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Studies of Iodine Trapping
Plant for Hot Cells

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Introduction

1. The hot cells and metallurgical lines operated by Windscale Nuclear Power Development Laboratories (WNL) and the Atomic Energy Establishment Winfrith (AEEW) have, built into their ventilation systems, fission product trapping plant comprising a high efficiency particulate filter, and a charcoal bed (for the trapping of radioiodine). This paper describes recent work done on different designs of the charcoal bed component of such plant in use, and one under consideration for use; previous similar work has already been published (Ref 1). These units are designed for the trapping of methyl iodide rather than molecular iodine, since that is the most penetrating vapour form of radioiodine confirmed as occurring in a release from irradiated fuel (Ref 2). Although of different mechanical design, all use one or both of two forms of activated charcoal as the absorber: a charcoal base impregnated either with 1.5% by wt. of potassium iodide (KI) or with 5% by wt. of triethylene diamine, (TEDA). A 50/50 mixture of the two is most frequently used. The tests carried out fall into two classes, tests done directly on the plant, and tests done on samples of the charcoal of the plant taken before and after the periodic renewal of the charcoal in the plant, and during its in-service use.

Test methods

2. Details of the method of testing the function of the plant directly have been published elsewhere (Ref 3). Briefly, the method involves the release of a small quantity of methyl iodide, labelled with the isotope I_{131} (8.05 days half-life) into the cell ventilation extract air, whilst the air flow into and out of the absorber bed is being sampled. Methyl iodide in the air samples is trapped in a suitable small mesh size impregnated charcoal and the upstream and downstream sampled iodine activities are compared by γ -counting. This test measures the performance of the plant under normal operating conditions, but because those conditions can vary widely from day to day (particularly in respect of relative humidity) it is a poor indicator of other than gross changes in the condition of the charcoal in the plant; however, early penetration, ie during the first half hour is usually an indication of gas by-pass of the charcoal or seal leakage (or extensive poisoning) whilst later increase in penetration is evidence of desorption resulting from in-service deterioration. The result of this test is usually expressed as a decontamination factor (DF), ie the ratio of inlet activity to outlet activity.

3. The more gradual deterioration in service of the charcoal can be determined from laboratory tests on samples of the plant charcoal, taken from the plant at intervals during its lifetime. The facility for taking such samples and ensuring that they are representative should, of course, be designed in; to shut down the plant, open up the charcoal bed, and extract a sample is not usually a convenient operation and such a sample may not be reliably representative. The standard laboratory test used at Windscale has been described elsewhere (Ref 3); briefly the test consists of the saturation of a sample of charcoal with water by passing through it a stream of air at 98% relative humidity (RH) until equilibrium water content is achieved, followed by a test of its efficiency for retaining methyl iodide, labelled with I_{131} , carried in the gas stream at the same humidity.

The test thus simulates in the laboratory a plant test at 98% RH. This humidity is chosen because the equilibrium water content of the charcoal and the resulting DF measured are consistently reproducible and because it represents the worst conditions for plant operation. Tests at other humidities can be done if specially required, but results tend to be inconsistent and depend on sample history. Laboratory results, when applied to the plant, are thus always pessimistic. The laboratory test is standard in terms of gas flow rate, sample volume, and temperature, as well as RH and methyl iodide loading on the charcoal and no attempt is normally made to reproduce plant conditions. The results for different charcoal samples are then directly comparable, and they may be applied to plant in terms of a parameter $K(s^{-1})$ defined as $K = \log_{10} DF \times (\text{Volume flow rate}) / (\text{Charcoal volume})$. The value of this parameter for a given charcoal sample depends on RH, as implied above; it is not entirely independent of other operating conditions (temperature, bulk density, flow rate, etc) but for most plant purposes may be regarded as so being. $(\text{Charcoal volume}) / (\text{Volume flow rate})$ is also referred to as "residence time" or "staytime" of the gas in the absorber bed.

Uses of the tests

4. The plant test measures the performance of the plant as a whole. Iodine penetration past the absorber bed may do so through inefficiency of the charcoal or through by-passing of the absorber through leaky seals, channeling through poorly packed material, settlement voids, etc. Some of these leaks may be present initially and others may appear in service. The charcoal will lose efficiency by ageing, and may be poisoned by some undesirable contaminant in the air stream. If the partial DF due to leakage is DF_L , and that due to the absorber bed is DF_B , then the effective plant DF as measured, DF_P , is given by

$$1/DF_P = 1/DF_L + 1/DF_B \quad \dots\dots\dots (1)$$

From the laboratory tests and a knowledge of the flow rate and bed volume of the plant we may calculate an expected minimum value DF_C for DF_B , so that

$$1/DF_C > 1/DF_B \text{ and } 1/DF_P < 1/DF_L + 1/DF_C \quad \dots\dots\dots (2)$$

