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THE BEHAVIOR OF HIGHLY RADIOACTIVE IODINE ON CHARCOAL IN MOIST AIR*

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Abstract

The behavior of highly radioactive iodine adsorbed on charcoal exposed to moist air (110 torr water vapor partial pressure) was investigated in a series of six experiments. The amount of radioactive ^{130}I on the well-insulated 28 cm^3 bed ranged from 50 to 570 Ci, and the relative humidity was 47% at the bed inlet temperature of 70°C.

Radioactive iodine was released from the test beds at a continuous fractional release rate of approximately 7 x 10^{-6}/hr for all types of charcoal tested. The chemical form of the released iodine was such that it was very highly penetrating with respect to the nine different types of commercial impregnated charcoals tested in backup collection beds. Two types of silver-nitrate-coated adsorption materials behaved similarly to the charcoals. Silver-exchanged type 13-X molecular sieve adsorbers were 20 to 50 times more efficient for adsorbing the highly penetrating iodine, but not as efficient as normally found for collecting methyl iodide. The chemical form of the highly penetrating iodine was not determined.

Methyl iodide was released from the main test beds only while the radioactive iodine was being loaded onto the test beds. An exception was a sample

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of charcoal that had been in service for four years in a reactor building air cleaning system at Oak Ridge National Laboratory. This charcoal, which contained 108 mg of adsorbed atmospheric contaminants per g charcoal, released a methyl iodide-like material continuously at a fractional rate of 80×10^{-6}/hr in addition to the highly penetrating iodine form. A different type of charcoal exposed for nine months at Savannah River Laboratory, which contained much less adsorbed organic impurity, released only the highly penetrating form of iodine.

When the moist air velocity was decreased from 28.5 fpm (25°C) to as low as 0.71 fpm (25°C), the charcoal bed temperature rose slowly and reached the ignition temperature in three of the experiments. At 0.71 fpm (25°C) the ignited charcoal beds reached maximum temperatures of 430 to 470°C because of the limited oxygen supply. The charcoal exposed for four years at Oak Ridge ignited at 283°C compared with 368°C for unused charcoal from the same batch.

Two of the experiments used charcoal containing 1 or 2% TEDA (triethylenediamine) and a proprietary flame retardant. The oxidation and ignition behavior of these charcoals did not appear to be affected adversely by the presence of the TEDA.

I. Introduction

We have completed a series of experiments which had the primary objective of determining whether the ignition of charcoal can occur from the decay heat of highly radioactive iodine before the iodine desorbs thereby preculding ignition. The second objective of our program was to provide supportive data for the calculation of charcoal bed temperatures, and the third objective was to study the movement of iodine within charcoal beds and the desorption of iodine from these beds during exposure to intense radiation fields and elevated temperatures. Seven experiments conducted with dry air were reported previously;(1) six experiments conducted with moist air are reported herein.

We used four types of charcoal in our moist air experiments, Runs Nos. 8-13, Table I. All of them are coconut-based charcoals except for WITCO Grade 42, which is petroleum-based. Each batch performed satisfactorily in other iodine or methyl iodide adsorption tests. Because some large differences in iodine desorption and oxidation rates or ignition temperature have been observed from batch to batch of a given manufacturer's type number, we caution that the experimental results presented here apply only to the particular batches tested.

Our experimental apparatus is shown in Fig. 1. The charcoal is contained to a depth of 2-1/8-in. (5.4 cm) in a quartz Dewar flask with an inside diameter of 1.03 in. (2.62 cm). A heater on the outside of the flask is adjusted to follow the temperature of the center of the bed in order to further reduce the chance for heat loss through the side in simulation of an essentially semi-infinite reactor adsorber system. Except for a small heat loss via conduction and heat radiation, the only cooling mechanism is the forced flow of air upward through the bed. Thermocouples measure the temperature distribution in the charcoal. The inlet temperature was controlled at 70°C in simulation of an accident ambient temperature, while the charcoal reached a higher temperature depending on the amount of radioactive iodine present and the air flow rate.

II. Experimental Procedure

The radioactive ^{130}I was obtained by irradiating a mixture of 86% ^{129}I--14% ^{127}I in the chemical form PdI_2 packed as powder in quartz ampoules and seal-welded in an aluminum capsule. We irradiated this material for 30 hr (except Run No. 13) in the High Flux Isotope Reactor (HFIR) at a thermal flux of 2.5×10^{15} n/cm^2·sec, which resulted in specific activities of 5.7 Ci ^{130}I/mg I and 1.4×10^{-3} Ci ^{131}I/mg I at the time of removal from the reactor.

After irradiation the capsule was transported to the hot cell and sealed into the recirculating loop system. The tips of the capsule and ampoules were then sheared off, thus allowing the ampoules to fall into a quartz-lined furnace where the PdI_2 was thermally decomposed to palladium metal and I_2 at 600 to 700°C. Air circulating at the normal flow rate carried the radioactive iodine through two layers of Cambridge 1G HEPA filter media into the test charcoal bed.

A collimated radiation detector was moved in 1/8-in increments to follow the distribution of iodine in the bed as the experiments progressed. The collimator contained a slit that was 1/16 in. wide and 8 in. long (8-1/2 in. long beginning with Run 10).

Iodine that penetrated or was desorbed from the test bed was collected in sequentially operated traps. HEPA filters (Cambridge 1G) were used to collect particulate matter, and charcoal and silver-exchanged zeolite were used to collect both elemental iodine and the more penetrating organic forms of iodine. We examined the distribution of iodine among the cartridges to determine the relative amounts of elemental and organic iodine.

The experimental procedure can be described as follows. The installed collection traps and heated loop were purged with dried and filtered air, using a once-through mode and a flow rate of 4.9 liters/min for 2.5 hr to dry the loop and remove volatile contaminants. A 20-cm^3 volume of the test charcoal in the as-received moisture condition was then poured into the insulated bed through the outlet tube. (The presence of many thermocouples and the restricted fall resulted in a packing density somewhat less than that measured by the method of ASTM-D-2854-70. Assuming a void fraction of 0.43 for normal packing, we estimate 0.45 for the test bed void fraction.) Subsequently, the loop heaters were turned on, the irradiation capsule installed, air circulation started, and the capsule sheared when the desired bed temperature was reached. The humidifier was heated to 54 or 55°C to maintain proper humidity.

The air inlet temperature to the bed was controlled at 70°C, and the charcoal bed was heated by the decay heat to some higher temperature, depending upon the level of radioactivity and air flow rate, as shown in Tables II-VII. The period of operation at normal air flow rate at the beginning of each experiment enabled observation of the iodine movement and desorption at nearly constant-temperature conditions. The air flow rate was then reduced in simulation of an accident situation (except Run No. 13), allowing the temperature to rise because of reduced air cooling. The operating conditions are also summarized in Tables II-VII.

III. Oxidation of Charcoal

The temperature behavior during several of the runs is shown in Figs. 2-5. The accelerating temperature rise during a period of each of these experiments was th result of an increasing rate of heat release from the oxidation of charcoal as the temperature rose. The rate of rise depended upon the amount of radioactive iodine decay heat, the oxidation characteristics of the charcoal, and the air flow rate. We calculated the heat balance for the test bed, described in more detail previously,$^{(1)}$ and obtained the rate of heat release also shown in Figs. 2-5. At the very low air flow rates employed in these experiments, the rate of heat release decreased following ignition, possibly a result of consumption of the chemically active sites and poisoning of the charcoal surface.

The calculated rate of heat release from oxidation is shown in Fig. 6 as a function of temperature. The heat release data prior to ignition can be correlated for each experiment using the Arrhenius-type equation

$$H = H_o e^{-A/RT} ,$$

where

H_o = a constant (cal/min.g charcoal),

A = an activation energy (cal/mole),

R = 1.987 cal/mole·°K,

H = the rate of heat release by oxidation (cal/min.g charcoal).

The values of H_o and A are given in Table VII. The experimental heat release data are more accurate within the range of 0.5 to 20 cal/min·g charcoal; the accuracy of extrapolations beyond this range is uncertain.

The rate of heat release for MSA 85851 during Run No. 8 was only approximately one-third that observed during Run No. 4 conducted in dry air using charcoal from the same batch and with nearly three times as much radioactive iodine. We believe that much of this difference resulted from the very slow rate of heatup during Run No. 8 which allowed consumption of chemically active sites in the charcoal. When the air flow rate was increased for the final 2 min of the experiment, the oxidation heat release rate and the bed temperatures increased rapidly as seen in Fig. 6.

Run No. 9, using WITCO Grade 42 charcoal, reached a maximum temperature of 184°C and did not evolve measurable heat from oxidation at that temperature. In Run No. 10, charcoal from the same purchase lot that had been used for four years in the High Flux Isotope Reactor (HFIR) air cleaning system at Oak Ridge released heat by oxidation at much lower temperatures and ignited at 283°C. Previous tests with unused charcoal from the same purchase lot indicated an ignition temperature of approximately 368°C.(1) It is quite clear that the adsorbed atmospheric contaminants promoted ignition of the base charcoal. The first 1-inch depth of our test bed contained charcoal from the first 1-1/8-in. bed in the HFIR system and the second inch of our test bed was taken from the second 1-1/8-in. HFIR bed. Infrared analysis of CCl_4 extracts revealed hydrocarbon concentrations of 145 and 71 mg hydrocarbons/g charcoal in the respective HFIR charcoals. Based on the reported air flow during the four years of service, the concentration of adsorbed hydrocarbons was approximately 50 x 10^{-9} g hydrocarbons/g air. This is compatible with the 290 x 10^{-9} g hydrocarbons/g air (excluding methane) found in the HFIR building in 1967.(2) The apparent density of the HFIR charcoal indicates a total weight gain almost twice that given by the CCl_4-infrared method.

