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SUBJECT: Fission Gas Holdup Tests on HRT Charcoal Beds

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Abstract

Fission gas holdup tests on the HRT charcoal beds under simulated operating conditions have been completed. A radioactive tracer technique developed for use in the laboratory adsorption study was utilized in these tests. The efficiency of the charcoal beds, in regard to holdup of fission gases, exceeds design specifications. On the basis of these tests, the charcoal beds should perform satisfactorily with the HRT operating at 10 MW with a total oxygen flow of 3 liters/min. or at 5 MW with a total oxygen flow of 3.5 liters/min., assuming that the maximum charcoal temperature in the first sections of the bed does not exceed 100°C and that the temperature in the 6 inch diameter section is in the 15° - 20°C range.

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I. Introduction

An important phase in the operation of a homogeneous reactor is the removal and disposal of the rare gas fission products. The activity of these gases will be such that discharging them directly into the atmosphere would produce a biological hazard. Disposal of these gases from the HRT without excessive contamination of the atmosphere will be accomplished with large charcoal adsorption beds. An oxygen stream, containing the fission gases (krypton and xenon isotopes), is removed from the reactor and passed into the charcoal beds. Gases leaving the charcoal bed are then released into the atmosphere through a stack. The fission gases are delayed in their passage through the charcoal bed by the process of adsorption-desorption on the charcoal surface. This holdup time allows the short-lived isotopes of krypton and xenon to decay to a very small fraction of their original concentration. If the charcoal bed is adequate for a particular reactor then the short-lived gaseous isotopes will decay almost completely before passing through an appreciable length of the bed. Long-lived isotopes are expected to pass through the charcoal bed without appreciable decay. Ideally, for an optimum sized charcoal bed, only krypton-85 (10.3 year) will pass through the bed. The amount of krypton-85 coming from the HRT operating at 10 MW will be less than 10% of the allowable activity discharge rate of 80 curies/day (10).

II. Design of the HRT Charcoal Beds

The essential design criteria and specifications for the HRT charcoal beds have been reported (5). The adsorption system was designed to process 500 cc/min of gases composed of injected oxygen and gaseous isotopes produced in the reactor at a 10 MW power level. This gas stream is then divided and passed through two parallel charcoal beds. Each bed consists of 40 feet of 1/2" pipe, 40 feet of 1" pipe, 40 feet of 2" pipe, and 60 feet of 6" pipe, containing approximately 520 pounds of activated charcoal. The beds are mounted in a horizontal position and placed in a water-filled pit. During operation, the charcoal in the beds will be heated by decay of the short-lived isotopes and cooling is necessary to prevent excessive oxidation of the charcoal. Thermocouples are installed at various points in the charcoal mass to monitor temperature during operation. The design allows a temperature not exceeding 100°C in the inlet of the bed. At the time of design only scanty experimental data were available on the adsorption of krypton and xenon by charcoal. Therefore, a safety factor of approximately 5 was included in the design. Later HRT operating experience indicated that the design gas flow rate of 250 cc/min through each charcoal bed must be exceeded to maintain required oxygen concentration in the fuel solution and that oxygen flows in the neighborhood of 1500 cc/min per bed may be expected. This fact in conjunction with experimental data on packing effects in horizontally mounted charcoal beds indicated the need of a dynamic holdup test on the HRT charcoal beds before the HRT reactor start-up.

III. Dynamic Holdup Tests

A radioactive tracer technique developed for use in laboratory adsorption studies was utilized for these tests (2). A pulse of krypton-85 (25 millicuries) was injected at the entrance of one of the beds and then swept through the bed by a measured stream of oxygen. Krypton-85 activity in the gas stream leaving the bed was then measured. The charcoal beds had been swept with dry oxygen for several days prior to introducing the radiokrypton. This test simulates the conditions that will be present during reactor operation with the exception of the decay heating and the concentration of rare gas fission products. The heating effect will tend to reduce the overall efficiency of the beds.