For a newly refilled bed, we can expect DF_C to be quite large, so that $1/DF_C$ tends to zero and $DF_P \approx DF_L$; if DF_P is then considered to be too small, by-pass leakage can then be looked for and remedied and the plant retested until a satisfactory result is obtained. Charcoal samples may then be taken at intervals, and tested in the laboratory and DF_C calculated. As long as DF_C is much larger than DF_P , plant tests only measure DF_L ; but the laboratory tests will show a fall in the value of K found from which deductions can be made about the conditions of the charcoal and from which predictions can be made of the probable performance of the absorber bed for wet gas conditions. Thus, where the design of the bed allows samples to be taken, maintenance of good performance of the bed is based on an initial measurement of DF under normal operating conditions to ascertain by-pass DF followed by the testing in the laboratory of charcoal samples taken from the bed during its life to ascertain the rate of charcoal deterioration. If the bed life allows, further plant tests serve to determine any changes in by-passing or confirm the effects of changes in the charcoal. If samples cannot be obtained, the effects of ageing on the charcoal may be estimated (Refs 4, 5) but not the cumulative effects of any poisoning.

Results

5. Plant No. 1

This plant was an experimental installation in the ventilation line of an empty cell; it was for the purpose of investigating commercially produced 'modular units' which would be readily changeable, similar in shape, size, sealing, access, etc to the usual HEPA units normally installed in ventilation systems for protection against particulate discharges. The arrangement is shown diagrammatically in Figures 1 and 2. Figure 2 shows in particular the method of mounting the units in such a fashion that the unit's external seals and all associated joints in the rig could be immersed in charcoal so that any leakage gas would have to pass through charcoal. In this way a leaky seal could first be detected at an open joint, and then its effect on the DF could be eliminated by adding the charcoal covering. (The cell is now in use and its ventilation system is no longer available but plant development using modular units continues at AEEW). Several different modules have been examined. All showed a tendency to suffer from similar problems to high efficiency particulate filter modules eg liability to damage by careless handling, and manufacturing faults attributable to volume production. This type of charcoal unit would obviously require the same degree of inspection as HEPA units. After plant testing, the modules have been opened up and the charcoal removed for testing in the laboratory. One of the disadvantages of these modules is that charcoal samples cannot be taken from the unit without destroying it; for in-service testing purposes, some indirect method would have to be employed, eg by arranging for samples of the original charcoal to be exposed in the plant so that they 'see' the same conditions of staytime, etc, as the charcoal in the module. Results for three different modules are in Table 1.

6. Module A was nominally filled with 1.5% KI charcoal. On analysis it was found to have a KI content only 0.6%; this will reduce the total load of radioiodine which the unit can carry, and possibly its ageing characteristics. Test results are in Table 1. The K value found in the standard laboratory test is much lower than expected, as is the value measured at 50% RH, and suggests a poor quality, or contaminated, or aged charcoal filling. The plant tests clearly indicate some leakage: the DF at first measured is not much above the pessimistically calculated value, and is considerably lower than the expected value. Use of equation (1) suggests about 3% of flow is by-passing. Packing the external seals with charcoal to remove methyl iodide from any seal leakage gave some improvement, but there were clearly still some sources of by-passing still remaining.

7. Module B had a nominal filling of 5% TEDA charcoal. On analysis only 2.7% TEDA content was found; this discrepancy, as with module A above, would reduce the maximum load of radioiodine which the unit could carry. Test results are in Table 1. The standard test result for K at 98% RH is not unexpected, but the DF found by plant test is lower than expected and suggests about 0.15% of total flow to be by-passing. If the measured DF and estimated leakage were acceptable, the expected life of the unit could be calculated: thus if a typical minimum DF of 100 were required, then DF_0 can be allowed to fall to 120, and the expected life of the unit would be 56 weeks (at the test airflow); thus to plan to change units every year would be reasonable.

8. Module C also had a filling of 1.5% KI charcoal; test results are also in Table 1. The measured K value is very low for fresh material of this type; a measurement of K at the plant test humidity RH ~ 70% was not made, but a value was estimated, from which an expected value of plant DF was estimated. The first measurement of plant DF gave a very poor result, but packing the external seals with charcoal gave a greatly improved value. Clearly the external sealing was very poor but other sources of by-passing must be quite small. Compare the effect of similar action on the results for module A. In view of the very poor K value, a further investigation was made to see if the charcoal had been contaminated during the manufacturing process: samples of a charcoal of known K value were exposed in the manufacturer's works under typical storage conditions, and then retested, and also retested after processing, but no source of contamination could be traced.

9. General conclusions which can be drawn from these results are that it is possible to manufacture replaceable modular units of adequate performance but that care must be taken to ensure effective sealing and since the charcoal in the unit cannot be non-destructively tested it is essential that a close control be kept on charcoal quality during manufacture.

