OPERATIONAL EXPERIENCES IN RADIATION PROTECTION IN FAST REACTOR FUEL REPROCESSING FACILITY

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Abstract. The COmpact Reprocessing facility for Advanced fuels in Lead cells (CORAL), situated at Indira Gandhi Centre for Atomic Research (IGCAR), Kalpakkam is a pilot plant to reprocess the mixed carbide fuel, for the first time in the world. Reprocessing of fuel with varying burn-ups upto 155 GWd/t, irradiated at Fast Breeder Test Reactor (FBTR), has been successfully carried out at CORAL. Providing radiological surveillance in a fuel reprocessing facility itself is a challenging task, considering the dynamic status of the sources and the proximity of the operator with the radioactive material and it is more so in a fast reactor fuel reprocessing facility due to handling of higher burn-up fuels associated with radiation fields and elevated levels of fissile material content from the point of view of criticality hazard. A very detailed radiation protection program is in place at CORAL. This includes, among others, monitoring the release of ^{85}Kr and other fission products and actinides, if any, through stack on a continuous basis to comply with the regulatory limits and management of disposal of different types of radioactive wastes. Providing radiological surveillance during the operations such as fuel transport, chopping and dissolution and extraction cycle was without any major difficulty, as these were carried out in well-shielded and high integrity lead cells. Enforcement of exposure control assumes more importance during the analysis of process samples and re-conversion operations due to the presence of fission product impurities and also since the operations were done in glove boxes and fume hoods. Although the radiation fields encountered in process area were marginally higher, due to the enforcement of strict administrative controls, the annual exposure to the radiation workers was well within the regulatory limit. As the facility is being used as test bed for validation of prototype equipment, periodic inspection and maintenance of components such as centrifuge, extractors and chopper consumed a sizable fraction of the external exposure dose expenditure. The frequent degradation of gauntlets due to the presence of organic medium resulted in contamination and airborne activity. There have been a few cases of personal contamination and internal exposure but the level of exposure was well within the prescribed regulatory limits. Minimizing the alpha-bearing waste was another major task. Efforts, such as decontamination of the gauntlets, were initiated to reduce the waste volume. The environmental discharges of particulate radionuclides were below detection limits and the gaseous release (^{85}Kr) was <0.1 % of the authorized limit. Health Physics unit acquired expertise in tackling unique situations and the challenging radiological surveillance at CORAL, one of its kind in the world, was rich and rewarding. The details of the same are reported in the paper.

KEYWORDS: Radiation protection, Health Physics, Reprocessing, criticality, CORAL

1. CORAL Facility – An Introduction

Reprocessing of spent fuel from fast reactors is the key to India's success in its three-stage nuclear power programme. However, it is a challenge for both plant operations as well as the health physics unit due to the diverse ways in which the solutions and the components of high activity are handled. Since there is a significant potential for both internal and external exposures, due to high fissile material content and higher burn-up respectively, utmost care needs to be taken during the design, construction and operation of the plant. CORAL (Compact Reprocessing of Advanced fuels in Lead cells) at Indira Gandhi Centre for Atomic Research (IGCAR), Kalpakkam, is a pilot plant, designed and constructed indigenously, to study and standardize various process parameters for the reprocessing of spent mixed carbide fuels from Fast Breeder Test Reactor (FBTR) [1]. Moreover, it is the only one of its kind in the world where reprocessing of carbide fuels, irradiated upto a maximum burn-up of 155 GWd/t at FBTR, have been carried out successfully. This paper highlights in brief, the radiological aspects associated with the plant and health physics experiences gained during the reprocessing operation.

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1.1 Process Description

Modified PUREX process is employed at CORAL for reprocessing of FBTR spent fuel [1]. Fuel pins, loaded in a fuel magazine, are received in a specially designed α-tight container. Fuel pins are chopped using a single pin chopper followed by dissolution. Solvent extraction is carried out by PUREX process for decontamination from fission products. The product solution containing purified Plutonium nitrate and Uranyl nitrate is separated into Plutonium oxalate precipitate and Uranyl nitrate supernatant by oxalate precipitation. The supernatant Uranyl nitrate is sent for precipitation by ammonium diuranate process. The solid products are calcined into plutonium oxide and uranium oxide.