A graph of activity in the effluent gases versus time after injection of the radiokrypton (elution curve) for the four existing charcoal beds is given in Fig. 1. The holdup performances of the beds are listed in Table I. For comparison, experimental t_{max} values, obtained from the tests normalized to a hypothetical bed containing 500 pounds of charcoal, 1 liter/min. oxygen sweep, 15°C, are also listed.

IV. Discussion of Results of Holdup Tests

The holdup performance of the beds was better than anticipated, based on data from small laboratory charcoal traps (2, 6, 7). Values for N (number of the theoretical chambers in a bed) were smaller than expected. The shape of the elution curve governs the value of N. For narrow, sharply rising elution curves the N value is large; for broad, slowly rising curves the N value is small. N values approaching 10^3 were expected for a bed the length of the HRT charcoal beds, whereas values in the 150-400 range were determined. A consequence of a low N value is that the interval between breakthrough time (t_b) and time of maximum concentration of activity in the effluent (t_{max}) is large. In designing a charcoal bed it is assumed that most of the undecayed fission gases will emerge from the bed at or slightly prior to t_{max}. If the interval between t_b and t_{max} is large, then the effective decay time of the short-lived gases is less and some of these gases may pass through the bed and emerge prior to t_{max} in concentrations greater than anticipated.

The low N values determined for the HRT beds may be the result of one of the following factors. The presence of horizontal void spaces or channels in the HRT beds would cause a broadening of the elution curve by allowing a portion of the fission gases to bypass some of the charcoal mass. An effect of this sort has been demonstrated experimentally (6). The rate of flow of sweep gas through the 6" section of the HRT beds was quite low, i. e., 0.18 feet/min, while laboratory traps were studied at linear velocities of 3.5 feet/min. At low gas velocities it has been noted that longitudinal diffusion of the adsorbable gas in the gas phase will effectively reduce the value of N (3, 8). It would be impossible to state the exact cause of the low N values determined for the HRT beds at this time. Additional experimental study of the effect of linear velocity in large diameter adsorber beds is needed.

Even though the N values are small the magnitude of the t_b and t_{max} values is such that the HRT beds should perform adequately for the disposal of the fission gases resulting from the HRT operation.

V. Calculated Performance of HRT Charcoal Beds

In order to predict the performance of the beds under various conditions several assumptions will be made. Values for t_b (time for breakthrough of activity) will be used in the calculations rather than t_{max} values. The magnitude of the decay heating in the small diameter charcoal filled pipes and its effect on the adsorption of the fission gases is not known exactly. Therefore, it is assumed that only the charcoal contained in the 6 inch diameter pipe contributes significantly to the holdup of the fission gases and that the temperature is 15°C. Since the large pipe contains 90% of the charcoal in the bed, the experimental t_b value is reduced by 10% to compensate for the decay heating in the first sections of the bed. Based on laboratory data, it is assumed that xenon gas is adsorbed 12 times more effectively than krypton gas (7). These assumptions will tend to under-estimate the actual holdup performance of the beds.

The experimental t_b values were normalized to a hypothetical bed containing 500 pounds of charcoal, 1 liter/min O_2 sweep, 15°C, and then averaged. After reducing the average value by 10% to compensate for the decay heating in the first section of the bed, a holdup of 180 hours is calculated for krypton. The corresponding t_b value for xenon is therefore 2160 hours (90 days). Holdup times for other flow rates were calculated from these values using relationships obtained in laboratory holdup tests (2, 6, 7). Table II lists these holdup times. The flow rate is of particular significance because bed performance is very sensitive to flow rate.

The expected rates of release of activity from the charcoal beds after equilibrium has been reached were calculated for each isotope of krypton and xenon. These calculations were based on data from reports by Van Winkle (9) and Kolb (4) as to amounts of gaseous fission products coming from the reactor core, and residence time of the gases in piping and equipment before reaching the beds. The isotopic composition of the gas stream at the entrance to the charcoal beds at various total gas flow rates from the reactor is listed in Table III. The rates of release of activity for the various isotopes are tabulated in Table II. Fig. 2 indicates the rate of release of activity as a function of total gas flow from the reactor operating at 5 MW and 10 MW power levels. It should be noted that these predicted discharge rates are lower than those previously calculated.[10] The earlier calculations were based on higher charcoal temperatures and on an adsorption coefficient derived from laboratory studies. The adsorption coefficient used in this report was determined in the holdup tests on the HRT adsorber bed.