Plant No. 2

10. Plant No. 2 is a single unit serving one laboratory building; the plant is under cover inside the building. Results of tests during the last 2 years are in Table 2. The ventilation flow through the bed (charcoal volume, 2.04m^3 , or 72 cu ft) can vary between about $2.1\text{ m}^3\text{ s}^{-1}$ and $2.8\text{ m}^3\text{ s}^{-1}$ (4500 and 6000 cfm) giving a bed staytime varying between 0.72 and 0.96 secs. This can make quite a large difference to DF. By-passing usually limits the DF of this bed to about 2000. At the start of Table 2, the charcoal is coming to the end of its useful life. The predicted DF is poor, and the measured DF is well below the bypass value. A laboratory measurement was made of the K value at 98% RH of new charcoal purchased to refill the bed. The refilled bed when tested had a DF of 2600 - this is the new by-pass value; its useful life was estimated to be about 3 years. A charcoal sample taken from the bed after 7 months had a K value lower than expected, suggesting that the bed was ageing more rapidly than usual, however, the predicted minimum DF was still acceptable and expected life still about $1\frac{1}{2}$ years. A retest after 1 year gave a DF only 800, so another charcoal sample was tested. The result 6.6 s^{-1} indicates that deterioration is not now as rapid, and the bed should still be good. The poor DF may then be the result of eg settlement in the bed leaving voids and the charcoal may need to be packed down (eg by vibration) and topped up.

Plant No. 3

11. The test results for plant no. 3 are in Table 3. The plant consists of two similar units in parallel, outside the main laboratory building and exposed to the weather; they are thus subject to condensation of moisture from the warm gas stream leaving the building, in cold weather. At the start of the two year period quoted, they were in poor mechanical condition; it was suspected that rain was entering the plant; one of the beds was permanently on line because of defective isolating dampers. The charcoal was in very poor condition, being contaminated by oil, and the measured DFs were very poor. The mechanical state of the units was improved, and no. 1 unit was refilled with a charge of fresh charcoal. There was little improvement although the charcoal was clearly in good condition. On further inspection it was found that the filling access arrangements were inadequate and the filling was very uneven. The beds had to be filled by pouring in charcoal through a few access holes at the top of the bed,

leading to uneven filling; the upper surface of the charcoal tended to the form of a few large heaps immediately below the access holes with voidage in the gaps between. The need was for more access holes for pouring, so that the upper levels of the bed were more evenly filled. The beds were thus modified to improve access, and the charcoal repacked evenly and topped up. The effects are clearly seen. In the meantime no. 2 unit had been attended to and similarly improved, and the units can now be run with one on line, one on standby. Because of its earlier history, the plant is being tested every three months, but because results are near to the limit of sensitivity for this plant no significance can yet be attributed to the changes in DF found. Charcoal samples from unit no. 1 after 6 months suggest that deterioration is taking place at the entry face.

Plant No. 4

12. Plant no. 4 consists of a group of similar units, each serving a separate facility in the laboratory building and all under cover inside the building. The nominal plant staytime is 0.375 s and the absorber beds are on the downstream side of the fans for reasons of space and construction convenience. They are thus not only under pressure so that any seal leakage is outwards to atmosphere, which is bad safety design, but any oil leakage from fan bearings is likely to contaminate the charcoal. All units in the group normally behave similarly. The charcoal is renewed annually as part of regular planned maintenance. Charcoal samples taken during and at the end of the life of the bed suggest that the charcoal in these units is consistently deteriorating more rapidly than it should, and that the probable cause is the absorption of oil which is blinding the available surface area. A likely source is the fan bearings. Redesign of this plant is under consideration.

Plant No. 5

13. This plant is at AEEW, and is being used for development of plant using modular units. The plant layout is shown in Figures 3 and 4; the original iodine fission product trap comprises a steel canister containing alternate layers of perforated metal, copper gauze, and beds of impregnated charcoal granules. The flow through the charcoal is upwards, and the packing fraction can vary considerably on filling, so that fluidisation of the beds may occur with consequent channelling of the air stream and loss of granules into the exit stream. Laboratory tests have shown that DFs of up to 10^7 should be achievable, but in practice plant DFs of only up to 10^4 have been obtained.

14. Current development on this plant is the substitution of modular units of type C above (paragraph 8) for the existing traps. Table 5 gives the results of an experiment performed on the plant to compare the efficiency of the existing traps with that of a modular unit installation. There are three similar extract ducts as shown typically in Figure 3, and in one, duct 'a', an assembly of modular units was substituted for the existing traps (Figure 5). The remaining two ducts 'b' and 'c', were unmodified. The duct flows were balanced to take equal volumes of air from the hot cell. Pollack counter tests to detect major leaks showed particulate penetrations of less than 1% for each trap. The test was done at relative humidity 65% at 21°C.

15. Although the efficiency of the installed charcoal beds is well within the operating limit of 1% penetration the 228 mm and 292 mm water gauge pressure drops are not acceptable and suggest a high packing fraction. From past experience, more loosely packed charcoal granules in this design of bed are liable to fluidise and ultimately join the extract air stream. Plenty of margin exists in the prototype assembly for an increase in bed depth, volume and pressure drop to produce a staytime of 1.5 to 2.0 secs, which would clearly give a far improved efficiency. Development of the prototype modular assembly will continue.

