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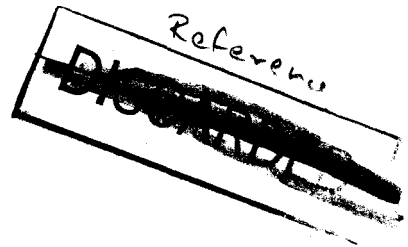
United Kingdom Atomic Energy Authority

EXPERIENCE IN TESTING INSTALLED FISSION PRODUCT TRAPPING PLANT WITH METHYL IODIDE

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EXPERIENCE IN TESTING INSTALLED FISSION
PRODUCT TRAPPING PLANT WITH METHYL IODIDE

by

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SUMMARY

A simple experimental procedure has been developed, based on labelled methyl iodide, for the full-scale in-site testing of the charcoal bed of an installed fission product trapping plant. A decontamination factor of 1000 is easily measurable for a 10,000 ft³/min. plant with a few mc Iodine-131.

Tests carried out to date on several plants indicate the need for such tests since the results have in some cases shown inadequate performance, which has usually been associated with mechanical leakage.

The most effective method of achieving reliable sealing appears to be to pass gas downwards through a bed of charcoal which completely fills the available area, rather than to attempt to seal an assembly holding a bed into a flange in the duct or vessel. Provision should be made for withdrawing representative samples of charcoal for laboratory testing.

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INTRODUCTION

1. Iodine-131 is generally recognised to be the most hazardous fission product resulting from possible reactor accidents, and an attempt was made to develop a technique for full-scale testing of installed fission product trapping plant based on passing labelled elemental iodine over hot reactor fuel components, to produce a proportion of other forms, e.g. aerosols. However, there was only a small yield of aerosols and the elemental iodine plated out so readily in the steel pipework that little of the original source reached the plant being tested. The only useful result from this early work (Table II, Test 1) was produced by an experiment with carrier-free iodine in which a sufficient proportion was converted to a more stable, penetrating form of iodine, now thought to have been methyl iodide.

2. Since then, methyl iodide has been identified as a component in many iodine releases⁽¹⁾ and, being at the same time both relatively stable and probably the most penetrating species in these iodine releases, appeared to form the basis for a satisfactory full-scale charcoal bed testing technique. Further, it has also proved necessary to develop special charcoals to deal with methyl iodide both at large⁽²⁾ gas loadings in dry gas⁽³⁾ and under conditions of high relative humidity⁽⁴⁾.

3. This paper describes a simple test which has been developed, based on labelled methyl iodide, which is readily applicable on a large scale (even at high pressure), and the results of a number of such tests which have been conducted on installed charcoal bed plant.

EXPERIMENTAL

4. Labelled methyl iodide is prepared by bubbling argon (or helium) through a U-tube containing labelled aqueous Sodium Iodide-131 or -132 in one limb and dimethyl sulphate in the other (Fig. 1). The methyl iodide (b.p. 42°C at 760 mm) volatilises readily, is dried and then condensed by cooling into a suitable glass container which is finally sealed off. Methyl iodide is occasionally confirmed quantitatively by passing the effluent carrier gas without condensation to a column of dinonyl phthalate or silicone oil on celite and carrying out conventional elution gas chromatography. The effect of the small amount of dimethyl sulphate carry over which might poison the charcoal has been shown to be negligible.

5. The ampoule containing methyl iodide, normally several mg and up to 10 mc ¹³¹I is then transferred to a point upstream of the plant to be tested and broken. It volatilises quickly and completely without heating in the gas stream leading to the plant. If the plant to be tested is in the extract of a shielded cave, remote fracturing arrangements are available. Again if the plant is in the extract from the containment of a reactor it may be more convenient to break the ampoule inside a ventilated glove-box and lead the methyl iodide into the extract by suction or applying a gas flow at slightly elevated pressure. The release of radioactive iodine is usually monitored by observing the fall in reading of a γ -monitor in close proximity to the source.

6. If the plant operates at high pressure the type of steel rig sketched in Fig. 2 can be used. After breaking the ampoule by rotation of a valve, the methyl iodide is displaced by purging with gas at higher pressure, adequate safety precautions being taken to minimise the escape of active high pressure gas which would result from failure of components or from maloperation.