1.2 Plant Description

As any other reprocessing plant, CORAL also is divided into CORAL operating area (COA), plutonium re-conversion lab (PuRL), active analytical lab (AAL), uranium re-conversion lab (URL), storage vault, waste vault etc.

CORAL operating area essentially houses a containment box (CBOX) of size 10.1x1.2x1.5m. This is leak-tight stainless steel glove box fitted with master slave manipulators where chopping, dissolution and solvent extraction process are carried out. The containment box is shielded with 200 and 250 mm thick lead in the sides and 400 mm thick mild steel at the top. The containment box houses single pin chopper, electrolytic dissolver, centrifuge, five extractor units and the related process tanks are located beneath the cell. Partition box (PB), an extension of CBOX, consists of second cycle stripping, partition cycle extractors and connected tanks.

Two glove boxes, called ‘top containment’ boxes (TC box), are placed above the containment box, namely aqueous and organic, in which the ejectors required for various operations, tank vents and other related utilities are located. Another glove box, called ‘Blister Box’ (BB), is connected to containment box through a tunnel to undertake maintenance work of any active component and/or to carry out any other CBOX related work. Dissolver-Off-Gas treatment box (DOG box) is yet another glove box, where three off gas scrubber columns and booster ejector that is required to create additional negative pressure during chopping and dissolution and related tanks are located.

2. Radiological Aspects of Fast Reactor Fuel Reprocessing

The fuel discharged from a fast reactor is high in fissile material content as well as in fission product activity, as compared to fuel discharged from a thermal reactor as can be seen in Table 1 [2]. For FBTR, these figures are still higher due to higher Pu content (70%) and higher burn-up (155 GWd/t) with shorter cooling periods.

Table 1: Typical comparison between thermal and fast reactor fuel reprocessing (TRFR & FRFR)

S. No	Particulars	TRFR	FRFR
1.	Pu/(U+Pu)	0.004	0.15 to 0.7
2.	Specific $\beta\gamma$ activity	~7400 GBq/l	~37000 GBq/l

The isotopic composition of Plutonium varies depending on the burn-up of the fuel at discharge and on the level of Uranium enrichment. It is also well known that the nuclear properties of the different plutonium isotopes differ as shown in Table 2 along with americium, which is included due to its gamma component [3].

As a result, the neutron flux, $\beta\gamma$ activity and the overall plutonium content are strongly dependent on the isotopic composition of plutonium. It is noted particularly that ^{241}Pu (half life 14.4 y) beta decays to ^{241}Am, which alpha decays with emission of a 59 KeV gamma to the ^{237}Np daughter. Consequently, the gamma radiation level resulting from ^{241}Am, is larger for Pu from high burn-up fuel than from low burn-up and it increases over time to a maximum that is reached after 72 y. As can be seen from the Table 2, external radiation hazard exists for radiation workers during the handling of Pu because the radioactive decay of commonly encountered Plutonium isotopes produce gamma rays and X-rays and ^{238}Pu, ^{240}Pu and ^{242}Pu produce spontaneous neutrons. Low and medium energy gamma rays, mainly from ^{241}Pu decay products (primarily ^{241}Am), are the major contributor to the external exposure. Therefore special attention is to be given to the growth of ^{241}Am. The gamma emission from other Pu decay products may become the major external gamma radiation source if the ^{241}Am is either removed or these low energy gammas are attenuated by shielding. The significance of neutron dose rate depends on the content of Pu isotopes handled, spontaneous fissions and (α, n) reaction with light elements. The growth of transplutonics, like ^{242}Cm and ^{244}Cm, albeit low, contribute significantly to neutron dose arising from high neutron flux in the form of spontaneous fission (10^7 n/s/g). Hence, handling of large quantities of plutonium requires special consideration from criticality safety point of view and high gamma emitting fission products due to high burn-up are the two major problems associated with fast reactor fuel reprocessing, from the standpoint of radiation protection.