Discussion

16. The results for the in-service plants nos 2, 3 and 4 illustrate some of the possibilities and essential requirements for the maintenance of adequate trapping of iodine in the ventilation systems of hot laboratories. The normal installation is some form of absorption bed filled with an impregnated charcoal, a 50/50 mixture of 1.5% KI and 5% TEDA impregnated coal-based charcoals is usual at Windscale. Both regular testing of the plant performance under normal conditions and the extraction and testing in the laboratory of samples of the charcoal are necessary, and good maintenance and care on the part of the operator is also required. The operator is in a difficulty when a poor result is due to some fault in the bed rather than the charcoal, work in the laboratory must be maintained, and a bed with a poor but just adequate DF is better than no bed at all, so that the search for faults and improvement of the bed, as in plant no. 3 may take months of work done when the opportunity allows. Consistent results must also be obtained from the tests otherwise the operator can be given little in the way of confident guidance; plant no. 2 is not as consistent as nos 3 and 4 in this respect. However, it is clear that good performance can eventually be got from plant initially in very poor condition. The results for plant no. 4 indicate that it is not enough to test a newly reconditioned plant, obtain a good result, and then forget it; standard ageing formulae which have been derived using normally clean laboratory air must be shown to apply to the plant in question by charcoal sample testing during the life of the plant, or misleading beliefs about the life of plant may be held. A contaminant in the ventilation air may accelerate deterioration of the charcoal, and an operator's only recourse may be to renew charcoal more frequently. In this event, the readily changeable modular units may be advantageous, but in their turn they clearly have their own troubles - poor sealing and charcoal filling of unknown quality are but two which have been found during the course of the investigative work at Windscale.

Conclusions

14. The outlined methods of investigating plant performance combine the full scale, in situ testing of plant with methyl iodide-131 and the testing in the laboratory under standard conditions of samples of absorber material (impregnated charcoals) taken from the plant. Recent work indicates that in general plant performance can be maintained at satisfactory levels, but examples of significant bypass leakage and unusually rapid charcoal deterioration have been detected, and continued vigilance is necessary for the maintenance of adequate performance. Studies of modular units indicate that they, like the refillable bed plants, have their own disadvantages, which include liability to faulty external sealing,

filling with charcoal of unknown and possibly poor quality, contamination of the charcoal during manufacture of the unit, and the impossibility of non-destructive testing of the charcoal during the life of the plant.

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TABLE 1

Test results, plant no. 1

Module	Laboratory tests			Plant tests					
	Result ref.	RH %	K (s-1)	Result ref	RH %	Flow m ³ /sec	DF found (=DF _P)	DF _B (expected)	DF _C
1A	1A.1	98%	5.5	1A.3	53	0.157	19.3	41.6	12.1
	1A.2	50%	8.2	1A.4	51	0.157	24.8		
				1A.5	52	0.156	24.8		
1B	1B.1	98%	13.5	1B.3	50	.137	600	1.4 x 10 ⁶	3.1 x 10 ⁴
	1B.2	50%	18.5	1B.4	53	.138	607		
				1B.5	53	.139	597		
1C	1C.1	98%	3.8	1C.2	79	0.132	4	(440)	14
		(70%)	(8.3)	1C.3	74	0.116	290		
				1C.4	74	0.113	145		
				1C.5	72	0.115	490		
				1C.6	76	0.115	270		

The values DF_B (expected) are calculated from the laboratory test results at low RH.
 The values DF_C are calculated from the laboratory test results for 98% RH.

TABLE 2

Test results, plant no. 2

Event No	K (98% RH) (s ⁻¹)	Plant staytime (s)	Plant DF _p	Predicted DF _c	Comment
2.1	(3.5)	(.77)	615	495	} Absorber at end of useful life, and due for change.
2.2	(3.0)	.77	730	204	
2.3	11.8				New charcoal for bed refilling.
2.4			2600	1.2×10^9	Refilled bed.
2.5	6.9	(.77)		2×10^5	Sample from bed after 7 months.
2.6			800		Test after 1 year.
2.7	6.6			1.2×10^5	Sample from bed after 13 months.

Bracketted values of K are calculated using assumed ageing characteristics.

Bracketted values of staytime are calculated using assumed flow rates.

TABLE 3

Test results, Plant No. 3

Event No.	K (98% RH) (s-1)	Plant staytime (s)	Plant DF _P	Predicted DF _C	Comment
3.1			53 59.5		No. 1 bed. Both beds in parallel.
3.2	1.2 1.9				Oil content 3.5% } Tests on Oil content 1.3% } samples of } bed charcoal
3.3	12.3	(0.7)		4.1×10^8	New charcoal for refilling of units.
3.4			76		No. 1, after refilling.
3.5			2100		No. 1, after improvements and topping up.
3.6	10.4 7.0	(0.7)		1.9×10^7 7.9×10^4	Sample after 6 months, from bed exit. Sample after 6 months, from bed entry.
3.7			1550 $\downarrow 1.5 \times 10^4$		No. 1 unit after 6 months. No. 2 unit after attention. Limit of sensitivity value.
3.8			$\downarrow 8000$ $\downarrow 1.7 \times 10^4$		No. 1 unit } after further 3 } months No. 2 unit }
3.9			7500 1.2×10^4		No. 1 unit } after further 3 No. 2 unit } months

TABLE 4

Test results, Plant No. 4

Event No.	K (98% RH) (s ⁻¹)	Plant staytime (s)	Plant DF _P	Predicted DF _C	Comment
4.1	9.29				New charcoal.
4.2			6.7×10^3		Typical value after 6 months service.
4.3	3.1				Typical value for sample after 1 year life. 4.8 s ⁻¹ expected.
4.4	11.5	(.375)		2×10^4	New charcoal for fresh charge.
4.5			4.0×10^4		Typical value for newly refilled bed.
4.6	3.3	(.375)		17	Typical value of sample after 9 months service. 7.0 s ⁻¹ expected. Oil content 1.9% typical.
4.7	2.8	(.375)		11	Typical value after 11 months service 2.6 s ⁻¹ expected from event 4.6.
4.8	10.7				New charcoal obtained for annual refill.

