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ABSTRACT

This report consists of two papers describing research being conducted at the Harvard Air Cleaning Laboratory in support of the design and development of the Fast Flux Test Facility. They deal with experimental and theoretical evaluations in the following subject areas: (a) the behavior and deposition of sodium aerosols within cell ventilating systems with particular emphasis on heat exchanger tubes; and (b) the adsorption and retention of noble fission gases on activated charcoal as a function of temperature and pressure.

The first paper presents data gathered in a series of tests using a model ventilating system and heat exchangers which simulate as closely as possible the equipment configurations, flow rates, and aerosol characteristics under consideration for the FFTF. Tests showed that, with an average sodium aerosol concentration of 0.69 gm/m^3 and a face velocity of 500 fpm, an 8-row deep finned-tube coil (operating at ambient temperature) remained fully functional for 150 minutes, but air flow resistance increased rapidly thereafter. After 225 minutes, total blockage was imminent. Sodium deposition was 57 grams per sq. ft. of face area. Tests of a tube-type heat exchanger at the design flow rate of 90 fps indicated it would remain functional for approximately an hour although total sodium deposited could reach 11 grams per tube which were one half inch in diameter by eight feet long. Particle deposition was shown to occur at a rate that was proportional to the sodium concentration in the aerosol and the time of operation. In all cases, the experimental results correlated with particle deposition theory. Experiments confirmed that placement of the recirculation blower upstream would reduce the deposition load on the heat exchanger but only at the risk of rapid blower failure by sodium deposition in the blower passages. Periodic one second bursts of compressed nitrogen (150 psi) introduced at the exit end of the tube-type heat exchanger and directed upstream were shown to be effective in removing deposited sodium.

The second paper presents data on dynamic adsorption coefficients for krypton on activated charcoal from streams of argon and helium at temperatures ranging from -140°C to 0°C and at pressures from 15 psig to 135 psig. Similar determinations were made for xenon but only at 37°C because measurements were not practicable at low temperatures. The data show that the adsorption coefficient is considerably higher from helium streams than from argon at all temperatures and pressures studied, and in both cases it increased markedly as the temperature is decreased. With helium as the carrier,

increased pressure caused a proportional increase in adsorption coefficient and the slope of the coefficient-pressure line increased as the temperature decreased. With argon, the slope was small and relatively unaffected by temperature above -120°C . At -120°C , pressure had almost no effect on the adsorption coefficient and at -140°C , the coefficient decreased markedly with increasing pressure up to about 75 psig and then increased gradually above that.

NOTE: In the interest of clarity, the figures and tables are included as an integral part of each paper and are numbered separately within each paper. Similarly, the references are numbered separately within each paper.

PAPER NUMBER ONE

PARTICLE DEPOSITION AND REENTRAINMENT ON FFTF H & V COMPONENTS FOLLOWING A SODIUM SPILL

M. W. First, E. Jochem,
T. W. Baldwin, R. Hall

I. Introduction

Closed loop cells in the FFTF contain major sodium recirculation equipment and must be cooled to protect the machinery enclosed in the space from over-heating. Plans call for the atmosphere inside the cell (0.5-2.0% oxygen; the remainder nitrogen) to be recirculated continuously through an externally-located heat exchanger by a centrifugal or axial flow fan. Because leakage of molten sodium from the equipment inside the closed loop cell will generate sodium oxide aerosols and, when the sodium spill is of sufficient magnitude to consume all available oxygen in the sealed space, metallic sodium aerosols, the system must be protected from excessive deposition of aerosol particles in the heat exchanger and blower if the cooling function is to be maintained during and after a spill.

Sodium aerosol filters located upstream of fan and cooler were considered to be a satisfactory method of preventing sodium deposition in these pieces of equipment (1,2), but space limitations in a redesigned FFTF containment housing made filters impractical. Therefore, studies were undertaken to examine critically the amount of deposition which would be likely on heat exchanger surfaces following a sodium spill of moderate size and to provide, if possible, sufficient excess heat exchanger capacity to overcome the loss in efficiency caused by the fouling of the surfaces. Deposition experiments have been conducted with finned-tube and with shell-and-tube heat exchangers, and the experimental results correlated with particle deposition theory.