The heat from oxidation measured for GX-176 during Run No. 11 was slightly lower than that measured previously during Run No. 7.(1) As shown in Fig. 5, the oxidation heat release rate decreased and the bed temperatures stabilized after ignition.

Run No. 12 used type G-615 charcoal that had been used for nine months at Savannah River Laboratory and contained 16 mg hydrocarbons/g charcoal according to infrared analysis of a CCl_4 extract. As shown in Figs. 5 and 6 the organic contaminants and/or TEDA impregnant apparently oxidized at relatively low temperatures but there was not sufficient heat released to ignite the base charcoal. We believe that a higher rate of temperature rise provided either by lower air flow or higher radiation decay heat could well have resulted in ignition of the base charcoal. Both G-615 and GX-176 are reported to contain a proprietary flame retardant that could influence the oxidation heat release rates.

A sophisticated computation of temperatures of a charcoal bed containing organic material would properly assume separate oxidation heat release rate equations for the base charcoal and for the organic material, and provide a maximum total heat release for the organic material depending on the amount present.

IV. Release and Desorption of Iodine

As mentioned previously, we used sequentially operated traps composed of high-efficiency filters, charcoal cartridges, and silver-exchanged zeolite to identify the iodine forms released during the course of the experiments. The results are summarized in Tables II-VII. Identification of the form of the collected iodine was uncertain in some cases. For example, most of the released elemental iodine plated out in tubing common to all of the individual collection traps. Many of our collection beds operated at unusually low air velocity, a condition for which very little I_2/CH_3I adsorption efficiency data have been reported. For constant residence time, adsorption efficiency decreases with velocity. Our rather scattered data suggest that for velocities in the range of 1 to 10 cm/sec, the adsorption (trapping) efficiency for methyl iodide on good quality impregnated charcoal will not exceed values extrapolated from the data of May and Polson(3) given by them for nearly dry air. Another departure from conventional adsorption techniques was the absence of clean air purging following a collection period. This permitted loosely adsorbed forms of radioactive iodine to be spread throughout the collection bed.

Highly Penetrating Iodine

Our first experiment with moist air, Run No. 8, revealed that most of the radioactive iodine released from the main test bed was in a chemical form usually penetrating with respect to impregnated charcoal. For subsequent runs we eliminated the silver-plated honeycombs (used for trapping elemental iodine) and increased the number of cartridges containing silver-exchanged zeolite (AgX).

The highly penetrating nature of the released radioactive iodine is illustrated in Fig. 7. The collection trap from which these data were obtained was operated for 2 hr, beginning 4 hr after loading was completed. The trap temperature was 130°C, air velocity 17.3 cm/sec at 130°C, and the total residence time (charcoal + AgX) was 0.43 sec. (The collection traps were heated in order to produce low relative humidity and therefore promote high adsorption efficiency. With the exception of Run No. 10, the distribution of radioactive iodine among the charcoal cartridges indicated loosely held or poorly sorbed species. For Run No. 10 we believe that the large amount of radioactive iodine on the first cartridge was methyl iodide. For the operating conditions of this collection bed, each charcoal cartridge should collect more than 95% of the entering methyl iodide. In addition to G-618, eight other types of commercial impregnated charcoal were tested in other collection traps without significant difference in adsorption efficiency.

The silver-exchanged zeolite was much more efficient for collecting the highly penetrating radioactive iodine. The decrease in slope between the AgX cartridges (Fig. 7) suggests that at least two forms of highly penetrating iodine were present. Our AgX material was prepared from type 13X molecular sieves in the 1/16-in. diam pellet form (mean particle diam 0.20 cm, void fraction 0.55) exchanged to < 95% silver content. Two other batches of the same size AgX prepared by others were tried in other collection beds and no efficiency difference was observed. AgX beads in the 10-20 mesh range and granular AgX in the 12-16 mesh range (particle diam 0.16 cm, void fraction 0.43) were also tried and both were found to have higher efficiency. The mean particle size and void fraction differences could easily account for the higher adsorption efficiency because of improved gas phase mass transfer.

Beginning with Run No. 10 we added high temperature backup beds in the circuits used during normal air flow rate operation. Beginning with Run No. 12 all flow lines were routed through these beds which contained 45 cm^3 total of Hopcalite, an oxidizing catalyst, MSA lot No. 21215.* This was followed by 42 cm^3 total of silver-exchanged zeolite pellets, all in a 2.84 cm diam tube. Table X shows the results of most of the high temperature backup traps. The AgX appears to have performed efficiently, yet a large amount of radioactive iodine reached the loop cleanup filter which contained 550 cm^3 of BC-721 charcoal.

Such large penetrations are difficult to understand. The distribution of radioactive iodine in the loop cleanup filter was examined to see if back-diffusion from the humidifier and main test bed during the 8-day cooldown period could be the source of the iodine. Some back-diffusion occurred, but only perhaps 10% of the total on the cleanup bed. Ackley and Davis tested this same catalyst and found it to be beneficial when CH_3I was used but detrimental when I_2 was being tested.(4) A single test with AgX only in a high temperature backup trap in our system was inconclusive because of possible penetration through a Hopcalite-AgX combination used later in the same run. Our calculations indicate that much of the improved performance of AgX with methyl iodide as the temperature is increased, can be accounted for by improved gas phase mass transfer.

Two other adsorbing materials were tested for their ability to trap the highly penetrating iodine. AC-6120** is an incombustible adsorbent which traps I_2 and CH_3I very efficiently.(5) It is a porous form of amorphous silicic acid impregnated with $AgNO_3$. We tested this bead-shaped material in our standard collection beds at 130°C during Run No. 13. Its performance was comparable

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*Mine Safety Appliances Co.

**Bayer

to the charcoals and inferior to AgX for trapping our highly penetrating iodine. Similar results were obtained for GX-135, a granular silver nitrate-coated aluminum silicate-based material which is no longer manufactured.

Characteristics of the Highly Penetrating Iodine

Beginning with Run No. 10 we added a sampling circuit, not shown in Fig. 1, in an attempt to collect and identify the highly penetrating iodine form(s). The circuit consisted of two HEPA filters, a condenser at 0°C, a freeze trap at -78°C, and a trap at -78°C containing cartridges of type 13X sodium zeolite, silica gel, Tenax, and Porapac-Q (the latter two are commercial chromatograph packings) alone or in combinations depending upon the experiment. The HEPA filters, condenser, and freeze traps collected only very small amounts of the radioactive iodine. All of the sorbents retained the radioactive iodine efficiently at -78°C, but the Porapak-Q appeared to release the iodine more easily when the sorbents were warmed later for transfer of the radioactive iodine to an evacuated stainless steel holding tank.

After several days to allow reduction of the radiation intensity by decay, the contents of the tank were admitted to a mass spectrometer for mass identification. Relatively large amounts of air and CO_2 adsorbed along with the radioactive iodine during the experiment were present in the holding tank and provided a high background which concealed iodine-containing mass peaks in the mass spectrometer. A positive peak at mass 130, presumable $H^{129}I$ was observed in the gas from Run 10, but we believe this to be a result of reaction with some equipment component, probably the type 13X sodium zeolite used as the original sorbent. In later experiments we decreased the air + CO_2 background by concentrating the radioactive iodine in a small trap containing cooled Tenax, but the mass results were inconclusive.

Our original radioactive iodine loaded onto the main test bed contained ^{129}I and ^{127}I in the ratio of 6:1. In the mass spectrometer we observed several paired mass peaks with approximately this same ratio. The larger of the paired peaks occurred at masses 131, 141, 168, 181, 186, 191, and 198. Mass peaks at 262, 278, 281, 331, and 354 were also seen frequently but appeared to be associated with the background emanating from within the needle valve, probe, and mass spectrometer internals.

None of these masses correspond to common iodine compounds, and we do not know whether any actually contained iodine. Since the charcoals in the main test bed all contained impregnant iodine (^{127}I) in greater quantity than the loaded radioactive iodine, we expected that the highly penetrating iodine leaving our test bed might have isotopic ratios of 129/127 much less than 6:1.

Several incidental non-iodine-containing masses were observed. From Run No. 11 (new GX-176) and Run No. 12 (new MSA 85851) we observed masses 101, 103, and 105 corresponding to the fragment $CFCl_2^+$. From Run No. 12 (G-615 used nine months at Savannah River Laboratory) we observed Freon 113 (1,1,2 trichloro-1,2,2 trifluoroethane), Freon 112 (1,1,2,2 tetrachlorodifluoroethane), and a large amount of trichloroethylene.

From the amount of radioactive iodine collected in the condenser in this circuit we could estimate the solubility of the form of iodine present. For Run No. 10 (which contained predominantly a methyl iodide-like form) and for later runs (which exhibited only the highly penetrating form of iodine) we calculated partition coefficients $\leq 10 \frac{gI/cm^3 \text{ water}}{gI/cm^3 \text{ air}}$ at 0°C. The radioactive iodine in the condensate was extracted readily by carbon tetrachloride. This behavior and the low partition coefficient suggest some organic iodide form(s). Formation and release from the main test bed was essentially independent of air velocity and temperature. Only traces of highly penetrating iodine were observed during the dry air experiments. We observed that with a long holdup time on most of the mentioned adsorbents (including charcoal) most of the radioactive iodine became tightly bound. J. G. Wilhelm(6) has shown that aryl iodides exhibit much of the behavior described above, but we have not been able to detect any of this class of compounds.

