EMERGENCY EXHAUST SYSTEM'S EFFICIENCY MEASUREMENTS FOR IODINE REMOVAL AT PARR-1

by

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The emergency exhaust system of the Pakistan Research Reactor–1 (PARR-1) incorporates charcoal bed filters for the removal of radioiodine from exhaust gases in case of an emergency involving the release of radioiodine. The filters adsorb radioiodine. Testing of the filters is necessary so as to assess their iodine retention efficiency, if they are to meet regulatory requirements. The retention efficiency of our filters has been measured and found to be 99.1%. This value is higher than the value of the activated charcoal filter efficiency (99%) employed to determine the source term for release through emergency exhaust with filtration, for dose calculations. In this paper, the procedure for testing the efficiency of charcoal filters has been discussed and results obtained presented.

Key words: Pakistan research reactor-1, emergency exhaust system, charcoal filter, radioiodine, retention efficiency

INTRODUCTION

The PARR-1 is a swimming pool-type research reactor using highly enriched uranium (HEU) fuel which reached its first criticality in December 1965. The reactor was operated at a power level of 5 MW. In 1991, the reactor pool was lined with stainless steel and the reactor core was converted to low enriched uranium (LEU). At the same time, reactor power was increased to 10 MW. The operating equilibrium core with LEU fuel is shown in fig. 1. The ventilation system of the reactor building was also renovated. The reactor building has an internal diameter of about 26 m and its height is 33 m. The free volume of the building is about 15000 m^3. The heating, ventilation and air conditioning (HVAC) system of the reactor building is designed to contain released radioactivity by filtering the exhaust air through absolute filters and charcoal activated filters. In order to avoid leakage from the containment building into the environment, a negative pressure of a 20 mm water column is maintained in the reactor building during the operation of the reactor. The normal exhaust system which contains an absolute filter remains in operation all the time, even when the reactor is in the shut down condition. However, there are some sub-systems which are functional only when the reactor is in operation. The expected gaseous

	1	2	3	4	5	6	7	8	9	
A		GR	GR	SFE	SFE	SFE	SFE	SFE	SFE	
B		GR	GR	SFE	SFE	SR#5 CFE	SFE	SR#1 CFE	SFE	
C		FC-A	GR	WB	SFE	SFE	WB	SFE	SFE	
D		GR	WB	SFE	SR#4 CFE	SFE	SFE	SFE	SR#2 CFE	
E		GR	GR	SFE	SFE	SFE	SR#3 CFE	SFE	SFE	
F		FC-B	GR	SFE	SFE	SFE	SFE	SFE	SFE	

Thermal column

Figure 1. Matrix representation of the equilibrium PARR-1 core – horizontal cross-section
Grid plate size 467 mm × 737 mm, A-F are rows and 1-9 columns, SFE – standard fuel element, CFE – control fuel element, FC – fission chamber, WB – water box, GR – graphite reflector and SR – shim rod

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waste to be produced during the postulated accident is diverted towards the exhaust stack through the emergency exhaust system and released into the atmosphere at a height of about 61 m. In the event of an increase in reactor power or build-up of pressure, or primary coolant activity, the normal ventilation system is shut off and the emergency ventilation system put into operation automatically. In addition to the pre-filters and absolute filters, the emergency exhaust has activated charcoal filters to adsorb the radioiodine from the air coming from the reactor hall. The air is pumped through the stack by the emergency exhaust fan, so as to release the pressure that might have built up inside the reactor hall in case of an accident and to restore it to a slightly negative one.

Experimental procedures for testing the efficiency of a charcoal filter [1] by using ^{127}I vapors at its inlet and operating the emergency exhaust system of PARR-1 have been described, the experiment conducted and results presented in this paper.

PRODUCTION OF IODINE ISOTOPES IN THE REACTOR CORE

The various isotopes of iodine produced by fission during the operation of a nuclear reactor are ^{129}I, ^{131}I, ^{133}I, ^{134}I, and ^{135}I. Except for ^{129}I having a very long half-life, all the above-mentioned radioisotopes reach their equilibrium (saturation) value in the reactor core. The equilibrium value of these isotopes is given in tab. 1.

Among these, ^{131}I is, effectively, the predominant isotope from the dose point of view. Short-lived isotopes, although more active, have a lesser contribution to the dose, because these decay within a short time. The main characteristics of these isotopes are given in tab. 1 [2, 3].