Table 2: Nuclear properties of plutonium isotopes and americium

Isotope	Half-life (y)	Decay mode	Sp.Activity (GBq/g)	Spontaneous neutron rate (n/g/s)	^{a} Dose rate (mSv/h)		
					X-rays	Gamma rays	Spontaneous neutron dose rate
^{238}Pu	88	α	600	2.6×10^3	5700	240	640
^{239}Pu	$2,4 \times 10^4$	α	2	0.03	89	3.2	<0.01
^{240}Pu	6.5×10^3	α	8	1.02×10^2	72	0.8	300
^{241}Pu	14.4	β	3700	8.8×10^2	--	120	--
^{242}Pu	3.8×10^5	α	0.1	1.7×10^3	1.3	--	310
^{241}Am	4.3×10^2	α	120	1.1	4000	27000	0.15

^{a} dose rate per kg of pure nuclides.

2.1 Criticality Control

Significant quantities of fissile materials are handled both in liquid and solid form at CORAL. Typical isotopic composition per kg of spent mixed carbide fuel (in gram) is given in Table 3. The data for 155 GWd/t burn-up is not given, as the campaign is currently in progress. In view of the potential for a criticality event, criticality control is absolutely necessary during all stages of the plant operations. Mass control is implemented where fissile material is handled in solid form such as fuel transport, chopping, process equipments and storage vault etc. Wherever fissile material is in solution form, control is ensured through both geometry and fissile concentration. The geometry of the tanks includes annular, slab and vertical cylinder types. The tanks are made up of material type AISI SS 304L to avoid corrosion due to high concentration of fissile material content. Fissile material storage containers are designed with mass control and birdcages are used for this purpose to avoid any interaction effect. Detailed theoretical analyses were done to study the effect of interaction between process tanks, effect of reflectors on criticality of the entire system by exact modeling. Results show that k-eff in all cases lie from 0.6 to 0.85 [4].

2.1.1 Criticality Accident Detection

Though a reprocessing plant is intrinsically stable and not susceptible to rapid fluctuations from normal conditions, it is postulated that there exists a probability of criticality accident, wherever unsafe geometry tanks are used. Hence, it is necessary to have criticality monitors and alarm annunciation systems at strategic locations for initiating immediate evacuation of personnel in the vicinity.

Table 3: Isotopic composition of spent mixed-carbide-fuel

Isotope	Burn-up			
	25 GWd/t	50 GWd/t	75 GWd/t	100 GWd/t
^{235}U	2.05	1.95	1.87	1.78
^{238}U	296.8	294.1	291.5	288.9
^{237}Np	9.2×10^{-3}	1.8×10^{-2}	2.8×10^{-2}	3.7×10^{-2}
^{238}Pu	4.7×10^{-3}	9.2×10^{-3}	1.4×10^{-2}	1.8×10^{-2}
^{239}Pu	630.3	601.2	573.5	547.2
^{240}Pu	45.8	49.0	52.0	54.8
^{241}Pu	3.8×10^{-1}	7.7×10^{-1}	1.16	1.55
^{242}Pu	1.8×10^{-3}	7.1×10^{-3}	1.6×10^{-2}	2.8×10^{-2}
^{241}Am	4.2×10^{-3}	1.6×10^{-2}	3.6×10^{-2}	6.4×10^{-2}
^{242}Cm	2.0×10^{-5}	1.4×10^{-4}	4.0×10^{-4}	8.2×10^{-4}
^{244}Cm	8.4×10^{-9}	1.35×10^{-7}	6.8×10^{-7}	2.1×10^{-6}

The majority of process tanks are geometrically safe. However, operational requirement demands the need for a few high-volume storage tanks, which are ought to be unsafe geometry ones due to space constraints. These tanks act as critical ones for mandatory continuous monitoring of accidental criticality. In all, five criticality alarm systems (CAS) are installed in CORAL plant at locations as deemed necessary. The locations of the CAS detectors were determined on the basis of plant layout and theoretical estimation of dose levels using QADCGPIC [5].

The CAS installed at CORAL is based on the detection of prompt gamma emission during a criticality incident and it is capable of detecting criticality accidents even when detectors are placed at a distance of 30m from an unshielded source of an accident. Each CAS comprises of three criticality monitors (ionization chamber) connected to alarm annunciation system. When any two of the three monitors of a Criticality Alarm System (2/3 logic) detect the dose levels exceeding the preset values for alarm condition, criticality alarm is actuated. In case of AC power failure, the monitors get automatically connected to battery thus ensuring continuous availability even during temporary power failures. The CAS monitor gives an alarm whenever gamma radiation dose rate at the detector exceeds 30 μGy during a fast criticality accident of duration ≤ 500 ms and also if the overall gross dose rate exceeds 40 mGy/h during a slow criticality accident, conforming to International Standards [6].