II. Experimental Results

A. Deposition in a Finned-tube Coil.

Figure 1 shows the arrangement of apparatus inside the aerosol chamber that was used to test an 8-row deep finned-tube coil. The blower is capable of moving 1400 cfm against a resistance of 4 in.w.g. The test tunnel was provided with a flow measuring orifice meter, temperature probes, pressure taps to measure filter resistance, and a flow regulating damper. Sodium samplers were installed up and downstream of the filter. Face velocity was 500 fpm. Coolant was not used during this experiment so all surfaces were at the same temperature as the aerosol. Oxygen concentration was 1% during this test, and airborne sodium concentrations ranged between 0.3 and 1 gram/m³.

The pressure drop across the 8-row coil at constant air flow rate was monitored as an index of particle deposition. During the first 150 minutes of steady flow at 500 cfm, the pressure drop increased from 0.145 to 0.155, a rise of 0.01 in.w.g. During the next 67 minutes, pressure drop rose to 2.7 in.w.g., indicating a more rapid rate of particle deposition which produced a progressive narrowing of the flow channels in the coil. During the next 8 minutes of operation, the pressure drop rose to 3.8 in.w.g. and the flow rate fell from 500 to 260 cfm. At 500 cfm (had the blower been capable of this flow at this resistance) the coil resistance would have been approximately 14 in.w.g. This behavior is shown graphically in Figure 2. An initial period of slow resistance increase was followed by a period of rapid rise, and, finally, nearly complete blocking of the flow channels. This may be seen in Figure 3, a photograph of the cooling coil section after deposition of sodium. If the experiments had been continued, this would have resulted in total blockage of these channels by deposited sodium aerosol particles. This behavior can be explained by reference to the mechanisms responsible for particle deposition. The largest weight fraction of the particles (i.e., > 80%) is above 0.5 μ m and therefore inertial deposition is the principal separating force. As the air passages in the filter and the coil become smaller by deposition of particles, velocity for the same volumetric flow rate increases and inertial separation, highly dependent on velocity, increases also. Thus, an accelerating rate of deposition accounts for an accelerating rate of pressure drop increase. In the case of the coil section, this suggests that operation at normal face velocities, i.e., 1000 to 1500 fpm, will result in more efficient particle deposition.

During the 225 minutes that the coil was exposed to a flow rate of 500 cfm, there was a total deposition of 44 grams of sodium. The average concentration of sodium in the aerosol during this period was 0.69 grams/m³ (toward the end of the period the sodium reservoir became depleted and the airborne concentration declined). This means that approximately 3000 grams of sodium passed through the coil during the experiment and that approximately 1.4% was retained in the coil. This is a small amount compared to the total coil volume, but as the photograph in Figure 3 indicates, deposition occurred principally on the upstream face of the coil and eventually formed a

FIGURE 1

Arrangement of apparatus for deposition tests.

FIGURE 2

Cooling coil air flow resistance as sodium particles deposit between fins.

Figure 3. Face of finned cooling coil showing sodium deposits.

substantial obstruction to flow.