7. Sampling procedures normally consist of separately withdrawing a known small proportion of the gas from upstream and downstream of the charcoal beds under test, and passing it through separate May Pack samplers, the essential component of which is the charcoal.

8. The test is conducted in two parts. The initial few minutes gives an indication of leakage paths in the bed (since even charcoal not specifically designed to trap methyl iodide in moist conditions will hold it up for a time by physical adsorption). Subsequent sampling tests the ability of the charcoal to retain methyl iodide. An aliquot of the source and sampler components are analysed by standard γ -counting techniques.

9. The stay-time in the charcoal sampler is usually about $\frac{1}{2}$ sec (200 ml charcoal, 20 l/min) to ensure efficient retention of methyl iodide. (Improved sensitivity could be achieved by reducing the stay-time or the same sensitivity could be obtained with lower source strength). Impregnated charcoal⁽²⁾ of small mesh size is also normally used in the samplers (-13 +52, compared with -8 +12 in the plants tested) for improved trapping efficiency. Occasionally a section of clean copper knitmesh has been introduced in the inlet sample upstream of the charcoal to show that no elemental iodine was present in the source. The appendix deals with the experimental procedure in a typical test. The mass of methyl iodide (a few mg) was chosen so that it was large enough for reproducible behaviour during preparation and also gave a negligibly small charcoal loading.⁽²⁾

RESULTS AND DISCUSSION

10. Table II gives the main results of the tests. It is clear from these results that the performance has been erratic, poor performance has been due to both the type of charcoal used and to mechanical defects with consequent passage of gas through the plant without contacting charcoal. These points are best illustrated by reference to one plant only (cave extract plant (1) Table II, which takes mainly horizontal but some upwards air flow) as in the following table:

TABLE I

Full-scale tests of an installed charcoal bed with methyl iodide

Test no.	Time after initial installation months	Methyl iodide penetration, % (period of test, h)	Remarks/Events
7	7	40 (0.5h) 100 (50 h)	Filled with untreated coconut charcoal
8	8	11 (70 h)	Repeat of test 7 coconut charcoal
9	12	0.02 (20h)	Charcoal replaced by KI/TEDA impreg. coal based variety
10	19	9 (0.5h) 11 (43h)	Repeat of test 9
11	22	<0.05 (0.5h) <0.15 (26 h)	Charcoal taking upwards gas flow removed and section blanked off
12	23	0.1 (9)	Repeat of test 11

11. The large rapid penetration of the bed in test 7, which was probably due to a large leakage path was not confirmed in the repeat, test 8, possibly because of varying distribution of charcoal in the trays of the bed allowing gas to by-pass charcoal in an erratic manner. The result of test 8 represents approximately the penetration expected by slow release of methyl iodide from the unimpregnated charcoal by a physical adsorption-desorption process and does not necessarily indicate a major leakage path.

12. The original coconut charcoal was replaced by a specially developed charcoal (impregnated coal based type, mixed KI/TEDA impregnation⁽²⁾), the sealing arrangements were improved, and the steel wire gauze which held down that part of the charcoal bed which took upwards flow was stiffened and spring loaded. The subsequent test (no. 9) showed satisfactory performance but a repeat (test 10) showed an unacceptably large penetration in a short time. A small representative sample of the main charcoal bed was next removed and shown to have satisfactory performance in a standard methyl iodide laboratory test but it was then found that a seal had failed and a substantial fraction of the charcoal taking upwards flow had been blown out. The defective section of this charcoal bed has since been effectively blanked off and further tests (nos 11, 12) have given satisfactory results.

13. The bulk density of this type of charcoal (-8 +12 mesh) is about 0.45 g/cm^3 so an upwards flow equivalent to 0.45 in. w.g. per in. deep charcoal is required to balance the gravitational forces. Taking the design conditions, i.e. a face velocity of 63 ft/min, the data in Fig. 3, obtained from laboratory measurements at Windscale, indicates an expected pressure drop across the charcoal of 0.6 in. w.g./in. deep of charcoal. These simple considerations imply that there was a tendency for upwards bulk movement to occur. Further, other Windscale laboratory experiments in 1 in. dia. glassware have indicated that movement of -8 + 12 mesh charcoal granules is just observable under these flow conditions. It is also possible that abnormally high flow conditions may have been produced either by movement of charcoal granules resulting in the development of shallower portions in the bed giving higher local gas flow (due to lower pressure drop), or by removal of some of the resistance to flow in another part of the plant (for instance by opening cave access doors).