Release of Methyl Iodide

A small amount of methyl iodide-like material either penetrated through or was released from the main test bed during the loading phase of each experiment. The identification was based only by its trapping behavior on impregnated charcoal. Methyl iodide-like material was not observed later in the runs except for Run No. 10 in which this type of material was continuously released. With the dry air experiments,(1) most of the form of iodine described as "penetrating" is believed to be have been methyl iodide although no positive identification was made. Small amounts of the highly penetrating form was present in at least some of the dry air experiments, but it was rarely detected because of smaller collection beds, only occasional use of AgX sorbent, and concealment by ^{131m}Xe radioactivity (a daughter of ^{131}I) which accumulated in the recirculating loop.

Desorption of Elemental Iodine

Since very little elmental iodine was released from the end of the main test bed, we used data collected by the collimated gamma scanner to calculate iodine adsorption/desorption coefficients. Gamma scan data are shown in Fig. 8 and in Table II. A computer program was written to calculate the partial pressure of iodine in equilibrium with the charcoal according to the observed rate of movement within the bed. The rate of mass transfer for elemental iodine is very rapid, so that there is an essentially continuous equilibrium between the charcoal and the vapor phase. Therefore, the movement of iodine should be directly proportional to the air velocity within the bed. The gamma scanner monitored 17 increments or slabs along the length of the bed. We assumed that iodine which desorbed from the first (inlet) slab was readsorbed on the second slab, etc. We assumed a linear adsorption isotherm; thus the partial pressure of iodine is directly proportional to the mass concentration on the charcoal, $P = x/k$, where P is the partial pressure of iodine (atm), x is the total concentration of iodine on the charcoal, including impregnant (0.03 to 0.1 g of iodine total per gram of charcoal), and k is the adsorption coefficient.

The calculation is most accurate for the first 1/2-in. of bed depth; hence we correlated experimental k values for this region with the temperature obtained from a thermocouple located in that region of the bed. Results are shown in Fig. 9. For each type of charcoal we can express the value of k satisfactorily with the equation $k = k_o e^{-A/RT}$, where A is the activation energy (cal/mole), R is the gas constant, T is the absolute temperature (°K), and k_o is a constant for each type of charcoal (g I/g charcoal·atm I).

The coefficients calculated for these moist air experiments are essentially the same as those determined previously for the dry air experiments.(1) The one exception is Run No. 10 which used heavily contaminated charcoal that desorbed iodine at a rate approximately six times greater than other charcoals.

When test bed temperatures remained constant, the calculated coefficients generally rose somewhat with longer times indicating stronger adsorption. The coefficients shown in Fig. 9 should therefore be used only for short-term situations such as in our experiments. The relatively large mass of radioactive iodine used in our experiments may have contributed to this behavior.

We had expected that significant differences in adsorption coefficient might be observed for different charcoals since previous observations have demonstrated a strong effect of relative potassium and iodine concentrations on iodine desorption.(1,7,8) Our assumption that the observed movement of iodine within the test bed was simple elemental iodine adsorption/desorption could be in error; the high radiation field could enhance the movement by some yet unrecognized mechanism. The ion chamber gamma scanner readings contained distortions introduced by collimator inefficiency and the inclusion of scattered radiation. No corrections were made for these distortions which made the low activity readings appear higher than their true levels. F. G. May performed a calibration which demonstrated the type of distrotion inherent in the system.(9)

V. Conclusions

Heat from the oxidation of charcoal and organic contaminants is an important contributor to attainment of ignition. Computer programs such as CHART(1) and TOOHOT(10) have demonstrated satisfactory calculation of charcoal bed temperatures including ignition, when heat release resulting from oxidation of the charcoal was included.

Movement of radioactive iodine within the charcoal bed and desorption from the end of the bed was trivial before ignition at the low air flow rates used in our experiments (0.7 to 3.8 fpm at 25°C reference). Higher flow rates would enhance radioactive iodine movement and desorption somewhat, but unusually large heat sources would be required to reach a critical temperature.

The decreasing heat release rate, stabalized bed temperatures, and very slow radioiodine desorption rates following ignition when low air velocities prevail provide time and opportunity for combatting the combustion if suitable methods can be developed. Increased air velocity or induced turbulence would be undesirable.

The formation and release of highly penetrating iodine, although occurring at a very low rate, appears to be a product of moisture and radiation. Identification and trapping of this material were not completely solved, but the use of some silver-exchanged zeolite is recommended in the collection system whenever the highly penetrating form is suspected to be present.

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Figure Captions

1. Flow diagram for experiments with moist air.
2. Temperature behavior during Run No. 8 with MSA 85881 charcoal.
3. Temperature behavior during Run No. 10 with WITCO grade 42 used four years in the HFIR.
4. Temperature behavior during Run No. 11 with GX-176 charcoal.
5. Temperature behavior during Run No. 12 with G-615 charcoal used nine months at SRL.
6. Rate of oxidation heat release during experiments with moist air.
7. Distribution of radioactive iodine among components of a collection trap.
8. Distribution and movement of radioactive iodine in the main test bed of Run No. 10.
9. Desorption coefficients calculated for radioactive iodine in the main test bed of experiments conducted in moist air.

TABLE I

CHARACTERISTICS OF CHARCOAL INVESTIGATED

Charcoal Type	ORNL Lot No.	Experiment No.[a]	Dry Mass in Each Exp. (g)	Dry Density (g/cm³)	Apparent Density[b] (g/cm³)	Mean Surface Particle Size[c] (cm)	K[d] (%)	I[d] (%)	pH[e]
MSA 85851[f]	51969	1, 2, 3, 4, 8, 13	11.3	0.420	0.45	0.161	2.85	4.41	9.1-8.6
BC-727[g]	1683	5	13.1	0.461	0.562	0.169	1.87	3.79	8.2-8.2
WITCO Grade 42[h]	62469	6, 9 10 (first in.) 10 (second in.)	10.9 6.77 6.14	0.384 0.513 0.449	0.398 0.564 0.513	0.171 0.168 0.174	0.90 0.83 0.96	3.07 2.72 3.32	8.3-7.8 3.6-3.3 7.1-6.8
GX-176[i]	SR-7	7, 11	14.7	0.524	0.563	0.151	1.56	1.30	9.1-8.8
G-615[j]	SR-6	12	14.9	0.539	0.686	-	1.0	1.3	7.3-7.1

[a] Experiments 1-7 reported in Ref. 1.

[b] Apparent density of the moist charcoal according to the method of ASTM D-2854-70.

[c] Particles collected on 8-mesh or passing 16-mesh (U.S.) screens eliminated. $D_p = W/\Sigma (W_i/D_{pi})$.

[d] Weight percent based on apparent density of moist (as-received) charcoal. Moisture content varied because of different storage conditions.

[e] First value obtained from 5 g charcoal in 20-ml water for 30 min at room temperature. Second value after heating to boiling and cooling to room temperature in covered container.

[f] Product of Mine Safety Appliances Company, Pittsburgh, Pa.

[g] Product of Barnebey-Cheney, Columbus, Ohio. Charcoal obtained from A. G. Evans, Savannah River Laboratory.

[h] Product of WITCO Chemical Chemical Corporation, New York, N.Y.

[i] Product of North American Carbon, Inc., Columbus, Ohio. This charcoal also contains 1% TEDA (triethylenediamine) and a proprietary flame retardant. Charcoal and chemical analyses obtained from A. G. Evans, Savannah River Laboratory.

[j] Product of North American Carbon, Inc., Columbus, Ohio. This charcoal also contains 2% TEDA and a proprietary flame retardant. The charcoal was obtained from A. G. Evans after 9 months service at Savannah River Laboratory.

Preliminary Summary of Conditions and Radioiodine Release, Run 8, MSA 85851 Charcoal

Time interval (min)	-340 to -240[a]	-240 to -120	-120 to 0	0 to 150	150 to 465
Main bed activity (Ci ^{130}I)[b]	465	415	370	320	240
Bed midpoint temp. range (°C)	72-93	80-93	79-80	79-147	147-42
Flow velocity (fpm at 25°C)[d]	28.5	28.5	28.5	3.8	(e)
Water vapor partial pressure (torr)	85	95	95	100	105

Iodine Form	Fraction of Radioactive Iodine Released (10^{-6})				
Elemental	<0.3	~~0.39~~ <0.4	<0.05	0.13	0.74
Particulate	0.004	0.001	0.001	0.001	1.20
Moderately Penetrating	~0.5	<0.4	<0.05	~0.1	<0.4
Penetrating	~~3.50~~ ~3.0	~~8.71~~ ~8.6	~~11.8~~ ~11.5	(f)	(f)
Total[g]	~~3.8~~ ~3.7	~~9.1~~ ~8.6	~~11.8~~ ~11.5	>2.4	~~>3.9~~ >3.4

(a) Loading of 100 mg of radioactive iodine took place during this period.

(b) Activity at end of time period. The half-life of ^{130}I is 12.3 hr.

(c) The ignition temperature (assumed to be 310°C) was reached at 396 min at approximately 0.8-in. bed depth.

(d) Moist air at 0.98 atm.

(e) Velocity was reduced to 1.43 fpm at 150 min (147°C), reduced to 0.71 fpm at 300 min (195°C), increased to 1.9 fpm at 457 min (369°C), and increased to 3.8 fpm at 460 min (412°C).

(f) These collection traps did not contain silver-exchanged zeolite. Total fraction found in the trap charcoal, loop cleanup charcoal, and humidifier was ~~44~~ 47 x 10^{-6}. Total of all penetrating iodine released was 68 x 10^{-6}.