B. Deposition in Tube-type Heat Exchangers

The above studies, combined with other considerations, resulted in design changes in the closed loop cell cooling system. A shell and tube heat exchanger with coolant in the shell was substituted for the finned tube cooler. The proposed shell and tube cooler contains 2600 0.56 inch I.D. gas flow tubes, each 8-ft. long. A 3-tube assembly of 0.53 in. I.D. tubes, 8-ft. long was constructed as a heat exchanger simulator for sodium aerosol deposition studies. Figure 4 shows the experimental arrangement. Aerosol was withdrawn from the chamber, passed through the tubes at the design flow rate of 90 fps, and blown back into the chamber through a second wall port. Sodium deposition was monitored by continuously observing the increase in the pressure drop of the tubes at constant volumetric flow rate (i.e., as the tube openings became narrowed by deposition, tube velocity increased). However, as the experiment progressed, the maximum static pressure capability of the recirculating blower was reached and from that time onward, when the experiment was continued, volumetric flow rate declined at constant pressure drop across the tubes. Clean tubes at design flow rate have a pressure drop of 11.4 in.w.g. Maximum measured pressure drop after deposition was 21.1 in.w.g.

Table 1 summarizes three test series. In each case, the chamber was totally filled with a sodium cloud before starting the recirculating blower that induced flow through the heat exchange tubes. For test number 1, two pounds of sodium were evaporated into the chamber gas in about 50 minutes. This produced very high particle concentrations (up to 6.8 grams, as Na, per cubic meter) and vigorous agglomeration. Visible suspended particles ($> 100\mu\text{m}$) were produced. Gas volume rate began to decline after 21 minutes and the experiment was terminated after 50 minutes. Figure 5 shows the tube inlet at the conclusion of the test (with an unused tube bundle for comparison) and Figure 6 shows the outlet. Figure 5 shows inlet deposition of sodium to form a pyramidal inlet contour but the tubes are open to almost their full diameter. This is shown more clearly in Figure 7 which is the inlet end after the deposit on the tube sheet had been carefully removed (again with a clean tube for comparison). Figure 8 is a similar photograph of the outlet end. (Please note that prior to the photograph we disturbed the sodium deposit in attempting to measure its thickness). In both cases, the deposit on the tube walls is obvious. At the inlet end it was about 0.05 in. thick; at the outlet, about 0.02 in. Thirty grams of sodium (as Na) were recovered from the interior of the three tubes after removal of the surface deposits on the inlet and outlet tube sheets.

Tests 2 and 3 were conducted with aerosols containing about one order of magnitude less particle loading. For test number 2, the run was halted after 69 minutes when the blower was no longer able to maintain the design gas flow rate. Total deposit in the tubes was 17.6 grams, as Na. Test number 3 was run 228 minutes. Again, it was possible to maintain rated flow for 69 minutes; but after that, flow declined steadily until, at the end, it was only 18% of the design value and pressure drop across the tubes was 21.1 in.w.g. at this low

Figure 4. Experimental arrangement for studies of sodium deposition in 0.53-in. tubes.

1. Sodium aerosol chamber	8. Valve
2. Inlet pipe	9. Recirculating blower
3. Cascade impactor	10. Chamber gas drier
4. Thermocouple	11. Cooler tube pressure taps
5. Tube bundle	12. Pressure drop monometer
6. Oxygen monitor	13. Set of replacement tubes
7. Flow meter	

Figure 5. Entry tube sheet before and after sodium deposition

TABLE I
SODIUM DEPOSITION IN 0.53-in. I.D. HEAT EXCHANGER TUBES

	Test #1 1/13/72	Test #2 1/18/72	Test #3 1/21/72
Loading - gm/M ³			
Maximum	6.8	0.90	1.12
Minimum	.26	0.24	0.21
Average	4.4	0.48	0.72
Running Time - Min.	71	69	228
Total Deposit in Tubes - gm Na	30	17.6	32
Velocity ft/sec.	92 for first 21 min.	90	90 for first 69 min.
	92-81 over last 50 min.		90-16 over last 159 min.
Temperature °F			
Outside tube	77	73	75
Inside tube	95	85-89	90
Pressure Drop in. of water			
Maximum	18.0	20.3	21.1
O ₂ - %			
Before test	1.4	1.1	1.2
After test	1.1	1.1	--
Water vapor lb. water/lb. N ₂			
Before test	0.00146	0.0009	--
After test	0.00088	0.00029	--