Due to relative high Pu content in fast reactor fuel reprocessing, special efforts were made in testing the criticality monitor for its fast transient response viz., a dose level of 30 μGy in 500 ms. Towards this, an innovative test rig facility was designed, where the rapid movement of the source from the lead shield results in sudden rate of rise of gamma field simulating a criticality event. Sensors for the precise estimation of the time of contact between the source and the detector have been fixed [7].

Another challenge was experienced when the high burn-up fuel pins were brought inside the CBOX for chopping, the threshold alarm limit of 40 mGy/h of CAS monitor exceeded due to high βγ activity in the fuel pins and the criticality alarm annunciated. To avoid occurrence of such non-genuine alarms from the criticality point of view, a complete review of locations of CAS was done and recommended provision of shielding to the CAS detector while at the same time ensuring that the actual criticality incidents do not get undetected.

As a sequel to this, in order to confirm the genuineness of CAS alarm, deployment of NaCl in pouches at strategic locations was resorted to. In case of an alarm from CAS, the pouches would be retrieved by remote means, without entering the operating area, and the induced gamma activity (especially due to ^{24}Na) measured from the pouches containing NaCl, which would confirm the occurrence of a criticality event or otherwise. As a prelude to this, experiments were carried out in a reactor by irradiating known amount of NaCl pellets for various time durations and power levels to arrive at exact quantity of NaCl that is to be deployed vis-à-vis the fission yield and minimum detectable level.

In order to estimate the dose received during a criticality accident, criticality badges are deployed at the operating area. However, before deploying them, it is essential to have a technical basis for selecting a suitable location for placement of such criticality badges. Towards this, a simple hypothetical criticality incident in the unsafe geometry tanks containing process solution in the containment box was simulated; the neutron leakage flux and the gamma dose are estimated as part of optimization study from the point of view of detection of minimum credible accident (10^{13} fissions) and the Minimum Detectable Activity (MDA).

2.2 Handling High Inventory Fission Products

The fission product activity encountered in the facility varied from 116 to 319 TBq/kg depending on the burn-up and cooling time. The major radionuclides that contribute to activity include: ^{85}Kr, ^{89}Sr, ^{90}Sr, ^{90}Y, ^{91}Y, ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{106}Rh, ^{129}Te, ^{131}I, ^{137}Cs, ^{140}Ba, ^{140}La, ^{141}Ce, ^{143}Pr, ^{147}Nd, ^{147}Pm, ^{129}I, ^{14}C and ^{3}H. Table 4 shows gamma activity from fission products for different burn-up of fuel with one-year cooling time.

Table 4: Fission product activity for different burn-up of spent fuel for one year cooling period

Burn-up (GWd/t)	Fission product activity (TBq/kg)
25	115.56
50	192.96
75	248.78
100	290.09
155	318.86

Irradiated fuel pins are transported to the plant in a specially designed cylindrical cask to ensure the dose rate outside of the surface of the cask not exceeding the regulatory limit of 2 mSv/h. All processes like chopping, dissolution and solvent extraction studies are carried out in the CBOX. The shielding adequacy of this box was evaluated by performing calculations using point kernel code QADCGPIC with accurate modeling of process tanks with maximum gamma activity [8]. It was recommended to provide additional shielding wherever it is found to be necessary during operational phase by wrapping the pipelines carrying high active solutions with lead sheets to reduce the dose rate levels in normal occupancy areas.

The burn-up of the FBTR fuel was increased based on the irradiation behavior of the fuel, progressively from 25 to 50 and then to 100 and finally now to 155 GWd/t. To reprocess this spent fuel it was felt essential that shielding adequacy for reprocessing higher burn-up fuels has to be periodically computed, updated and checked. Towards this, the gamma dose rates have been computed using QAD-CGPIC for a one year cooled fuel of 155 GWd/t irradiated fuel pins at critical locations and around the operating area of the containment box, along the line of sight of each source in the cell and at CAS detector locations on top of the lead cell. It was found that the existing shielding was found to be adequate.