These predictions are based on the assumption that the temperature of the charcoal contained in the 6" diameter pipe remains at 15°C during operation. A higher temperature will reduce the holdup time of the fission gases. Raising the charcoal temperature in the 6" diameter pipe by 10 degrees is equivalent to increasing the flow rate by 18%. For example, if the reactor is operating at 10 MW with a total gas discharge to the charcoal beds of 3000 cc/min, the calculated discharge rate is 41 curies/day, assuming that the temperature is 15°C. A temperature of 25°C would be equivalent to increasing the flow rate to 3540 cc/min, which gives a discharge rate of 115 curies/day. Assuming the same conditions at 5 MW power level, the activity discharge rate would change from 21 curies/day to 53 curies/day.

It should be noted that the time for establishment of equilibrium rates of release is essentially equal to the t_b values for xenon holdup. Caution should be exercised in deciding upon successive increases in flow rate based upon activity levels observed over short time intervals. For example, the time to reach equilibrium at 3 liters/min oxygen flow from the reactor is 60 days.

In these calculations it is assumed that operation of the reactor and off-gas system is managed in such a way that local overheating of the charcoal bed does not occur. Charcoal in an atmosphere of pure oxygen is potentially unstable; and uncontrolled heating could cause combustion of the mixture with sudden release of some of the accumulated fission gases. This could result in radiation exposure of personnel nearby and in the surrounding area. The temperature of the various parts of the charcoal bed are monitored, and action will be taken to prevent or control a fire if the temperature should exceed safe levels. Stopping the oxygen flow from the reactor will control a fire, if it should develop. After the oxygen at the burning zone is consumed the resultant combustion products (CO and CO_2) will extinguish the fire. Ignition of charcoal in an oxygen stream has been observed at 290°C (1). It is possible that a lower ignition temperature could apply at high radiation levels because of ozone produced in the oxygen.

VI. Conclusions

The existing HRT charcoal adsorber system (two beds in parallel) should perform satisfactorily for disposal of fission product gases produced in the reactor at the 10 MW power level with a total oxygen flow from the reactor of 3000 cc/min, provided the maximum charcoal temperature does not exceed 100°C in the first sections of the bed and that the temperature in the 6" diameter pipe does not exceed the 15° - 20°C range. Under these conditions, the calculated activity discharged from the beds will be 42 curies/day consisting of Kr-85, Xe-131m, and Xe-133. This amounts to 52% of the allowable discharge rate of 80 curies/day. The allowable discharge rate would be exceeded at a total oxygen flow of approximately 3300 cc/min.

For operation of the reactor at a 5 MW power level under the same oxygen flow and temperature limitations, the calculated activity discharge will be 21 curies/day, or 26% of the allowable discharge rate. A total oxygen flow from the reactor of 3700 cc/min would be required to exceed the allowable discharge rate.

Continued operation of the charcoal beds is not recommended under such conditions that produce temperatures in the charcoal much in excess of 100°C because of the decreased adsorption efficiency and increased oxidation rate of the charcoal at temperatures above 100°C.