14. This postulation suggests that if upwards flow must be accepted due to particular circumstances than an allowance must be made so that the charcoal cannot fluidise under the most adverse conditions envisaged.

15. The tests on the reactor coolant discharge plant (Table II) have shown satisfactory performance both in air and carbon dioxide after loading impregnated charcoal while test 2 illustrates the gradual penetration of the untreated coconut charcoal. However, a pre-commissioning inspection of the reserve plant (also horizontal flow) showed that charcoal granules had settled below the level of the baffle plate; this was probably due to inadequate filling as well as subsequent settling and points to the need for careful supervision during installation.

16. The void ventilation plant was satisfactory whether or not the charcoal was impregnated because the high operating temperature (50°C) resulted in low relative humidity. The only plant with wholly downwards flow through the charcoal was in the extract cave 2 and this showed consistently good performance.

17. The plant in the extract from Cave 3 was found on inspection to have large leakage paths in upwards and downwards flowing sections, which accounts for the high penetration observed in tests 17, 18. In this case, where charcoal granules had been observed to have moved away from the edges of the trays to leave gaps, the height:diameter ratio of each tray was small. This feature made it difficult to prevent movement of charcoal by holding it down under a gauze and appears to have allowed vibration inspired movement of the charcoal granules away from the edges. A similar phenomenon can be observed during a simple hand sieving operation.

18. A suggested solution is to increase the height:diameter ratio substantially (e.g. by adopting an 'egg-box' arrangement), and thereby minimise the reduction in bed depth near the edge due to the tendency for the upper surface of the charcoal to assume a characteristic angle of repose. In addition the perforated horizontal section of the tray which supports the charcoal could be provided with a solid flange round the edge so that the shallower section of the charcoal bed, i.e. at the edges, would not then take any direct flow of gas.

CONCLUSIONS

19. A simple technique has been developed for the in-situ testing of charcoal beds of installed fission product trapping plant, and involves the release of labelled methyl iodide upstream of the plant. Little additional plant is required.

20. The test measures mechanical leakage as well as charcoal retention characteristics and results in negligibly small poisoning of the charcoal.

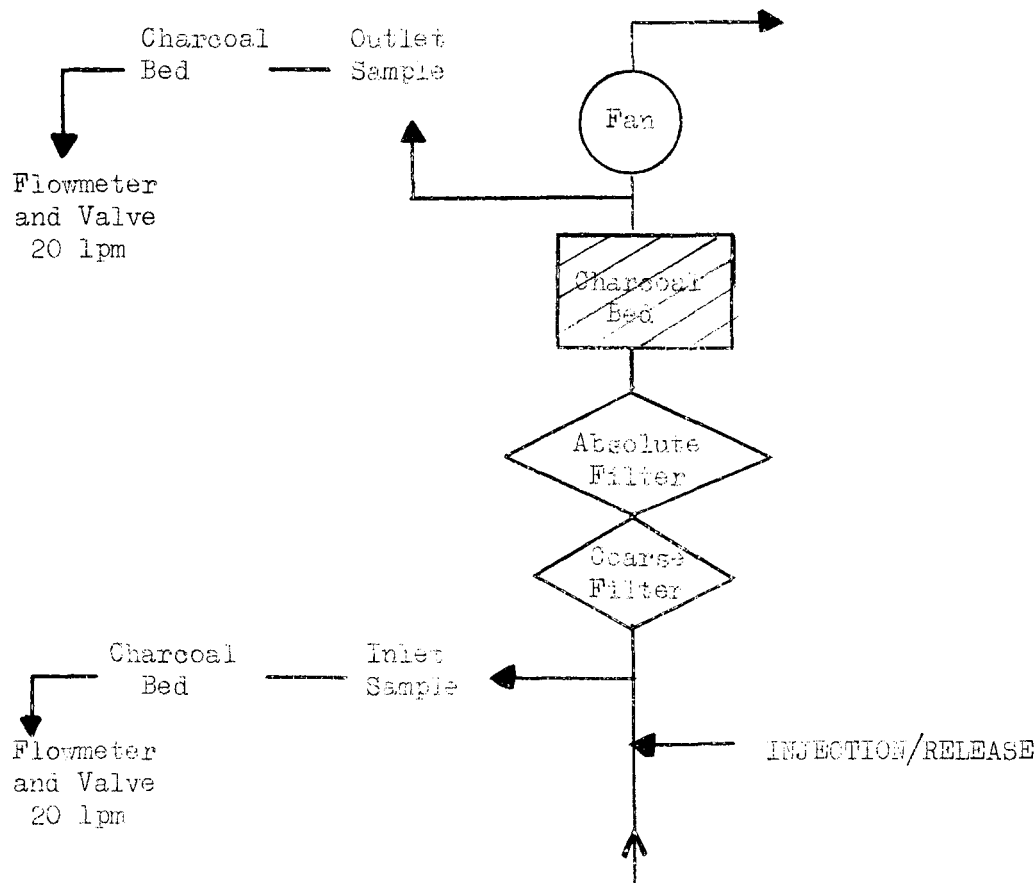
21. Experience with tests carried out on a number of plants has shown that to achieve good performance of charcoal beds, careful attention should be paid to direction of flow, which should be downwards, sealing methods and bed filling arrangements, as well as to the choice of a properly developed charcoal. The possibility of bed movement under a combination of gas forces and vibration must also be considered.