Table 1. Main characteristics of different isotopes of iodine

Isotope	Half-life	Fission yield [%]	Main radiation emitted in MeV/disintegration		Saturation activity of iodine in the core at 10 MW [Bq]
			β	γ	
^{131}I	8.04 days	2.89	0.19	0.38	$3.54\ 10^{15}$
^{132}I	2.28 hours	4.31	0.49	1.29	$1.24\ 10^{16}$
^{133}I	20.8 hours	6.69	0.41	0.61	$1.93\ 10^{16}$
^{134}I	52.6 minutes	7.79	0.61	2.61	2.54 10
^{135}I	6.58 hours	6.29	0.377	1.65	1.81 10

RELEASE OF IODINE AND ITS RISKS

Iodine can be released in case of a core meltdown. Minor leakage is also possible due to a pinhole in any fuel plate. About 50% of the iodine will be released from the core and will escape into the containment in the form of iodine gas in the case of a loss of coolant accident (LOCA) [3–7], resulting in core meltdown.

It has been reported [2] that about 91% of the released activity due to iodine will be in the form of elemental iodine, 5% in the form of particulate iodine, and 4% in the form of organic iodides. Iodine will get adsorbed on paint and metal surfaces. It will also be entrained in steam and water vapors and condensed on the floor. As a conservative assumption, 25% of the equilibrium radioactive iodine inventory will reach the filters. The activated charcoal filters will adsorb a major portion while, depending on their retention efficiency, a part will escape and enter the environment from the top of the stack. Estimated radioiodine emissions in the case of a LOCA resulting in core meltdown are given in tab. 2. Maximal concentration will occur at a distance of approximately 300 m from the stack, resulting in a thyroid dose of 21.7 mSv in 30 days (for releases through the emergency exhaust with filtration [3]), under assumed metrological data:

– stack height: 61 m,
– average wind speed at stack top: 2.9 m/s,
– average wind speed at ground level: 2.0 m/s, and
– wind direction: WSW to ENE.

Table 2. Maximum possible iodine activity released into the environment from the stack in case of core meltdown

Isotope	Activity [Bq]
^{131}I	$8.85\ 10^{12}$
^{132}I	$3.1\ 10^{13}$
^{133}I	$4.82\ 10^{13}$
^{134}I	$6.35\ 10^{13}$
^{135}I	$4.5\ 10^{13}$

ADSORPTION OF IODINE BY ACTIVATED CHARCOAL FILTERS

Activated charcoal is a form of commercially pure charcoal with a microscopic physical structure. Each grain of charcoal contains millions of tiny passage-ways providing a tremendous internal surface area of the order of 700-1800 m^2/g [2] which accounts for its adsorptive power. Charcoal is used as an air-purifier and available in a wide variety of grain sizes, powders, pellet forms, cloth, sheets, *etc*. Different types of fabricated filters for various uses are available. Iodine is removed very efficiently by acti-

vated charcoal filters but, methyl iodide, released along with it, cannot be removed efficiently by activated charcoal filters alone. The charcoal is impregnated with potassium iodide and iodine so as to improve the efficiency of the filters for removing methyl iodide. The reaction of CH_3I with the impregnate of charcoal is:

$$K^{127}I + CH_3{}^{131}I \leftrightarrow K^{131}I + CH_3{}^{127}I \qquad (1)$$

Charcoal Airborne Charcoal Airborne

The emergency exhaust filter contains 27 kg of activated charcoal enabling it to adsorb the potential radioiodine.

EXPERIMENTAL WORK

Experimental set-up

The experimental set-up shown in fig. 2 includes the system generating iodine vapors in the reactor hall exhaust in front of the charcoal filters. Iodine vapors are passed through the charcoal filter by operating the emergency exhaust system. The operation of the air samplers at the inlet and outlet of the charcoal filters is required to adsorb iodine on the samplers' charcoal from the air being passed through the emergency exhaust system. The irradiation of charcoal samples collected from the two samplers in the reactor core is, then, carried out. Irradiated samples are measured for ^{128}I and the efficiency of the charcoal filters is determined.