(g) Grand total fractional release of radioactive iodine was 71 x 10^{-6}.

Preliminary Summary of Conditions and Radioiodine Release, Run 9 – WITCO Grade 42 Charcoal

Time interval (min)	–465 to –360[(a)]	–360 to –240	–240 to –120	–120 to 0	0 to 180	180 to 360
Main bed activity (Ci ^{130}I)[(b)]	535	480	430	380	320	270
Bed midpoint temp. range (°C)	70–92	81–92	80–81	79–80	79–166	166–181[(c)]
Flow velocity (fpm at 25°C)[(d)]	28.5	28.5	28.5	28.5	3.8	0.71
Water vapor partial pressure (torr)	95	100	95	95	95	95
Iodine Form	**Fraction of Radioactive Iodine Released (10^{-6})**					
Elemental[(e)]	~ 0.4	< 0.3	< 0.3	< 0.3	< 0.5	< 0.5
Particulate	0.03	0.05	0.06	0.001	0.4	0.002
[illegible]	0.4	0.2	0.2	≤0.2	<0.2	<0.2
Penetrating[(f)]	>> 1.2	>> 7.0	>> 2.6	>12.	15.	3.1
Total[(g)]	>> 1.6	>> 7.1	>> 3.2	>12.	15.5	3.2

(a) Loading of 123 mg of radioactive iodine took place during this period.

(b) Activity at end of time period. Half-life of I-130 is 12.3 hr.

(c) Ignition was not reached because of the low radioactivity and the high ignition temperature of WITCO Grade 42 charcoal.

(d) Moist air at 0.98 atm.

(e) Total fractional release of elemental iodine was probably in the range 0.4×10^{-6} to 1.4×10^{-6}.

(f) The fraction which penetrated the four collection beds used during normal air flow operation and was collected in the backup bed and humidifier was 21×10^{-6}. Total fractional release of penetrating iodine was 63×10^{-6}.

(g) Grand total fractional release was 65×10^{-6}.

Run 10 Preliminary Summary of Conditions and Radioiodine Release - WITCO Grade 42 Charcoal from HFIR Air Cleaning System

Time interval (min)	-470 to -360[a]	-360 to -300	-300 to -120	-120 to 0	0 to 180	180 to 296
Main bed activity (Ci ^{130}I)[b]	485	460	390	345	290	260
Bed midpoint temp. range (°C)	70-88	83-88	78-83	77-78	77-139	139-376[c]
Flow velocity (fpm at 25 °C)[d]	25.7	28.5	28.5	28.5	3.6	0.71
Water vapor partial pressure (torr)	100	100	110	110	110	110
Iodine Form (Tentative)	**Fraction of Radioactive Iodine Release (10^{-6})**					
Elemental	-	~ 1.4	-	-	-	1600
Particulate	< 0.2	< 0.03	2.5	< 0.7	< 0.03	< 1
Moderately Penetrating	130	~ 76	266	156	24	≥ 630
Very Penetrating	8	~ 8	25	15	20	18
Total[e]	139	86	294	171	44	2200

(a) Loading of 100 mg of radioactive iodine took place during this period.
(b) Activity at end of time period. Half-life of ^{130}I is 12.3 hr.
(c) Ignition temperature of 283 °C reached at 275 min at appx. 0.3-in. bed depth. Ignition temperature is bas on thermocouple rate-of-rise first reaching 20 °C/min. Maximum recorded temperature was 473 °C.
(d) Moist air at 0.99 atm.
(e) Grand total fractional release was 2950 x 10^{-6} or 0.295%.

Run 11. Summary of Conditions and Radioiodine Release

Type GX-176 Charcoal

Time interval (min)	-470 to -360[a]	-360 to -300	-300 to -120	-120 to 0	0 to 180	180 to 360
Main bed activity (Ci ^{130}I)[b]	570	540	455	410	345	291
Bed midpoint temp. range (°C)	70-93	89-94	80-87	79-80	79-120	120-396[c]
Flow velocity (fpm at 25°C)[d]	26-28	16-26	28.5	28.5	5.8	1.9
Water vapor partial pressure (torr)	105	105	110	110	110	110

Iodine Form	Fraction of Radioactive Iodine Released (10^{-6})					
Elemental	< 0.3	< 0.2	< 0.2	< 0.2	< 0.2	6.4[e]
Particulate	0.05	0.26	3.0	1.5	0.29	0.59
Moderately Penetrating	0.34	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
Very Penetrating	6.3	5.2	28.1	20.7	27.7	19.7
Total[f]	6.7	5.5	31.1	22.2	2[illegible].9	26.6

(a) Loading of 133 mg of radioactive iodine took place during this period.
(b) Activity at end of time period. Half-life of ^{130}I is 12.3 hr.
(c) Ignition temperature of 307°C was reached at 319.5 min after first flow reduction. Maximum recorded temperature was 534°C.
(d) Moist air at 0.98 atm.
(e) This amount of iodine was leached from tubing network between the main test bed and the absorber traps. Most of the release probably occurred at the high bed temperature and was probably mostly elemental iodine.
(f) Not included were 7.2×10^{-6} found in loop cleanup bed and 0.4×10^{-6} in humidifier. Grand total release was 124.6×10^{-6} or 0.0125%.

Run 17. Summary of Conditions and Radioiodine Release from Type G-615 Charcoal Used 9 Months at SRL

Time interval (min)	-491 to -361[(a)]	-361 to -300	-300 to -120	-120 to 0	0 to 180	180 to 450
Main bed activity (Ci ^{130}I)[(b)]	480	455	380	340	290	225
Bed midpoint temp. range (°C)	71-85	85-95	80-85	74-80	74-113	113-219
Flow velocity (fpm at 25°C)[(c)]	21.5	9-22	22	23	5.8	2.9
Water vapor partial pressure (torr)	110	110	115	115	110	115
Iodine Form	Fraction of Radioactive Iodine Released (10^{-6})					
Elemental	< 0.6	< 0.6	< 0.3	< 0.6	< 0.5	6.0[(d)]
Particulate	0.09	0.06	0.5	0.3	0.02	0.003
Moderately Penetrating	0.6	< 0.6	< 0.3	< 0.6	< 0.5	< 2.9
Very Penetrating	8.5	10.9	44.7	28.8	35.0	73.7
Total[(e)]	9.2	11.0	45.2	29.1	35.0	79.7

(a) Loading of approximately 110 mg of radioactive iodine took place during this period.

(b) Activity at end of time period. Half life of ^{130}I is 12.3 hr.

(c) Moist air at 0.98 atm.

(d) This amount of iodine was leached from the tubing network between the main test bed and the adsorber traps. Most of the release probably occurred at the high bed temperature and was probably mostly elemental iodine.

(e) Not included were 10.9×10^{-6} found in the loop cleanup bed and 0.3×10^{-6} in the humidifier water. Grand total release was 221×10^{-6} or 0.022%.

Run 13. Summary of Conditions and Radioiodine Release from Type MSA 85851 Charcoal

Time interval (min)	-145 to 0[(a)]	0 to 60	60 to 195	195 to 315	315 to 2895	2895 to 3015
Main bed activity (Ci ^{130}I)(b)	50	47	42	37	3.3	2.9
Bed midpoint temp. range (°C)	70-71	67-71	69-71	70	68-70	70
Flow velocity (fpm at 25°C)(c)	28.5	21.1	28.5	28.5	28.5	28.5
Water vapor partial pressure (torr)	120-150	120-150	120-150	140	140-145	140-145
Iodine Form	Fraction of Radioactive Iodine Released (10^{-6})					
Elemental(d)	< 0.2	< 0.05	< 0.06	< 0.1	< 20	< 0.6
Particulate	0.001	0.003	0.008	0.0006	0.004	0.007
Moderately penetrating	0.2	< 0.05	< 0.06	< 0.1	< 20	< 0.6
Very penetrating	8.3	6.6	13.9	15.3	111	1.6
Total(e)	8.5	6.6	13.9	15.3	111	1.6

(a) Loading of approximately 77 mg of radioactive iodine took place during this period.
(b) Activity at end of time period. Half-life of ^{130}I is 12.3 hr.
(c) Moist air at 0.98 atm.
(d) The fraction 0.8×10^{-6} was leached from the tubing network between the main test bed and the adsorber traps.
(e) Not included were 2.6×10^{-6} found in the loop cleanup bed and 0.3×10^{-6} in the humidifier water. Grand total release was 160×10^{-6} or 0.016%.

VIII

TABLE IX.

CHARCOAL OXIDATION CHARACTERISTICS

Charcoal type	Run No.	Air velocity (fpm at 25°C)	Oxidation rate parameters HO (cal/min)	A (cal/mole)	Ignition Temp. (°C)
MSA 85851 Lot no. 51969	8	0.7	2.05×10^8	20,000	[a]
WITCO Grade 42 HFIR, 4 yr service	10	0.7	1.25×10^9	20,000	283
GX-176 SR-7	11	1.6	3.37×10^6	14,500	307
6-615 (9 mo. service) SR-6	12	2.5	5.41×10^6	14,500	[a]

[a] Not determined.