Figure 6. Exit tube sheet after sodium deposition
Figure 7. Inlet tubes after removal of tube sheet deposit
Figure 8. Outlet tubes after removal of tube sheet deposit

flow rate. Figure 9 shows the inlet tube sheet at the conclusion of test number 3. It is obvious that the tubes are almost totally closed off. Figure 10 shows the entry tube sheet after the deposit at the entry has been removed. Again, it is clear that the deposit has almost completely plugged the tubes. Probing with a stiff wire indicated that the thick wall deposit extended 12 to 14 inches into the tube. Figure 11 shows the exit end of the tubes after the deposit on the tube sheet had been removed.

It is clear from these three tests that substantial wall deposition may be expected in 0.56 I.D. tubes at a tube velocity of 90 fps when the tubes are not protected by entry filters.

It is possible to deduce from the data in Table 1 that particle deposition occurs at a rate that is proportional to the sodium concentration in the aerosol and the time of operation. Other factors, such as gas temperature and particle size, shape, and specific gravity are important as well, but by generating the test aerosol as uniformly as possible and as closely similar to the predicted source term as possible, particle concentration and time of exposure remain as the principal deposition variables in the current series of experiments.

C. Deposition in a Centrifugal Recirculating Blower

It was proposed that blowers on closed loop cell cooling systems be placed upstream of the heat exchanger to remove some of the particulate sodium before it reaches the exchanger tubes. Therefore, two runs were conducted with the recirculation blower upstream of the heat exchanger tubes. In the first, conducted for 87 minutes, sodium aerosol concentrations averaged 0.08 gm/m^3 . This is about one order of magnitude less than aerosol concentrations of previous experiments and total deposition in the heat exchanger tubes was approximately one order of magnitude less, indicating the direct relationship between aerosol concentration and mass deposition rate in grams per minute. During this period, only 1.8 grams of sodium (as Na) deposited in the tubes and airflow resistance increased 1.5 in.w.g. but a total of 29 grams of sodium (as Na) deposited in the blower casing. In the second run, conducted for 140 minutes, the sodium aerosol concentration entering the blower was 1 gm/m^3 . Design flow rate, equivalent to 90 fps through the tubes, began to decline after 70 minutes and reached a low of 33 fps before the run was terminated. Tube deposition over the 140 min. exposure period totaled only 2.7 gm. This is approximately proportional to 1.8 grams deposited in 87 minutes during the previous run and demonstrates the proportionality of deposition with time of exposure for equal aerosol concentration flowing through the tubes. However, 54 grams (as Na) had deposited in the blower casing and almost completely plugged it with solid sodium oxide as shown in Figure 12a, the blower outlet, and Figure 12b, the blower inlet. Details of the two runs are summarized in Table 2.

From these two runs, it is evident that placing the recirculation blower upstream of the heat exchanger will, indeed, reduce the deposition load on the heat exchanger but only at the risk of rapid blower failure by sodium deposition in the blower passages.

Figure 9. Inlet tube sheet at end of test 3.

Figure 10. Inlet tube sheet at end of test 3 after removal of entry deposit.

Figure 11. Outlet tube sheet at end of test 3 after removal of exit deposit.

Figure 12. Recirculation blower upstream of heat exchanger tubes after 140 min. exposure to 1 gm/m³ sodium oxide aerosol. a. outlet; b. inlet.

Figure 13. Cleaning caked dust from heat exchanger tube with compressed nitrogen pulse (calcium carbonate).