TABLE 5

Comparative tests on installed conventional and modular trapping plant
(plant 5)

Duct	Flow	Pressure drop (mm water)	Staytime (secs)	Penetration (%)	DF	$K = \log DF / \text{staytime}$ (s-1)
a	400	10.2	0.7	1.1	96	2.9
b	400	292	2.2	0.1	1017	1.4
c	400	228	2.2	0.3	318	1.15

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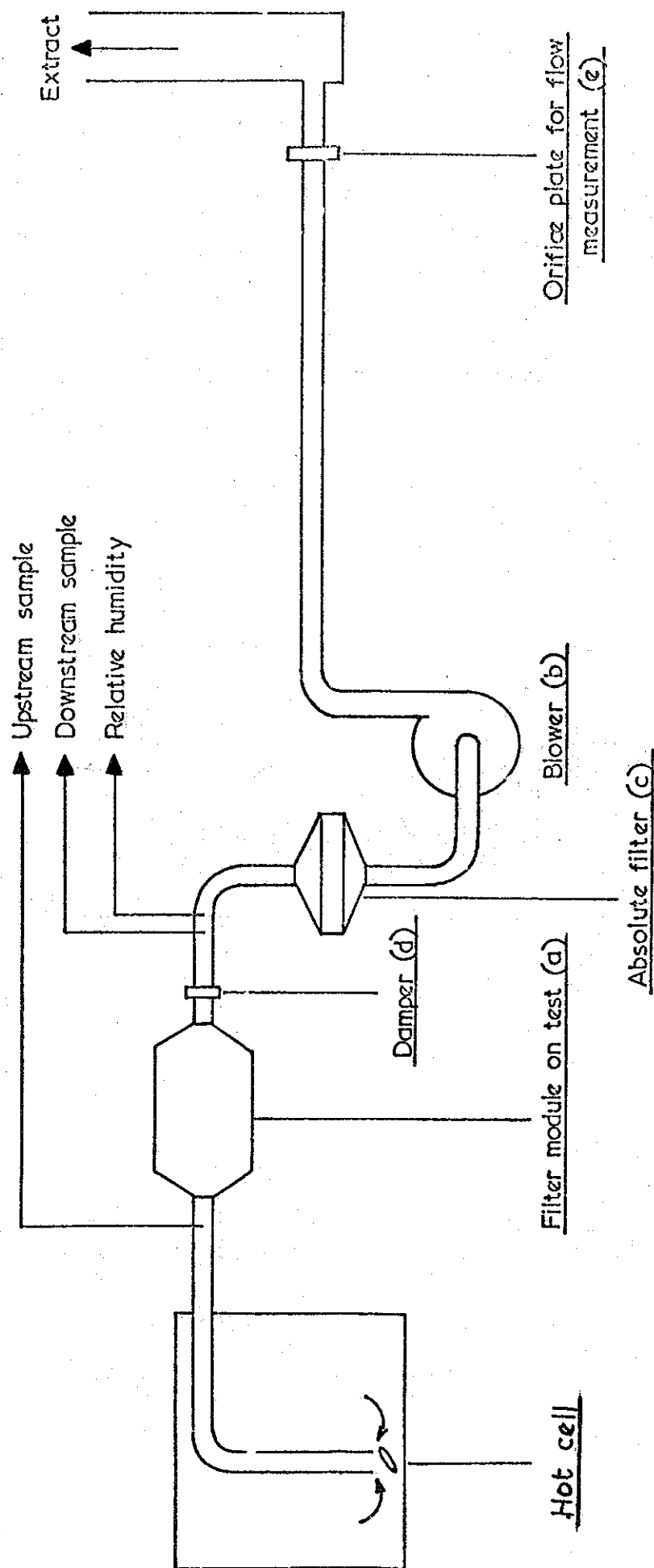


Fig. 1. PLANT USED TO TEST CHARCOAL FILTER MODULES.