Table 9. Rate of release of penetrating forms of radioactive iodine

Run No.	Amount of radioactive iodine (Ci ^{130}I)	Time from first flow reduction (min)	Ave. Temp. (°C)	Partial pressure of water (torr)	Release rate x 10^6 (fraction/hr)	
					Moderately penetrating iodine	Highly penetrating iodine
8	0 - 465	-340 to -240	82	85	0.3	∼2
	465-370	-240 to 0	82	95	<0.1	∼5
	370-240	o to 465	∼150	105	<0.1	∼6
9	535-380	-360 to 0	81	95	0.2	∼7
	380-320	0 to 180	125	95	<0.1	5
	320-270	180 to 360	174	95	<0.1	1.0
10	0-485	-470 to -360	79	100	71	4.4
	485-345	-360 to 0	79	110	83	8.0
	345-290	0 to 180	∼110	110	16	13
	290-260	180 to 296	∼260	110	<325	9.3
11	0-570	-470 to -360	82	105	0.2	3.4
	570-410	-360 to 0	82	110	<0.1	9.0
	410-345	0 to 180	∼100	110	<0.1	9.2
	345-290	180 to 360	∼260	110	<0.1	6.6
12	0-480	-491 to -361	78	110	0.3	3.9
	480-340	-361 to 0	80	115	<0.3	14
	340-290	0 to 180	∼94	110	<0.2	12
	290-225	180 to 450	∼170	115	<0.6	16
13	0-50	-145 to *	70	135	0.08	3.4
	50-37	0 to 315	70	135	<0.04	6.8
	37-3.3	315 to 2895	69	142	<0.5	2.6
	3.3-2.9	2895 to 3015	70	142	<0.3	0.8

Table X. Performance of High Temperature Backup Beds

	Approximate Temperature (°C)	Run No. 10 (10^5 dpm ^{131}I)	Run No. 11 (10^5 dmp ^{131}I)	Run No. 12 (10^5 dmp ^{131}I)	Run No. 13 (10^8 dmp ^{130}I)	
HEPA Filter	150	0.0005	0.0010	< 0.002	0.004	0.0001
1st Hopcalite	~~300~~ 250	0.41	0.52	0.34	0.010	0.24
2nd Hopcalite	~~330~~ 320	0.52	0.65	0.63	0.009	0.26
3rd Hopcalite	360	0.78	1.64	1.5	0.036	0.30
1st AgX	380	17.7	72.2	1.9	13.2	20.0
2nd AgX	400	0.0034	18.2	0.007	3.3	0.14
3rd AgX	420	0.0069	1.1	0.005	0.0002	0.006
HEPA Filter	200	< 0.0005	0.0003	not used	not used	not used
Loop Cleanup Bed*	70	16.5	31.8	37.7	5.98	
Time in Use (min)		300.0	410.0	491.0	460.0	2700.0

*Apparent penetration of above high temperature backup beds.

Distribution of Radioactive Iodine in Main Test Bed

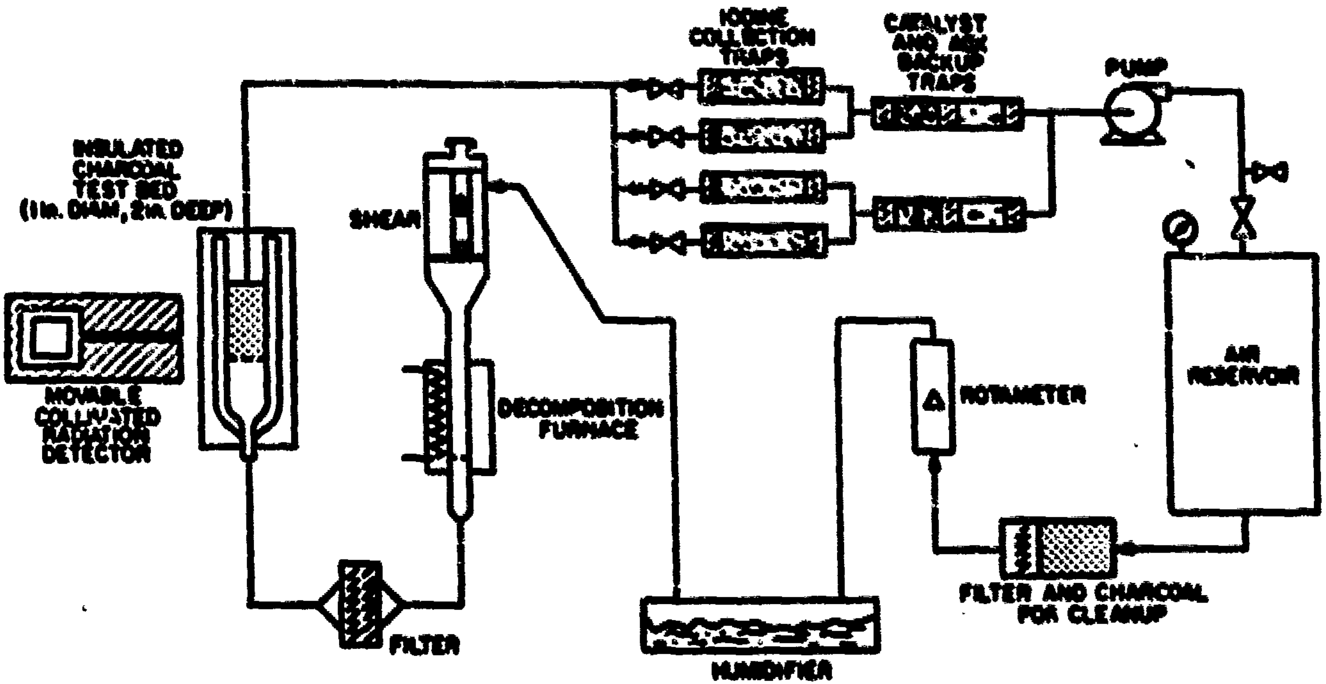
Percent of total radioactivity in each slab

Slab No.[a]	Run 8				Run 9			Run 11			
	Time from flow reduction (min)				Time from flow reduction (min)			Time from flow reduction (min)			
	-235	0	300	465	-360	0	360	-360	0	180	360
1	17.7	12.4	7.5	3.2	16.1	13.9	6.3	6.3	4.7	2.9	0.5
2	59.7	61.4	57.4	26.2	55.2	53.3	46.8	64.2	61.4	59.7	7.8
3	13.3	15.9	23.3	22.8	16.2	18.3	26.9	20.2	23.5	26.4	11.2
4	3.92	4.48	5.93	17.7	5.04	6.33	10.2	4.62	5.37	5.78	11.6
5	1.62	1.89	2.27	12.7	2.45	3.11	4.21	1.71	1.97	2.07	12.2
6	0.86	0.95	1.03	7.74	1.01	1.24	1.56	0.91	1.00	1.02	11.8
7	0.52	0.55	0.55	4.09	0.60	0.75	0.84	0.59	0.61	0.62	10.8
8	0.39	0.42	0.40	2.34	0.38	0.47	0.55	0.42	0.43	0.44	9.47
9	0.27	0.28	0.27	1.18	0.23	0.29	0.34	0.30	0.31	0.31	7.59
10	0.18	0.19	0.16	0.55	0.14	0.18	0.21	0.21	0.22	0.23	6.10
11	0.13	0.13	0.11	0.28	0.10	0.11	0.14	0.15	0.16	0.16	5.50
12	0.09	0.09	0.07	0.13	0.07	0.08	0.10	0.10	0.11	0.11	2.75
13	0.08	0.07	0.04	0.08	0.07	0.07	0.08	0.07	0.07	0.07	1.44
14								0.04	0.04	0.05	0.61
15											0.26
16											0.18
17											0.16
Background	0.49				0.56			0.34			
Bed mid-point temp.(°C)	85	82	196	429	87	81	178	85	78	120	394

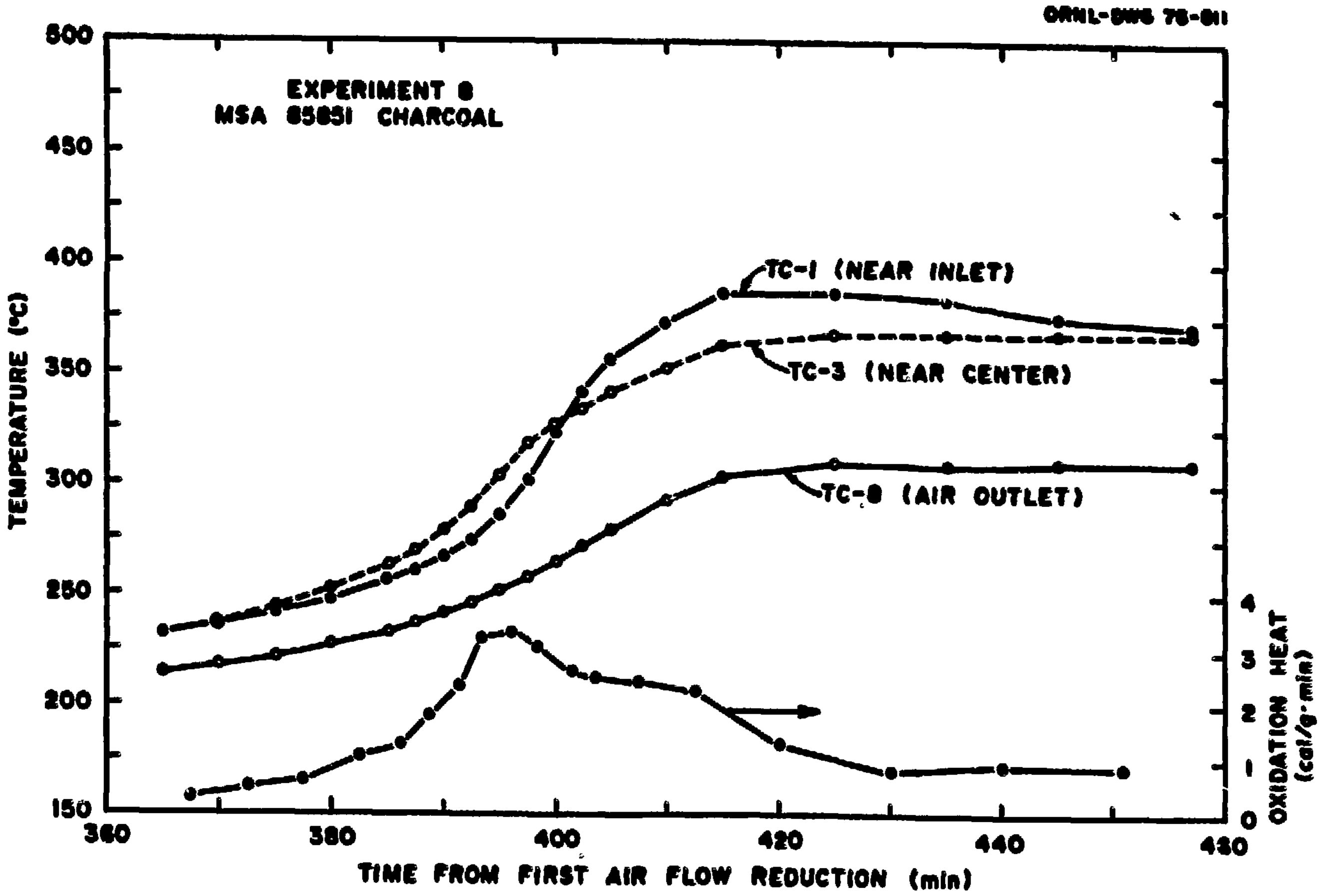
[a] Each slab is determined by collimator movement of 0.125 in. (0.318 cm).