TABLE 2

TWO DEPOSITION RUNS WITH RECIRCULATING BLOWER
UPSTREAM OF HEAT EXCHANGER TUBES

	1/28/72 Test #1	2/8/72 Test #2
Loading at tube entrance (gm Na/M ³)		
Maximum	0.10	0.28
Minimum	0.033	0.06
Average	0.08	0.16
Loading at fan entrance (gm Na/M ³)		
Average	--	1-2
Running time - min.	87	143
Total deposit in tubes (gm Na)	1.8 calc. (2.5)	2.7
Total deposit in blower (gm Na)	29	55
Tube velocity-ft/sec.	90	90 for first 70 min. 90 to 50 for last 73 min.
Temperature °F		
Outside tube	75	75
Inside tube	102	104
Pressure drop in. of water		
Maximum	12.9	(4.5 at end)
O ₂ -%		
Before test	1.3	1.9
After test	1.1	--
Water vapor (lb. water/lb. N ₂)		
Before test	.0009	.00014
After test	--	--

From previous experiments, it is clear that with the blower downstream of the tubes, failure of the heat exchanger may occur from excessive deposition if the aerosol concentration reaching it is abnormally high or aerosol flow is abnormally prolonged. Therefore, methods of removing sodium deposits from heat exchanger tubes while the unit is in normal operation were investigated as an alternative to filtration.

D. Tube Cleaning during Normal Service

Tube cleaning tests were conducted first in normal air using finely ground calcium carbonate as a sodium oxide simulant. It was found that short (1-sec) bursts of compressed nitrogen (150 psi) introduced at the exit end of the tube and directed upstream were effective in removing deposited dust. Figure 13 shows dust being discharged from a tube during a simulation test. Next, similar static tests were conducted with tubes in which freshly formed sodium oxide had deposited by standard exposure methods described previously. Tubes were cleaned with one, two, and four successive one-second compressed nitrogen blasts. The expelled sodium was collected in filter bags and the residual sodium washed out. Both fractions were analyzed for total sodium and gave the following results:

Cleaning cycles of 1-sec at 125 psi	Amount Na blown out of tube (gm)	Amount Na remaining in tube (gm)	Total Na deposited (gm)
1	0.94	4.6	5.7
2	2.2	3.5	5.6
4	3.8	0.9	4.7

Next, dynamic cleaning tests were conducted using one-second compressed nitrogen blasts each time the heat exchanger tubes reached an elevated airflow resistance signifying excessive sodium oxide deposition. The three compressed nitrogen jet tubes installed in the discharge header of the experimental heat exchanger tube bundle are shown in Figure 14a. Figures 14b and 14c show the inlet and outlet ends of the heat exchanger tubes at the conclusion of a multi-cycle run during which the pressure drop through the tubes (and hence the thickness of the sodium coating which affects heat exchange rate) had been well controlled by occasional nitrogen blasts. The nitrogen jet tubes shown in Figure 14a had been removed prior to recording Figure 14b. These tubes were centered on the tube axes and mounted approximately 0.1 inch back of the tube end. Figure 15 shows the compressed nitrogen cylinder, pressure reduction valves, pulse timer, and external attachments to the blast cleaning tubes. Table 2 summarizes the details of two runs using repeated dynamic tube cleaning gas blasts during continuous operation. It was found that 150 psi pressure was not adequate for satisfactory tube cleaning under dynamic conditions and the pressure was raised to 250 psi. Total nitrogen released during a 1-second blast at 150 psi was 6.4 liters; at 250 psi, 13.6 liters; both at NTP. Two runs of approximately

Figure 14 a. Nitrogen jets at downstream end of heat exchanger tube bundle.
b. Discharge end of tube bundle
c. Entry end of tube bundle.

Figure 15. Jet tubes attached to compressed nitrogen cylinder, showing jet pulse timer and pressure reduction valves.

200 minutes were conducted at sodium concentrations averaging 1.3 and 4.2 grams/m³ without significant rise in tube resistance. After each of the 8 cleaning cycles of Run 2 in Table 3, tube resistance was lowered to a value close to clean starting resistance and it seemed probable that this run could have carried on indefinitely with similar results. Lower nitrogen gas pressures (and less total nitrogen consumption) would be equally effective if the recirculating blower could be shut down for a few minutes during the cleaning process.