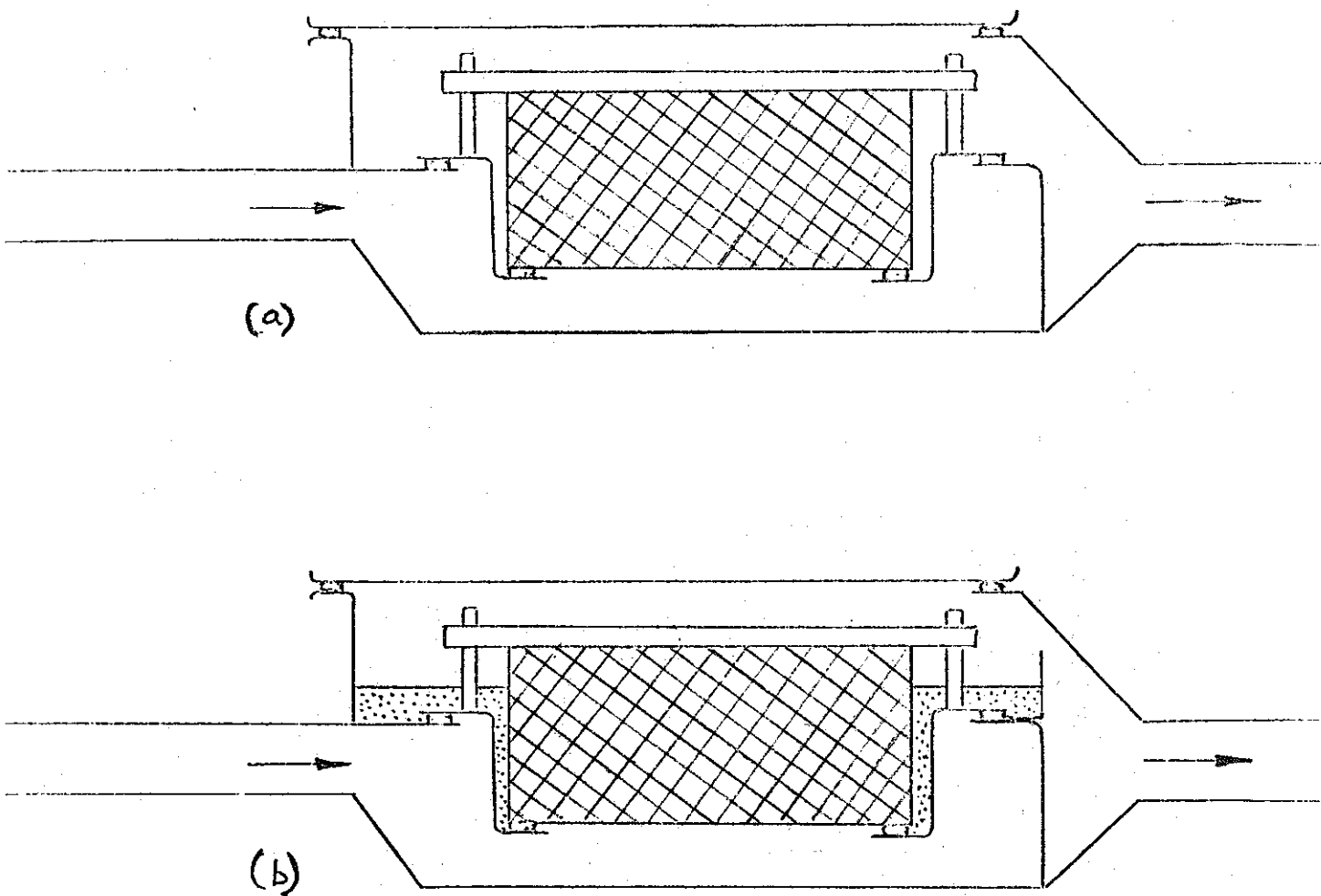


FIG. 2 Arrangement of charcoal filter in housing for methyl iodide penetration test.

(a) original arrangement,

(b) after packing seals with charcoal.