3. HP Experience gained in the Operation of CORAL

3.1 Routine Monitoring

Area Gamma Monitors (AGM) have been installed in various locations of the plant in order to monitor the ambient radiation levels. Radiation levels in the normal occupancy areas are around 2-3 μGy/h. However, during maintenance operations of some active equipment the radiation levels increased up to 20-30 μGy/h. In addition to the installed AGMs, radiation survey is carried out at various locations using portable survey instruments to find out any increase in levels. Any work involving an exposure rate greater than 100 μGy/h, is linked to Radiological Work Permit (RWP) system for control of personal exposure.

Prior to taking up the maintenance of active components that are housed inside CBOX, as part of ALARA concept, it was decided to evaluate remotely the gamma dose rate level on the components in question. Towards this, TLD powder containing $CaSO_4$:Dy in capsule forms was used to estimate the dose rate. This method was beneficial in planning the man-rem expenditure and recommending the precautions to be taken during maintenance activities. On similar lines, neutron dose estimation was carried out using bubble detectors.

3.2 Air Activity Monitoring

Continuous Air Monitors (CAM) are installed at potential areas to continuously monitor the ambient air for any suspended air-borne radioactivity. Besides, air-borne activity levels are monitored in every shift as well as during any special operations, using portable air samplers. During the early stage of commissioning, it was observed that the CAMs in the operating area annunciated alarm frequently. The investigation revealed that the alarms were due to diurnal variation in the levels of short-lived radon / thoron daughter products. Based on this, improvement in the area ventilation balancing was carried out by the plant, which drastically reduced the frequency of false alarms. The alarm levels were arrived at after considering various factors such as diurnal background fluctuations, detector efficiency etc. Very few incidents related to air activity encountered during the operation of the plant are experienced. Even in these incidents, the air activity levels were negligible from the point of view of internal exposure and the cause for the same could be identified and arrested immediately.

On one occasion, during gauntlet changing operation in a glove box, alpha air activity, around 1.2 DAC was detected by air sampling. Nasal swab from one of the persons who were involved in the operation showed 1.5 Bq. As a precautionary measure, the concerned person was promptly referred for special bioassay monitoring and the results indicated committed effective dose of 3.7 mSv.

On another incident, upon alarm by a CAM near deep bed filter, investigation was carried out. The filter papers were subjected to gamma ray spectral analysis and presence of ^{134}Cs and ^{137}Cs was confirmed. The cause for the alarm was traced to a leak in the valve in a pipeline leading from deep bed filter. However, there was no internal exposure in this incident. The leak in the valve was promptly arrested.

3.3 Radiation Data Acquisition System (RDAS)

All the installed radiation monitors are connected to a centralized radiation data acquisition system installed at control room. It has the capability to store data from all monitors, which can be retrieved at any point of time. These data are displayed on the monitor for instant reference and effective monitoring.

3.4 Environmental Monitoring

The gaseous and particulate effluents generated from the plant are discharged through a 75 m stack. The stack-monitoring system consists of a CAM, which monitors the sample air drawn through iso-kinetic sampling technique. While the alpha and beta particulate activities are monitored thus, the gaseous activity, predominantly ^{85}Kr, is monitored by passing the sample air through a 2-liter leak tight cylinder containing a glass walled GM. Whilst the gross alpha and gross beta particulate activities released through stack, in each year, were below detection limit (BDL), the activity of ^{85}Kr released was far below the annual limit prescribed by the regulatory authority (7×10^7 GBq). The activity of ^{85}Kr released through stack during the past five years is presented in Table 5.

To improve the quantification of the discharges more accurately, an experimental set up was designed, fabricated and installed to estimate the release of ^{85}Kr and particulate activities on a continuous basis near the source of generation instead of at stack where there is large dilution. For this purpose, a system was installed in the VOG line in upstream of Deep Bed Filter. This has given encouraging results and this should be one of the lessons learnt for nuclear plants in general and in particular for future reprocessing facilities.

Table 5: Gaseous effluent (^{85}Kr) released through stack

Year	Krypton-85 (GBq)
2003	4.44
2004	2.38
2005	7.85
2006	5.89
2007	18.5

Bore wells are provided around the delay tank and underground waste vault. Periodic samples are collected from these wells and analyzed for gross alpha, gross βγ activity to check the breach of containment of the systems.