III. Deposition Theory

Neither the phenomenon of turbulent particle deposition on surfaces nor particle reentrainment due to the action of the turbulent motions of the carrying stream has yet been fully elucidated. A review of the recent deposition theories and experimental studies and also of the less well understood subject of particle reentrainment reveals accuracies at best to within about one order of magnitude, and many times much less than this. Even the various theories which have been advanced lack agreement. This could be due, in part, to differing assumptions and simplifications of the various models, disagreement of the correct experimental methods to be used and errors in particle generation and sizing, flow and concentration measurements and undetected reentrainment.

All deposition models are considered only for a monodisperse aerosol, radial inertial and diffusive removal mechanisms, and the deposition surface as a perfect sink. Many authors assume the equality of the eddy diffusion coefficient of fluid momentum transfer, ϵ , and the eddy diffusion coefficient of material transfer, D_E . From all indications this assumption seems to be an over simplification. The assumption is also made that particle transport from the bulk fluid to the region next to the walls is very rapid with respect to particle transport to the wall. Then particle deposition is assumed to take place by a 'free flight' mechanism whereby the particle traverses the final boundary layer in projectile fashion. All the models differ, however, in their prediction of the starting point of the particle's free flight and its velocity.

Sehmel (3) tested by statistical methods four published models and one of his own against results calculated from experimental deposition velocities. These statistical tests showed that the five models did not adequately represent the experimental data.

A review of different experimental investigations show that in their presentation some authors omitted necessary information for data comparison. For example, such important parameters as geometric standard deviation, agglomeration rate, surface stickiness and roughness, thermophoretic and electrostatic effects, and inlet and outlet effects many times are not mentioned. Therefore, it is difficult to evaluate the different models while at the same time comparing the experimental results with each other, in order to make deposition predictions within a reasonable error.

Despite a data scatter of one order of magnitude, two empirical equations, derived from the experiments of four workers, seem to give

TABLE 3

COMPRESSED NITROGEN HEAT EXCHANGER TUBE CLEANING
DURING CIRCULATION OF SODIUM OXIDE AEROSOLS

(all cleaning by three 0.02 min. blasts each tube at
250 psig* unless noted otherwise)

Loading at tube entrance (gm Na/M ³)	2/29/72	3/3/72
	Exp. # 1	Exp. #2
Maximum	1.9	6.2
Minimum	0.76	0.72
Average	1.3	4.2
Velocity-ft/sec	90	90
%O ₂		
Before experiment	0.99	1.25
After experiment	1.11**	1.15
Running time-min.	192	156
Deposit in tubes at end (gm Na)	10	2.2

Experiment #1					Experiment #2			
Clean- ing Cycle	Deposit Time (min)	Pressure Before Clean- ing	Drop (in.w.g.) After Clean- ing	Diff.	Deposit Time (min)	Pressure Before Clean- ing	Drop (in.w.g.) After Clean- ing	Diff.
1	77	19.2	16.0	3.2 (150 psi)	32	16.25	12.9	2.35 (2 blast
2	25	19.7	17.6	2.1 (150 psi)	17	15.65	11.85	3.8
3	10	19.6	15.2	3.4 (200 psi 2 blasts)	14	15.3	11.9	3.4
4	28	19.2	12.2	7.0	15	14.3	11.0	3.3
5	44	16.1	11.9	4.2 (2 blasts)	10	14.5	10.6	3.9
6					17	14	11.1	2.9
7					11	13	11.1	1.9
8					10	12.7	10.7	2.0

*At 150 psi = 6.4 l per blast per tube
At 200 psi = 10.8 l per blast per tube
At 250 psi = 13.6 l per blast per tube.