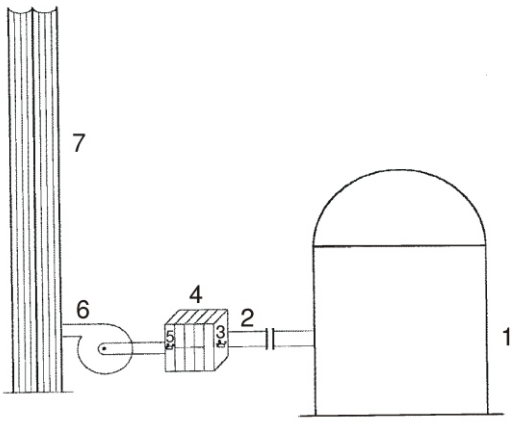

Figure 2. Experimental setup
(1) – reactor building, (2) – emergency duct, (3) – inlet sampler, (4) – activated charcoal filter, (5) – outlet sampler, (6) – emergency exhaust fan EF-15, (7) – exhaust stack

Air sampler

The experimental arrangement for collecting airborne iodine samples at upstream and downstream sampling points (inlet and outlet of the charcoal filters) is shown in fig. 3. Air sample collectors were fabricated by sandwiching a thin layer of activated charcoal between filter papers which were then attached to the two air samplers placed at the inlet and outlet sampling points. For identical sampling, the loaded amount of charcoal in each sample collector was precisely weighed and made equal. A valve was introduced between the sample collector and sampler pump to assure the same flow rate in both inlet and outlet air samplers. Said experimental procedures are described in the following subsections.

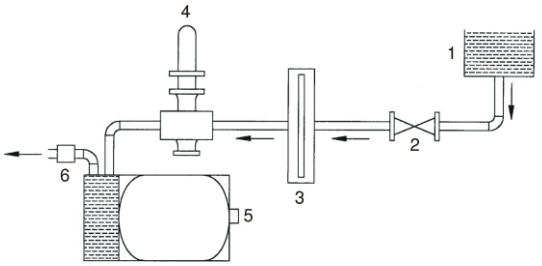

Figure 3. Air sampler
(1) – activated charcoal sandwiched between filter papers, (2) – valve for flow control, (3) – flow meter 0-100 l/min, (4) – automatic flow control valve, (5) – sampler pump (rotary vane type), (6) – filter

Experimental procedures

- Inlet sampler placed at the inlet sampling duct of the activated charcoal filter.
- Outlet sampler placed at the outlet sampling duct of the activated charcoal filter.
- Samplers switched on and airflow adjusted to 40 l/min for both.
- Grill on the intake duct leading to the emergency exhaust system removed.
- Hot plate placed in front of the intake duct leading to the emergency exhaust system.
- Ten grams of iodine crystals placed in a glass dish on the hot plate.
- The emergency exhaust system switched on.
- The hot plate turned on, approximately five minutes later, iodine vapors seen, heating continued for another two minutes, hot plate switched off.
- When the hot plate and glass dish were cooled to room temperature, the iodine crystals from the glass dish were collected and weighed and about 5 g of iodine vaporized and passed through the emergency exhaust activated charcoal filter.
- The grill on the intake duct was placed at its proper position.
- Five minutes after removing the glass dish, the emergency exhaust system and inlet and outlet samplers were switched off and the latter removed.

Neutron irradiation of ^{127}I adsorbed on charcoal

The activated charcoal with the adsorbed ^{127}I was collected separately from the two samplers and labeled as inlet and outlet. Activated charcoal samples taken from both inlet and outlet samplers, weighing 0.355 g in each vial, were prepared for thermal neutron irradiation of the ^{127}I which was adsorbed on the charcoal of the samplers. The samples were irradiated in the reactor core at a thermal neutron flux of 10^{12} n/cm^{2}s in a period of 600 seconds. The neutron activation of ^{127}I resulted in the production of ^{128}I which is a radioactive isotope of iodine with a half-life of 25 minutes. After irradiation, the two samples from the inlet and outlet were counted for ^{128}I.

EXPERIMENTAL RESULTS AND DISCUSSION

The instrumentation used to measure 443 keV gamma rays from ^{128}I consisted of a high purity germanium (HPGe) detector operated at liquid nitrogen temperature with associated electronics and a computer-based multi-channel analyzer (computer/MCA). Standard sources were used for energy calibrations. Gamma rays spectra due to ^{128}I of 443 keV for both irradiated samples were recorded on the HPGe detector with the provision for dead time correction. Radioactivity measurements of samples for both the outlet and inlet samplers were carried out on the same detector, for 300 s; therefore, the difference in the starting time of the measurements for the two outlet and inlet samplers, *i. e*. the delay time of 420 s, was taken into consideration. Based on the delay time, corrected counts were determined using the radioactivity formula [4]

$$A \quad A_o e^{\lambda t} \qquad (2)$$

where A_o is the activity measured at zero delay time, A – the activity measured after delay time of t seconds, λ – the decay constant, and t – the delay time between the two counts.