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References

1. Adams, R. E., Browning, W. E., "Estimate of Probability and Consequences of Ignition of the HRT charcoal beds", CF Memo (in preparation).
2. Browning, W. E., Bolta, C. C., "Measurement and Analysis of the Holdup of Gas Mixtures by Charcoal Adsorption Traps", ORNL-2116.
3. Keulemans, A. I. M., "Gas Chromatography", Reinhold Publishing Corp., New York, 1957, p. 119-128.
4. Kolb, J. O., "Shielding for the HRT Charcoal Pit and Off-Gas Line", ORNL-CF-56-1-106.
5. Leland, T. W., "Design of Charcoal Adsorbers for the HRT", ORNL-CF-55-1-12.
6. McNees, R. A., et al., <u>HRP Quar. Prog. Rep.</u>, July 31, 1957, ORNL-2379, p. 138.
7. McNees, R. A., et al., <u>HRP Quar. Prog. Rep.</u>, Oct. 31, 1957, ORNL-2432, p. 147.
8. Phillips, C., "Gas Chromatography", Academic Press Inc., New York, 1956, p. 76-77.
9. Van Winkle, R., "Heating in HRT Cold Traps from Radioactive Fission Product Gases", ORNL-CF-55-7-141.
10. Weeren, H. O., Lee, W. J., "Performance of HRT Charcoal Beds", ORNL-CF-57-6-120.

Table I

Experimental Holdup Tests

Charcoal Bed	Wt. of Charcoal (lbs)	Temp. Range (°C)	Average Oxygen Sweep (1/min)	Exp. t_b (hours)	Exp. t_{max} (hours)	N Value	Normalized* t_{max} for hypothetical Bed (hours)
10A	520	10-15°	1.251	190	227	373	273
10B	520	10-15°	0.869	195	249	148	215
10C	520	16-19°	1.022	186	211	370	232
10D	360	16-19°	0.947	140	170	137	250

*Experimental t_{max} values normalized to hypothetical charcoal bed containing 500 pounds charcoal; 1 liter/min oxygen sweep, 15°C.

Table II

Rate of Release of Activity at Various Total Oxygen Flow Rates from Reactor (10 MW). Two Charcoal Beds in Parallel

Oxygen Flow from reactor (liters/min)	Holdup Time (hours)		Rate of Release of Activity at Equilibrium (Curies/day)				
	Kr	Xe	Kr^{85m}	Kr^{85}	Xe^{131m}	Xe^{133}	Total
0.5	720	8640	-	7.0	-	-	7.0
1.0	360	4320	-	7.0	-	-	7.0
1.5	240	2879	-	7.0	-	-	7.0
2.0	180	2160	-	7.0	0.7	0.7	8.4
3.0	120	1440	-	7.0	4.6	29.8	41.4
4.0	90	1080	-	7.0	11.3	216	234.3
5.0	72	864	3.1	7.0	19.0	700	729.1

Table III. Composition of Rare Gas Stream at Entrance of Charcoal Beds

Isotope	Half Life	No. of Atoms Leaving Core (Sec^{-1} x 10^{-15})	Number of Atoms Entering Charcoal Beds (sec^{-1}x10^{-15}) at Various Oxygen Flow Rates from Reactor (Liters/minute)							
			0.5	1.0	1.5	2.0	2.5	3.0	3.5	4.0
Kr^{83m}	114m	1.518	0.077	0.341	0.56	0.718	0.836	0.921	0.988	1.04
Kr^{85m}	4.36h	2.83	0.767	1.47	1.84	2.04	2.17	2.28	2.35	2.40
Kr^{85}	10.27y	0.9	1.31	1.17	1.10	1.06	1.03	1.01	0.996	0.986
Kr^{87}	78m	8.57	0.105	0.968	1.99	2.86	3.58	4.13	4.58	4.92
Kr^{88}	2.77h	12.4	1.6	4.44	6.25	7.43	8.22	8.82	9.26	9.59
Xe^{131m}	12.0d	0.095	0.093	0.094	0.094	0.095	0.095	0.095	0.095	0.095
Xe^{133m}	2.3d	0.541	0.487	0.515	0.523	0.526	0.53	0.535	0.536	0.537
Xe^{133}	5.27d	22.4	21.4	21.9	22.0	22.2	22.3	22.4	22.4	22.4
Xe^{135}	9.13h	21.5	11.5	15.7	17.4	18.4	19.0	19.4	19.7	19.9