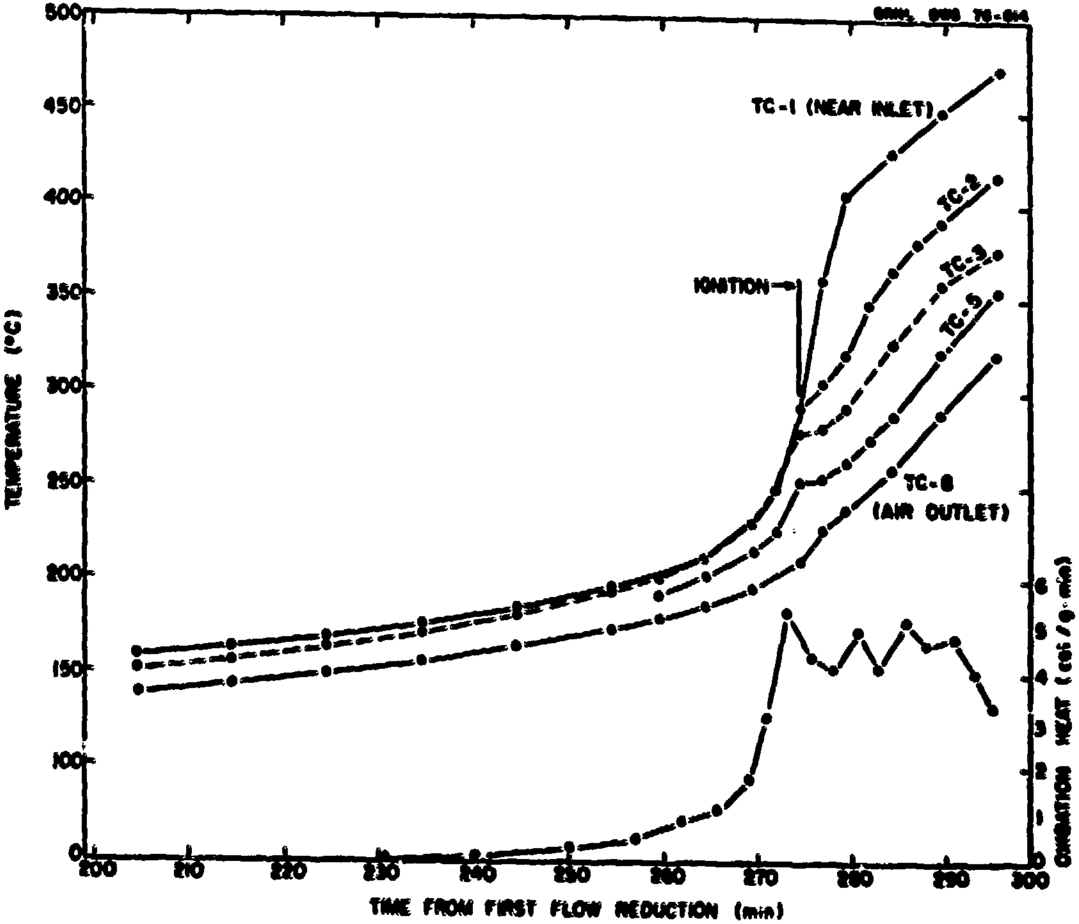
Slab No.ª	Run 12				Run 13		
	Time from flow reduction (min)				Time from end of loading (min)		
	-360	0	180	450	0	1440	2880
1	4.9	3.2	2.6	1.4	2.5	2.1	2.0
2	59.0	55.3	54.3	37.2	61.3	58.2	56.4
3	22.6	25.9	27.0	34.8	25.2	27.9	29.0
4	7.36	8.60	8.90	15.2	5.56	6.16	6.46
5	2.47	3.01	3.12	5.53	2.12	2.31	2.38
6	1.24	1.46	1.49	2.44	1.00	1.06	1.11
7	0.75	0.86	0.88	1.23	0.65	0.70	0.72
8	0.49	0.54	0.55	0.72	0.44	0.43	0.51
9	0.33	0.36	0.36	0.43	0.31	0.31	0.38
10	0.24	0.25	0.26	0.31	0.23	0.23	0.29
11	0.17	0.19	0.18	0.22	0.16	0.16	0.22
12	0.11	0.12	0.12	0.15	0.12	0.11	0.16
13	0.07	0.08	0.08	0.10	0.06	0.07	0.10
14	0.05	0.05	0.05	0.07	0.04	0.04	0.09
15							
16							
17							
Background	0.35				0.37		
Bed mid-point temp.(°C)	83	78	113	219	73	73	73