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APPENDIX

Typical procedure for testing in an installed charcoal bed



<u>Time (min)</u>	<u>Procedure</u>	
-ve	Set up flow through main charcoal bed and start both sampling flows.	
0	Release MeI source by breaking. Check γ -radiation.	
30	Stop inlet sample Change outlet sample	INLET SAMPLE OUTLET SAMPLE (1)
120	Change outlet sample	OUTLET SAMPLE (2)
Next day	Change outlet sample	OUTLET SAMPLE (3)

Note: If the source is released into say the large inner containment volume of a reactor sufficient time should be allowed during the first sampling period for the majority of the methyl iodide to reach the plant under test.

TABLE II

Methyl iodide tests of full scale charcoal bed plant results

Test No.	Plant Designation	Charcoal bed data			Flow conditions (as tested)			Duration of test	% Penetration	Remarks
		Type	Volume ft ³	Surface area, ft ²	Direction	Gas composition	Gas staytime sec.			
1	Reactor Coolant discharge plant	Coconut	70	70	Horizontal	Dry CO ₂	~11	1.0	0.1	
2	"	Coconut	70	70	"	Undried air	1.3	16.0	3.1	
3	"	Coal based, KI and TEDA impregnated (mixed 50 50)	70	70	"	Undried air	1.3	-	<4.0	Charcoal changed
4	"	"	70	70	"	Dry CO ₂	~1.0	1.0	0.1	
5	Reactor void ventilation plant	Coal based, (unimpreg.)	51	77	"	Undried air + little CO ₂	0.75	70.0	<0.14	Low humidity
6	"	Coal based, KI and TEDA impregnated	51	77	"	"	0.75	-18.0	<0.1	
7	Cave extract plant (1)	Coconut	76	144	Horizontal and upwards	Undried air	0.5	0.5 50.0	40.0 ~100.0	
8	"	Coconut	76	144	"	"	0.5	70.0	11.0	
9	"	Coal based, KI and TEDA impregnated	76	144	"	"	0.5	20.0	0.02	Charcoal changed
10	"	"	76	144	"	"	0.5	0.5 43.0	9.0 11.0	
11	"	"	64	144	"	"	0.4	0.5 26.0	<0.5 <0.16	Upward flow part of bed blanked off
12	"	"	64	144	"	"	0.4	9.0	0.1	
13	Cave extract plant (2)	Coal based piperazine impregnated	6	4	Downwards	"	0.8	48.0	<0.05	
14	"	"	6	4	"	"	0.8	0.5 2.5	<0.02 <0.6	
15	"	"	6	4	"	"	0.8	6.3 11.0	0.006 0.07	
16	"	"	6	4	"	"	0.8	0.5 10.0	0.003 0.02	
17	Cave extract plant (3)	Coal based, KI and TEDA impregnated	48	96	Upwards and downwards	"	1.0	0.5 6.0	100.0 100.0	
18	"	"	48	96	"	"	1.0	0.5 6.0	~100.0 ~100.0	