3.5 Personnel Monitoring

About 200 radiation workers are issued Thermo Luminescent Dosimeters (TLD) every month and their dose records are well documented in a computerized dose management system with a feature to view the dose history of an individual at any given time. Control of exposures, during special operations under RWP, is exercised by issuing Direct Reading Dosimeter (DRD). The annual cumulative dose received by radiation workers during the period 2003-2007 is shown in Table 6. It is observed that none of the workers exceeded the annual dose limit in spite of the fact that virgin solutions with high specific activity were handled and the maintenance of components with high beta-gamma activity were undertaken.

Table 6: Dose distribution for the period 2003-2007

Year	Total dose received (mSv)					
	0	0-5	5 - 10	10 -15	15-20	>20
2003	205	37	1	0	0	0
2004	140	92	0	0	0	0
2005	133	57	0	0	0	0
2006	55	121	8	3	1	0
2007	89	142	16	5	1	0

Assessment of internal exposure is done through whole body counting and bioassay. While the routine monitoring of whole body counting is carried out for all occupational workers and contractors on annual basis, bioassay is done for the critical group of personnel to have the base line data. For all those who join newly the plant, bioassay and whole body counting are mandatory prior to enumerating them as a radiation worker and allotment of TLD badges. Internal dose monitoring is sub divided into three distinct types: 1) Routine monitoring, 2) operational and 3) special monitoring. Routine monitoring is essential in all such cases where, apart from the likelihood of annual exposures exceeding 30 % ALI, contamination of work place may occur resulting from either normal operations or random events or due to minor incidents. Operational monitoring refers to the monitoring of workers as well as contractors who have carried out a particular operation in which internal exposure is anticipated. Special monitoring is the monitoring of a worker involved in a minor incident leading to spillage or spread of contamination, which might have resulted in an internal contamination of the worker. The hospital attached to IGCAR is fully equipped with conventional Zn/Ca-DTPA therapy procedures to deal with internal exposure cases involving Pu.

An elaborate OPEC (Operating Procedures under Emergency Conditions) document in case of a criticality accident has been brought out highlighting the roles and responsibilities of the plant personnel as well as Health Physicists. A brief outline for the reentry into the plant after such a postulated criticality incident scenario is also included.

3.6 Radioactive Waste Management

The classification, limits and management of the solid and liquid wastes generated from the plant are done as per guidelines issued by the regulatory authority. Solid wastes are collected, segregated and stored until collection by Centralized Waste Management Facility (CWMF) for disposal. All types of particulate activity filters are handled as solid wastes whenever they are replaced and sent for disposal. It may be mentioned that the activities of both solid and liquid wastes disposed from the facility were well within the regulatory limits. High-level liquid wastes are stored within the plant complex with adequate cooling provision in the storage tanks. The inventory of the tanks is regularly checked and recorded. Sumps are provided to collect the sample in case of leakage of solution. Low-level radioactive liquid wastes are transferred to CWMF through delay tanks for further processing and disposal. The activities of wastes disposed were in the range of alpha: 10 to 300 MBq and beta 50 to 2175 MBq respectively.

4. Training and Emergency Preparedness

Training in radiation protection to all occupational workers and contractor workers is given utmost importance. The syllabus for the training program includes, inter alia, the criticality safety aspects, handling of fissile material and emergency preparedness procedures. Followed by class room training program, examinations and viva voce are conducted. Refresher training is arranged to all concerned once in three years.

Plant emergency exercises are periodically conducted once in three months as per plant document on emergency preparedness and improvements made on the basis of feedback. Postulated hypothetical scenarios were constructed and appropriate action plans formulated. In addition, a detailed criticality emergency preparedness action plan is available in the plant [9].

5. Conclusion

Despite the inherent challenges involved in the reprocessing of fast reactor fuel, the Health Physics Unit at CORAL could effectively implement ALARA principle during the entire operation of the plant, with the active involvement of the plant management. Each and every stage of the process was a wealth of information both for operations as well as for the health physics personnel. The rich and rewarding experience gained during this landmark achievement in Indian reprocessing programme will go a long way in building confidence to meet future goals.

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