ORNL DWG 73-730 R1

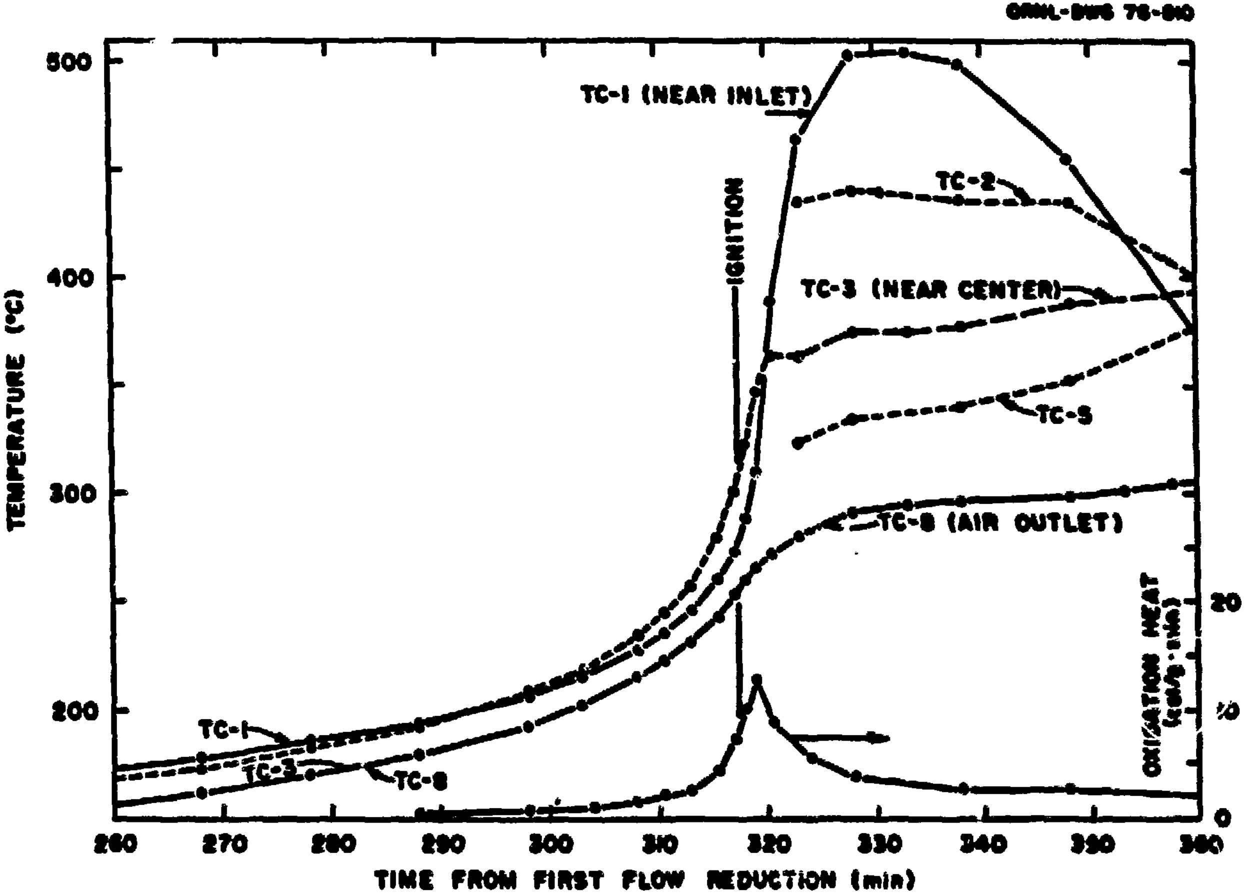


FLOW DIAGRAM FOR EXPERIMENTS WITH MOIST AIR

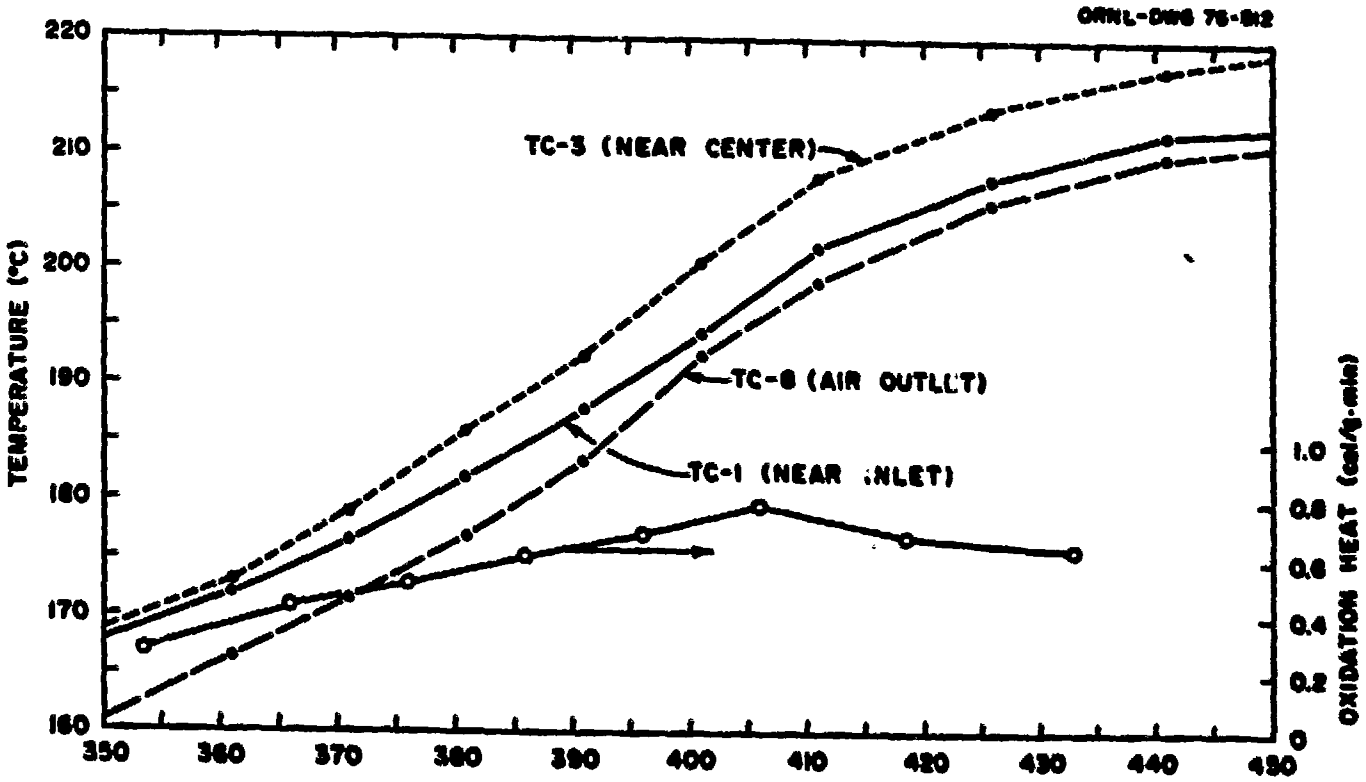

ORNL-DWG 76-811
EXPERIMENT 8
MSA 85851 CHARCOAL
TC-1 (NEAR INLET)
TC-3 (NEAR CENTER)
TC-8 (AIR OUTLET)
TEMPERATURE (°C)
500
450
400
350
300
250
200
150
360
380
400
420
440
460
TIME FROM FIRST AIR FLOW REDUCTION (min)
OXIDATION HEAT (cal/g·min)
4
3
2
1
0



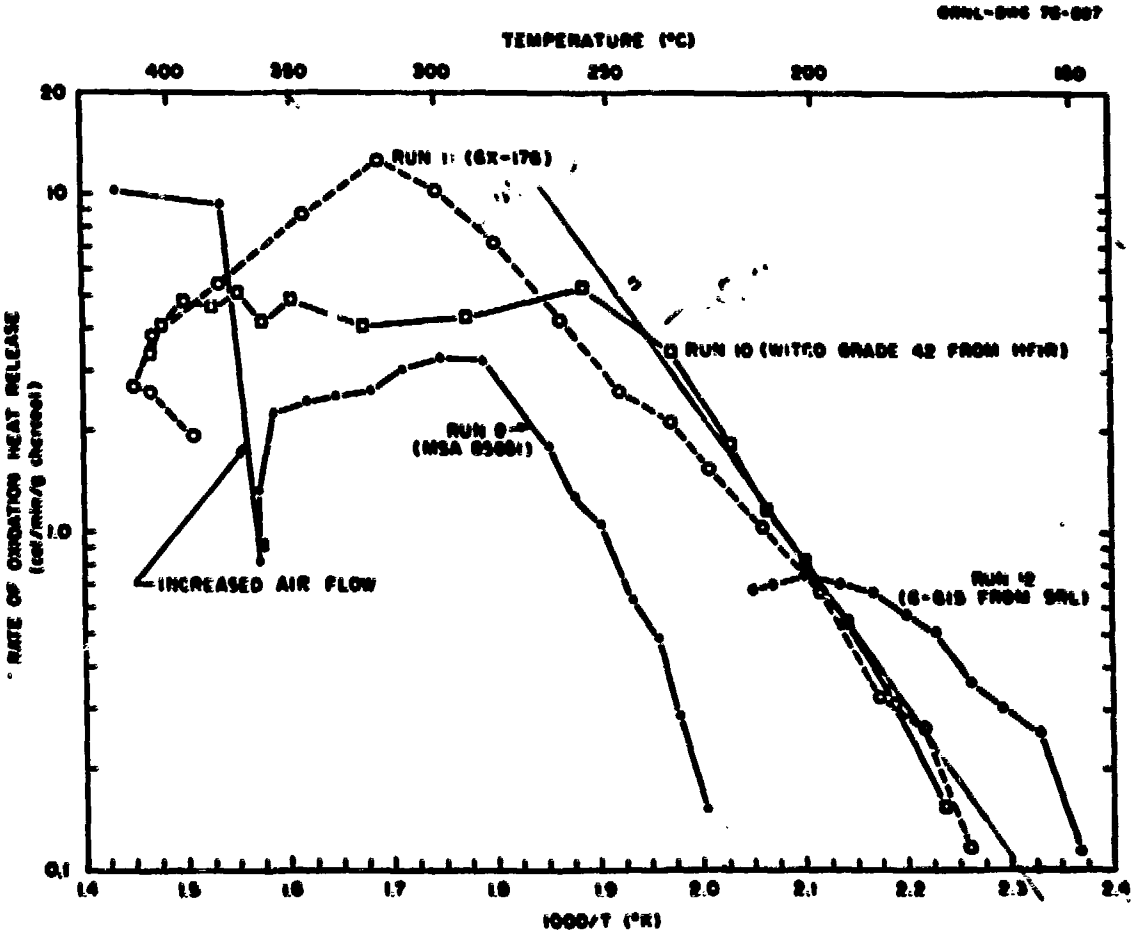
EXPERIMENT NO. 10 WITCO GRADE 42, 4 YRS. IN HFIR



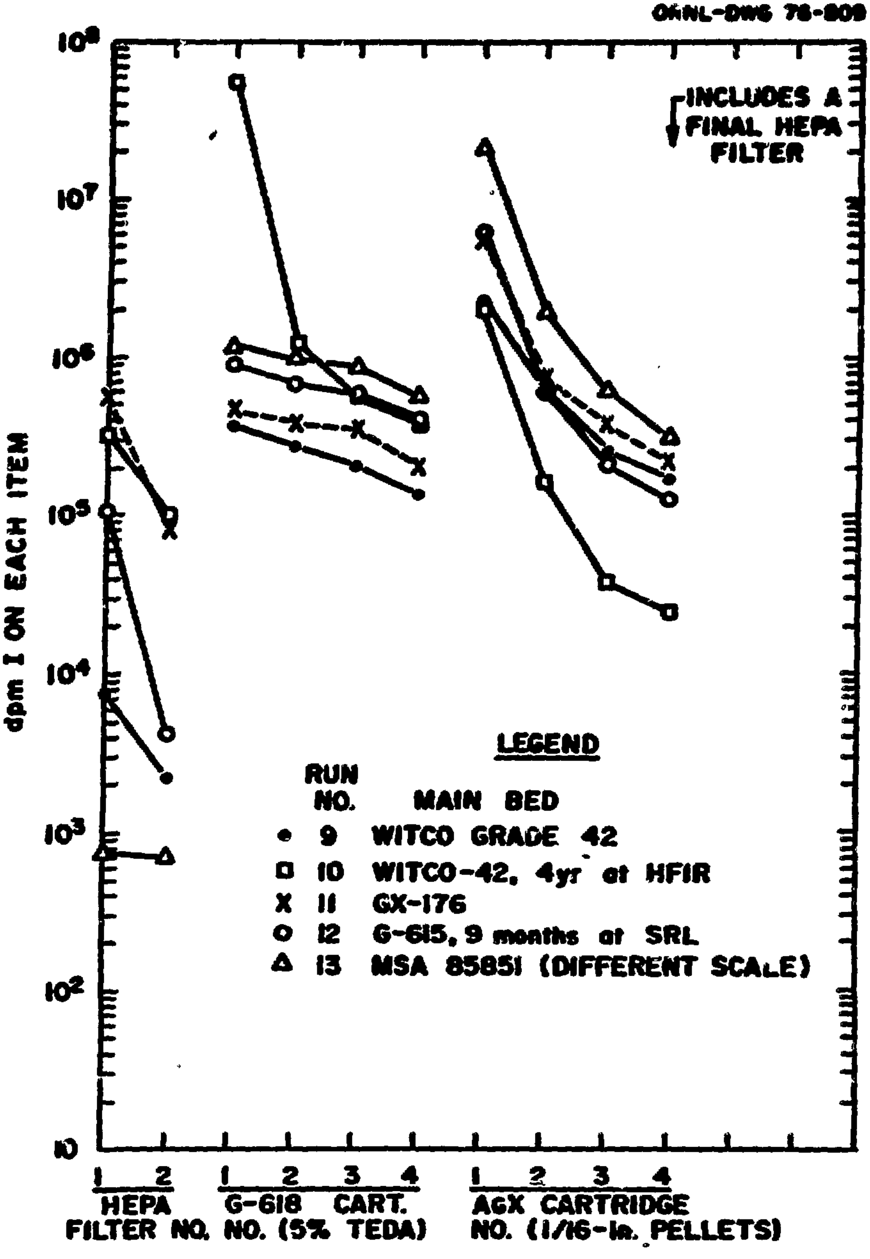
EXPERIMENT NO. 11, GX-176 CHARCOAL (1% KI + 1% TEDA + FLAME RETARDANT).