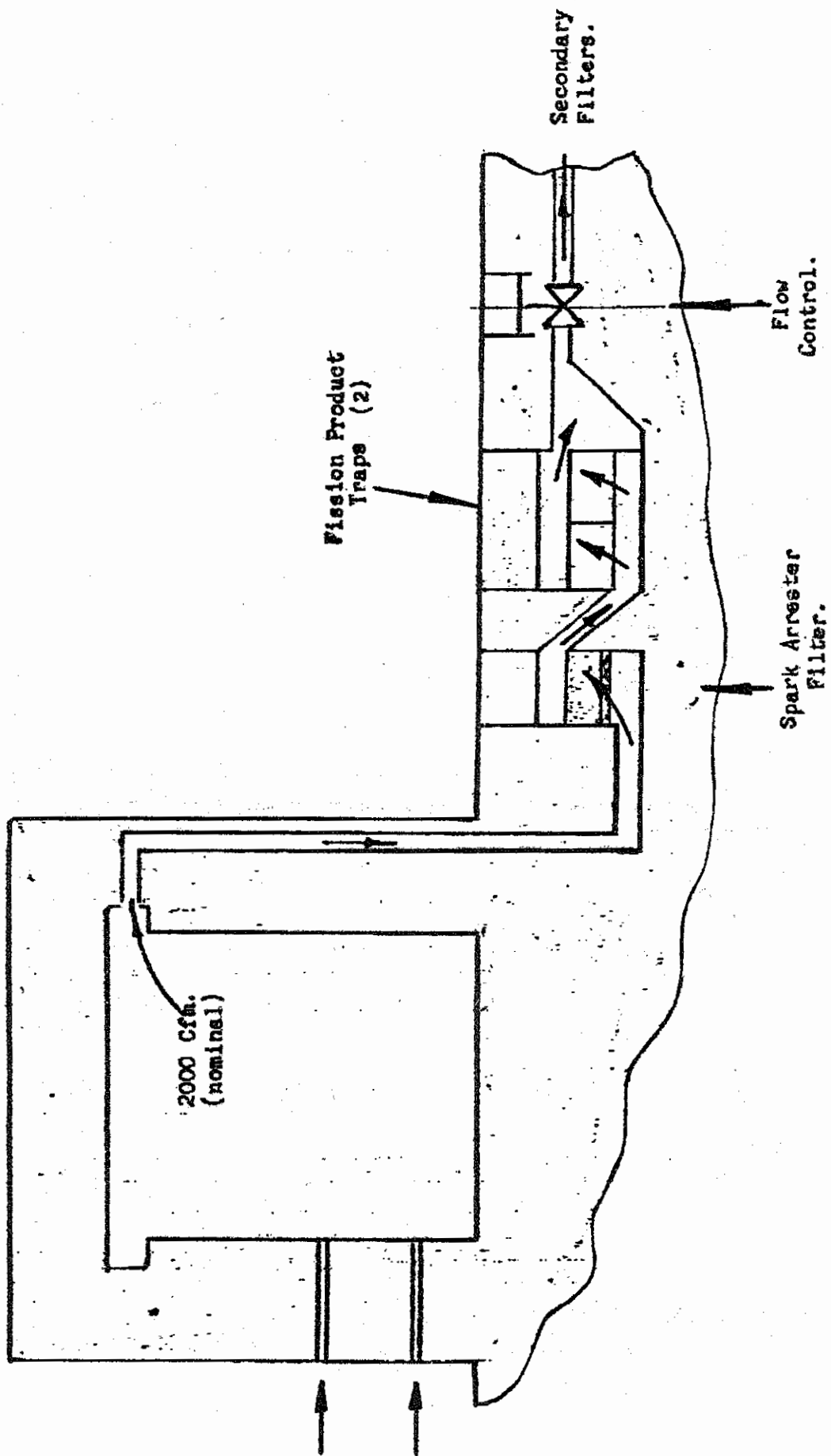


Fig 3. CAVE EXTRACT - Typical Section thro' extract duct

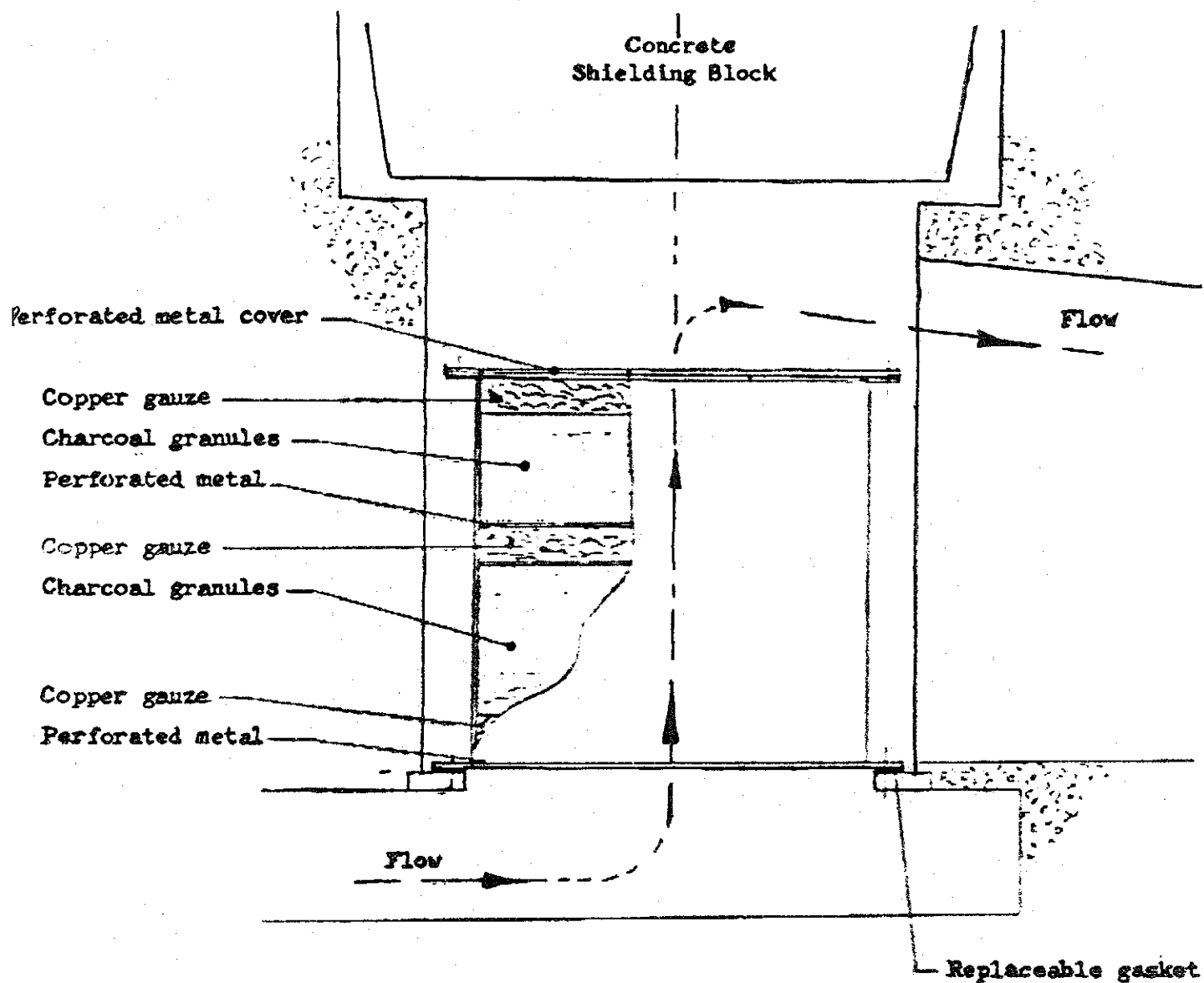


Fig. 4 Fission Product Trap - Existing Installation

