541	N	379
D	827	I	1069	O	473
E	166	J	1540	P	569

From the estimated ratio of contact to chest level dose rates, maximum extremity exposure could be of the order of 9 R.

Air Contamination Control

Each glove box premise has a suction port with an air sampling head connected to a central air sampling pump. Filter paper samples obtained with this system as well as with annular impactors when analysed for long lived activity, did not show any air contamination in the laboratory. In addition a Pu-in-air monitor located in the laboratory detects air borne Pu by alpha spectrometry. The detector is of a silicon surface barrier type. The unit is pre-set to sound an alarm at 8 MPC hours in presence of natural radioactivity whose spill-over in 4.1 - 5.1 MeV plutonium channels is estimated to be less than 10% of the total.

During glove changing operations, respirator area was maintained. Maintenance work was carried out once on an induction furnace for which frog suit and air line respirator were prescribed. Only one instance of air contamination due to a small tear on glove arose. Due to immediate corrective action, no personnel exposure occurred.

Bioassay and whole body counting of the operating staff showed that there was no internal exposure. About 34 members of staff were monitored, after completion of the programme for Pu deposition in lung with a thin NaI(Tl) crystal with a Be window. The count rates obtained were of background levels after repeat monitoring.

Environmental Contamination Control

Air-borne effluents were discharged after monitoring downstream through a stack. The glove box and laboratory exhaust had negligibly small long lived activity. The liquid wastes, both high active ($>10^{-4}$ $\mu Ci/cm^3$) and low active ($< 10^{-4}$ $\mu Ci/cm^3$) ones were collected in separate tanks and sent for disposal. Maximum levels of alpha and beta-gamma activity of the liquid effluents discharged from the fuel fabrication facility for processing were nearly 1.2×10^{-6} $\mu Ci/cm^3$ and 5.6×10^{-6} $\mu Ci/cm^3$ respectively; the net activity figures over a year for alpha and beta-gamma were nearly 2 mCi and 10.5 mCi respectively. Solid wastes, suspected to contain Pu were segregated in standard containers marked 'active' while non suspect wastes were handed over to the waste management facility. Low active solid wastes generated were to the extent of 15 - 20 packets, each of 2 c.ft. volume and the packets had a maximum surface dose rate of 1 mR/h and these were also sent for disposal.

Criticality Safety

Preliminary clearance was limited to 250 g Pu in the sintering furnace glove box, taking into account the possibility of the coolant line rupture. Presence of two batches, each of 250 g Pu, was permitted in either of the high active halls at any time. Later, on the basis of operating experience, the quantity of Pu for sintering was progressively increased to 500 g Pu and the same handling limit, was enforced for the welding box too. Administrative control ensured that water or other hydrogeneous materials were not brought inside the box; however small quantities, required for specific operations, were permitted after special clearance.

Birdcages have been fabricated to store and transport the fuel pins. The birdcage consists of a mild steel slotted angle frame work with an aluminium container, rigidly fixed at its centre. Inside the Al container is a square cluster of nine aluminium tubes welded together at the top and bottom to form a bundle. Each of the tubes accommodates one fuel pin in a PVC bag; thus nine pins, amounting to 1.26 kg PuO_2 can be stored in the birdcage. The central Al container is provided with a tight fitting cap with a neoprene gasket to render it leak tight. A prototype birdcage was subjected to drop and water leakage tests and was approved for use.

The birdcages carrying the complete core charge of 'PURNIMA' reactor ($\approx$22 kg Pu) were stored in the Pu store room in a plane array. The birdcages of size 60 x 40 x 40 cm^3 have been designed to maintain between the central Al containers a minimum surface-to-surface separation of 30 cm, to isolate the containers in the event of flooding. Effective neutron multiplication factor of a birdcage with 9 fuel pins, under flooding conditions(including internal flooding) has been estimated as about 0.67. Thus the nuclear safety of the individual birdcage as well as the array was ensured in the event of flooding.

Criticality Monitor

Criticality monitors are located in the two high active halls and Pu store room. The sensing device consists of an ion chamber, connected to a period amplifier. The amplifier gives an indication of the rate of rise of the gamma field during an excursion. The criteria for alarm setting of the system were fixed as follows:

(i) the system shall sound a positive alarm if a criticality burst of 10^{15} fissions occurs at a distance of 30 ft. from the detector and delivers prompt gamma dose in 100 milli-seconds,

(ii) the system shall not give an alarm as a result of handling 10 Ci ^{60}Co source at a distance of about 10 ft from the chamber.

The above criteria will be satisfied if a change in radiation level by 6 decades (i.e. 10 mR/h background to 10^4 R/h) triggers the alarm.

In order to study the response of the monitor, a criticality event was simulated by shooting a ^{60}Co capsule of 1 Ci strength past the ion chamber. The source was ejected with compressed air over a distance of $8\frac{1}{2}$ ft in 0.25 sec, giving a change of field from 10 mR/h to 10^4 R/h. The alarm limit was set at 50% of the maximum deflection obtained during calibration.

Estimation of Pu in Waste and Fuel Pins/Pellets

As a measure of inventory control, instruments were developed to estimate Pu content in solid wastes, PuO_2 pellets and finished fuel pins.

Assessment of Pu in solid waste was carried out by counting low energy X-rays from Pu in the channels, corresponding to 11.0 to 21.5 KeV, with a 1 mm thick and 25 mm diam. NaI(Tl) crystal, having a Be window. Measurements with 1 μCi Pu source, in a 125 mm diam and 175 mm high standard waste container, gave twelve times the background counts in four minutes and this indicated a feasibility of estimating μg levels of Pu in solid wastes.

A BF_3 filled annular counter was developed to measure the neutron emission from PuO_2 pellets and fuel pins. Since Pu has been obtained from reprocessing of low burn-up fuel, it was possible to estimate ^{239}Pu and ^{240}Pu content in the pellets and pins knowing the neutron yields from spontaneous fission and (alpha, neutron) reactions in PuO_2. An approximate assessment of the neutron dose rate could also be made from the measurements.

Conclusion

Safe operation of the facility has been amply demonstrated by low personnel exposure and absence of unsafe incidents and this has given an incentive to fast reactor fuel development programme.

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