FIG 2. ACTIVITY DISCHARGE FROM TWO PARALLEL CHARCOAL BEDS

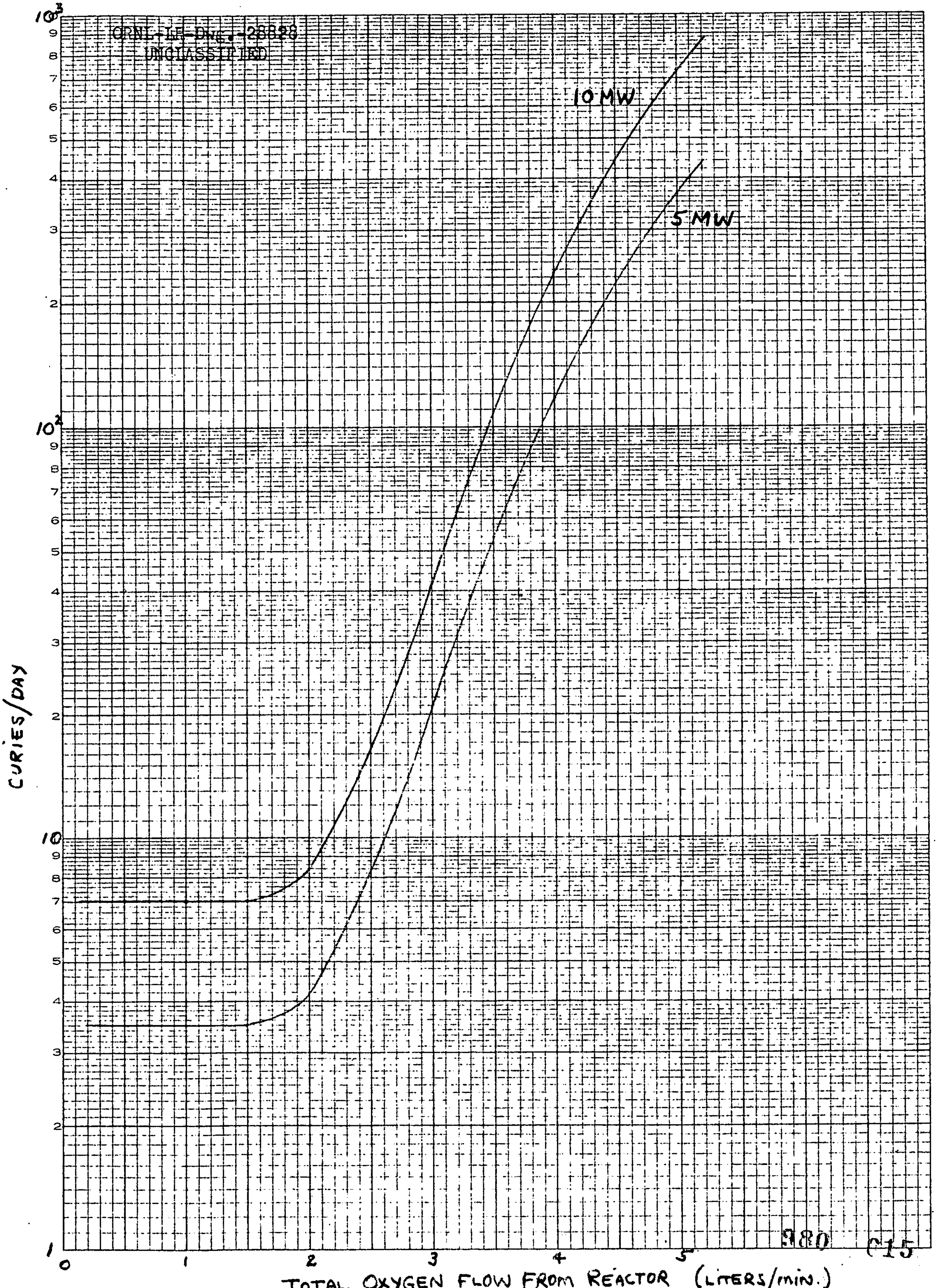

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