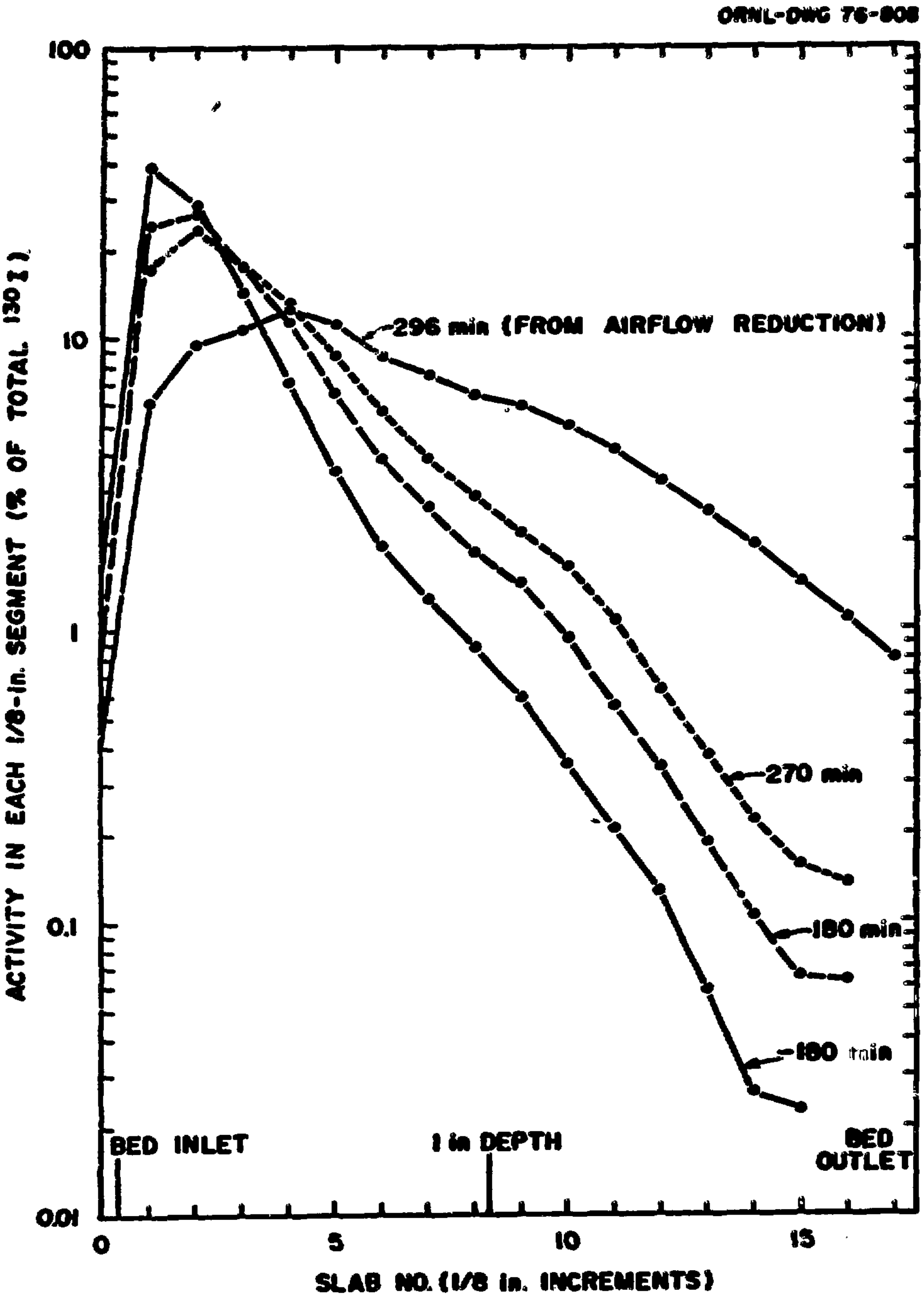
EXPERIMENT NO. 12, G-615 USED 9 MOS. AT SRL (2% KI + 2% TEDA + FLAME RETARDANT).



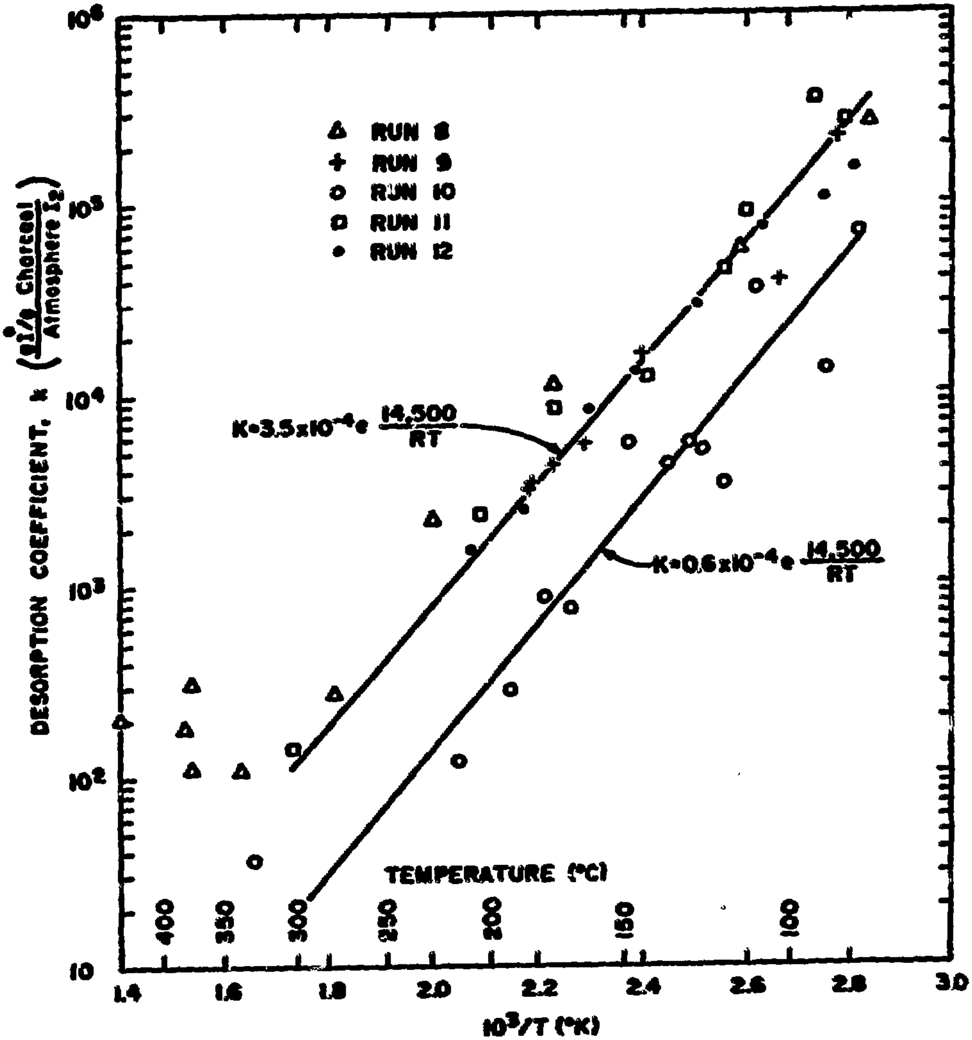
OXIDATION HEAT RELEASE DURING EXPERIMENTS WITH MOIST AIR



DISTRIBUTION OF IODINE ON COLLECTION TRAPS.


ORNL-DWG 76-808
ACTIVITY IN EACH 1/8-in. SEGMENT (% OF TOTAL ^{130}I)
100
10
1
0.1
0.01
296 min (FROM AIRFLOW REDUCTION)
270 min
180 min
180 min
BED INLET
1 in DEPTH
BED OUTLET
0
5
10
15
SLAB NO. (1/8 in. INCREMENTS)

ORNL-DWG 76-813



DESORPTION COEFFICIENTS