Experimental results are given in tab. 3. The energy spectra of inlet and outlet samples have been shown in figs. 4 and 5. The percentage of efficiency was calculated as

$$\text{Efficiency} \quad 1 \quad \frac{C_{out}}{C_{in}} \quad 100\% \qquad (3)$$

where C_{out} is the counts in the outlet sample (for the 443 keV peak, *i. e*., the area under the peak), and C_{in} – the counts in the inlet sample (for the 443 keV peak, *i. e*., the area under the peak). Counting time is 300 s.

Table 3. Activity measurement of ^{128}I in inlet and outlet samples and calculations of filter efficiency

Reading set	Sample No.	Net counts	Decay time [s]	Net counts corrected for decay	Efficiency [%]
1	Outlet Inlet	666 60648	0 420	666 73635	99.10
2	Outlet Inlet	324 45705	420 0	393 45705	99.14

The experiment has been carried out at the PARR-1. Experimental data in tab. 3 show that iodine removal efficiency of the charcoal filters is estimated to be 99.1. This is higher than the 99% used for source term calculations of the PARR-1 during the core meltdown accident, in a scenario in which the emergency exhaust system is operative and fission products are released through the emergency exhaust system [3].

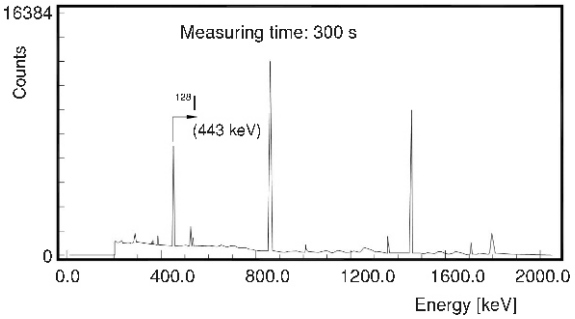

Figure 4. Measurement of ^{128}I in the inlet sample

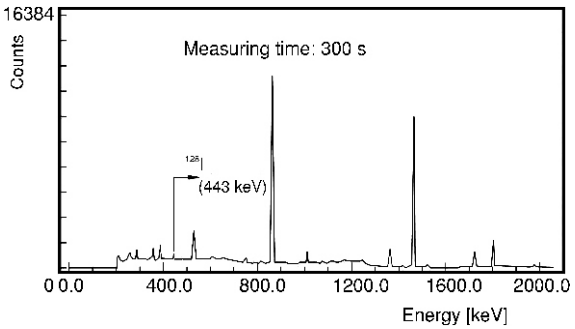

Figure 5. Measurement of ^{128}I in the oultel sample

CONCLUSION

The body of the filter was loaded with activated charcoal a year ago. After a year of operation, the retention efficiency of the activated charcoal filter in the emergency exhaust has been measured and found to be 99.1%, a satisfactory result. The retention efficiency

of the filter should not be allowed to go below 99%, which can be ensured by testing and replacing the ageing charcoal by a fresh supply. Care should also be taken to perform the test with a minimal amount of iodine-127 vapors (say about 2-3 g), so as not to reduce the holding capacity of the charcoal filters, which can be done by reducing the heating time of iodine crystals.

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МЕРЕЊЕ ДЕЛОТВОРНОСТИ СИСТЕМА УДЕСНЕ ВЕНТИЛАЦИЈЕ ЗА УКЛАЊАЊЕ ЈОДА НА ПАКИСТАНСКОМ ИСТРАЖИВАЧКОМ РЕАКТОРУ

Систем удесне вентилације Пакистанског истраживачког реактора (PARR-1) обухвата слој филтара са дрвеним угљем за уклањање радиоактивног јода из испуштених гасова у случају удеса који укључују ослобађање радиоактивних изотопа јода. Филтри адсорбују радиоактивни јод. Тестирање филтара потребно је ради оцене њихове способности задржавања јода, а у циљу постизања регулаторних захтева. Мерена је ефикасност задржавања ових филтара и утврђено је да износи 99,1 %. Ова вредност је виша од вредности ефикасности филтара са дрвеним угљем (99%) која је коришћена да се одреди извор при удесном испуштању са филтрацијом, а ради израчунавања дозе. У овом раду размотрен је поступак за проверу делотворности филтара са дрвеним угљем и приказани су добијени резултати.

Кључне речи: Пакистански истраживачки реактор, систем удесне вентилације, филтар са дрвеним угљем, радиоактивни јод, ефикасност задржавања