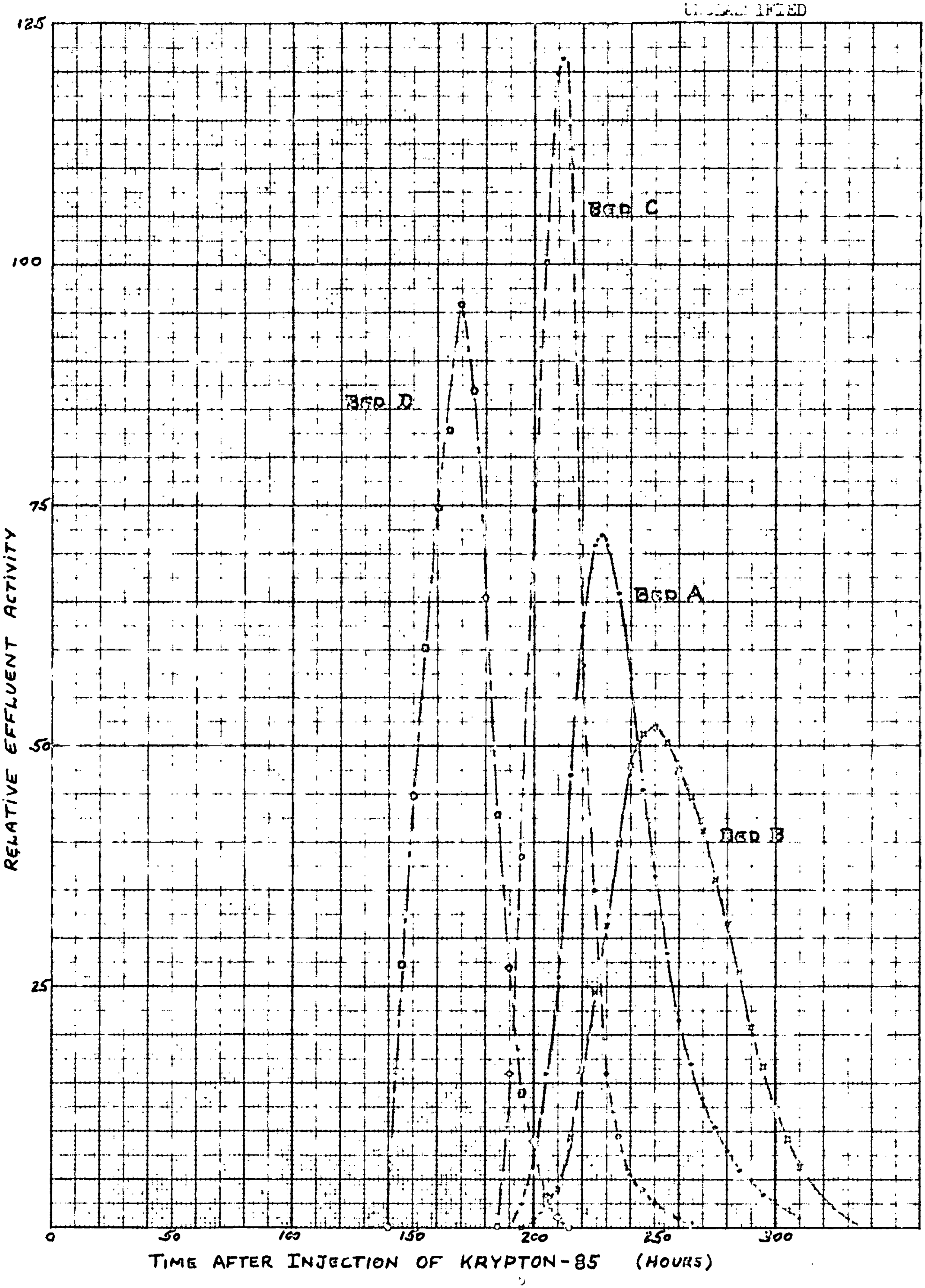
Fig I EXPERIMENTAL HOLDUP TESTS



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