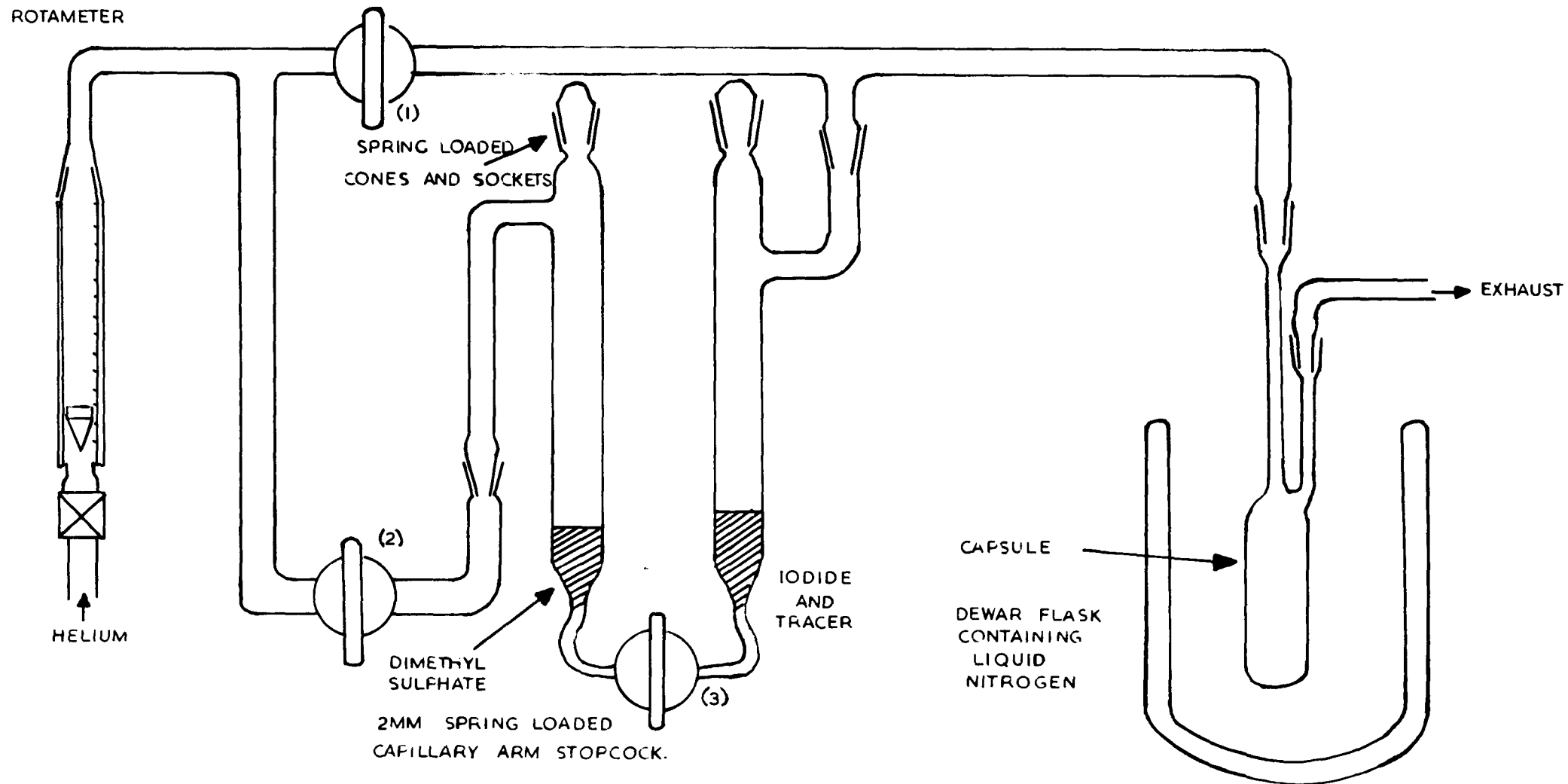


FIG. 1. METHYL IODIDE ENCAPSULATION APPARATUS.