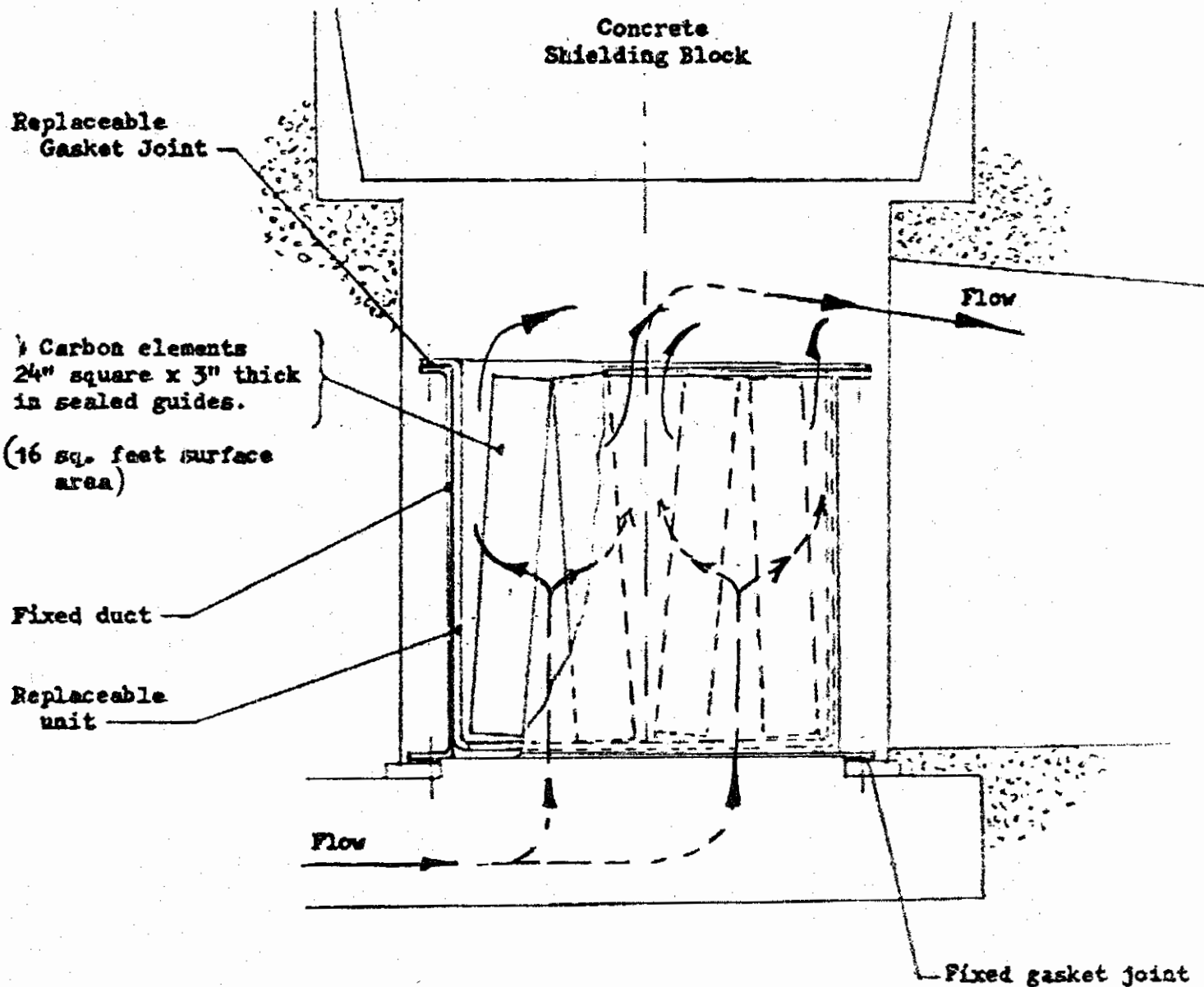


Fig. 5 Fission Product Trap - Proposed replaceable element

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