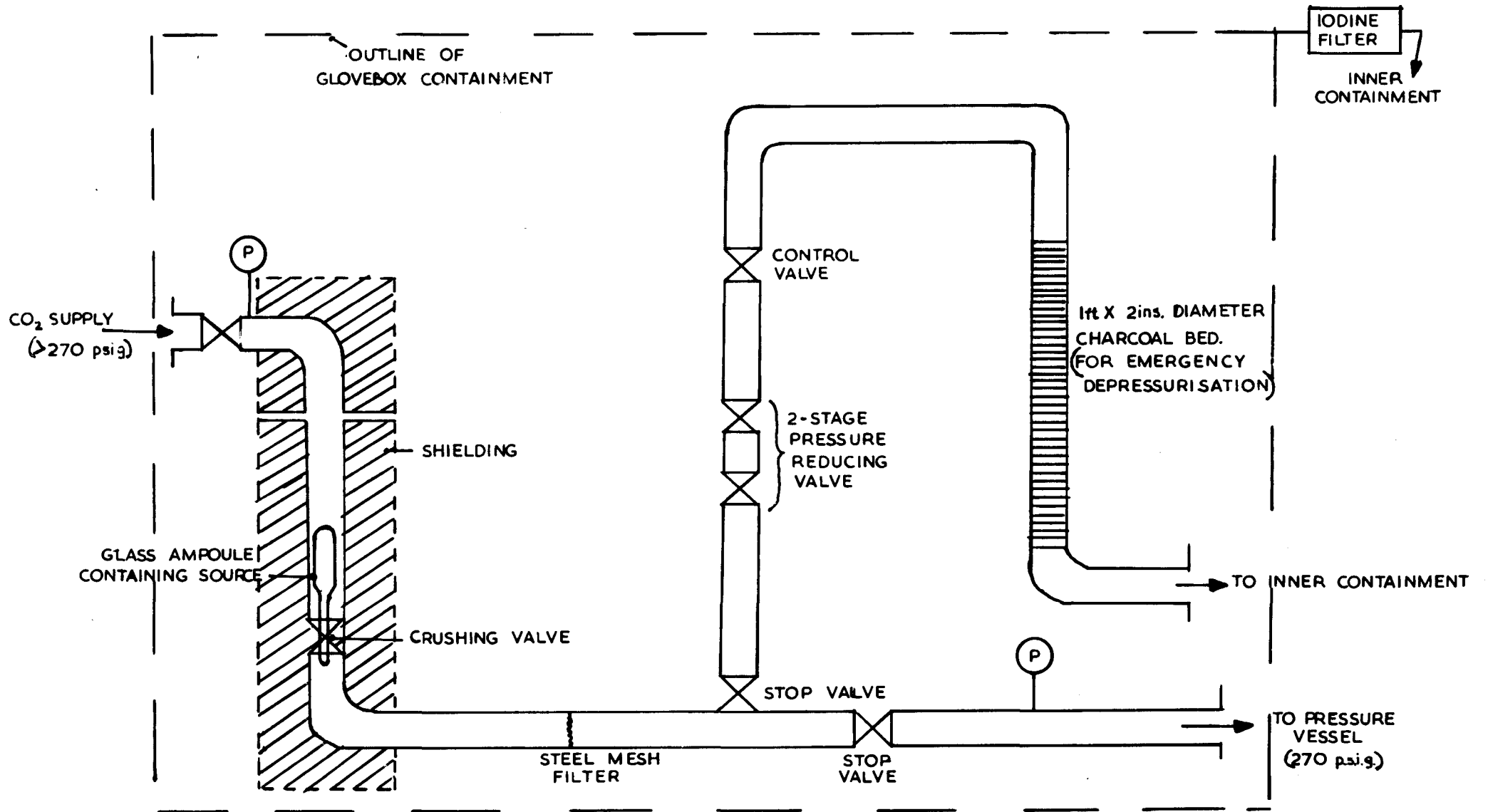


FIG. 2. METHYL IODIDE INJECTION APPARATUS. (HIGH PRESSURE)

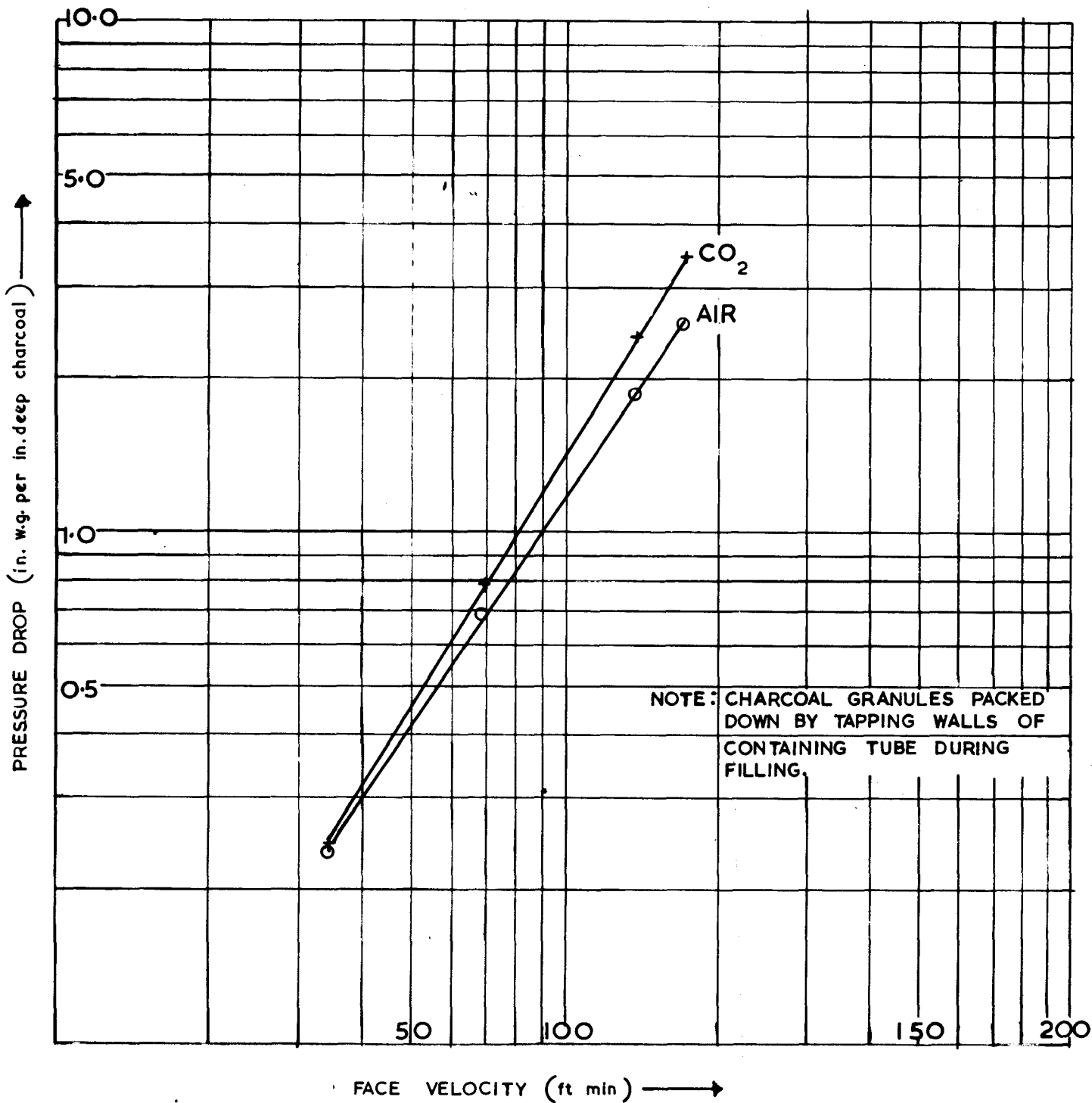


FIG.3. PRESSURE DROP vs. FACE VELOCITY FOR -8+12 MESH GRANULATED CHARCOAL (1in. DIAMETER BED, DOWNWARDS FLOW 760mm. PRESS., 20°C.)