**Air leak into negative pressure side of blower.

the most satisfactory results in predicting turbulent deposition. These equations come from Sehmel's semi-empirical effective eddy diffusion model and are as follows:

$$K = 1.47 \cdot 10^{-6} \cdot \rho_p^{1.01} \cdot d_p^{2.1} \cdot \left(\frac{D \cdot U}{\nu}\right)^{3.02} \cdot \left(\frac{f}{2}\right)^{0.5} \cdot U$$

and

$$K = 1.10^{-17} \cdot \rho_p^{1.83} \cdot d_p^{2.99} \cdot \left(\frac{D \cdot U}{\nu}\right)^{3.08} \cdot \left(\frac{f}{2}\right)^{0.5} \cdot U$$

where

K = deposition velocity

ρ_p = density of particles

d_p = ratio of particle diameter in μm to duct diameter in cm.

D = duct diameter

U = average flow velocity

ν = kinematic viscosity

If the state-of-the-art of turbulent deposition theories is little developed, then an adequate theory to account for reentrainment does not agree with reality in some cases as it predicts no reentrainment for 20 to 10 μm particles under conditions where it has been observed experimentally. A kinetic model cannot be applied because of the lack of knowledge of the particle removal velocity profile within the laminar sublayer.

Experimental results are somewhat confusing. It is possible to maintain a certain rate of reentrainment despite increasing deposition due to increasing Reynold's number or particle size. The fact that particles as small as 2 μm can be removed in turbulent reentrainment (4) is not sufficient for the prediction of quasistationary deposition-removal conditions. In addition, there appears to be no data available on friction forces of deposited layers which would permit prediction of reentrainment by breaking up of whole chunks of deposit. Thus, there is little available for application to the practical problem at the present time.

Nonetheless, the information was applied to the specific problem of deposition and reentrainment of sodium aerosols in a turbulent flow of an inert atmosphere which passes through a shell and tube heat exchanger. It is expected that this should give results accurate to within one order of magnitude.

In comparing deposition models with experimental observations, a decrease of tube diameter caused by the deposition layer has not been considered. This may be why calculated values of K_{cal} underpredict measured deposition velocities, recognizing that for constant flow rates, Reynolds number, average flow velocity U , and the diameter

ratio, d_g , increase with decreasing open tube diameter.

Therefore, to calculate K_{cal} for a three tube assembly of 1.346 cm I.D. tubes, a mean diameter D_m during deposition was defined. Assuming a linear decrease of Δs_m from the inlet to the outlet, we find the mean diameter at $x = L/2$ and $t = \Delta t/2$:

$$\frac{\pi}{4}(D^2 - D_m^2) = \frac{\pi}{4}(D_m^2 - D_{1/2}^2)$$

where

$$D_{1/2} = \frac{D_1 + D_o}{2}$$

D_1 and D_o were determined by analyzing photographs taken of inlet and outlet ($D_1 = 0.99$ cm; $D_o = 1.07$), following a deposition run.

The deposition velocity is determined by

$$K_{exp} = \frac{m_{exp}}{n_t \cdot (\pi \cdot D_m \cdot L) \cdot C_{av} \cdot \Delta t}$$

where m_{exp} is the total deposited mass.

The average density of the depositing sodium aerosol is taken as the bulk density of Na_2O . Comparing predicted and experimental values of K_D and $K_{D,cal}$, the close agreement was unexpected (see Table 4). For test number 2, deposition has been calculated for initial conditions of open tube diameter $D_o = D$ and flow velocity U and secondly for average conditions with constant flow rates. With a few additional runs, the validity of the design equations could be checked, although for the purposes of this study the number of runs made was adequate. Accurate results require accurate determinations of the mass median diameter, MMD, and the deposition layer determining D_1 and D_o . In our recent deposition studies, we found a layer density of the deposit P_l of about 0.17 g/cm³, assuming a linear increase of the open tube diameter from the inlet to the outlet. The experimental set up of the study did not permit us to measure reentrainment, but there is some evidence that reentrainment did not occur during the tests reported:

1. Using Schmelt's relationship 35 for reentrainment, the critical distance y_a^* is less than 0.5 in all cases (see Table 4).

2. The good agreement between experimental and calculated deposition was obtained by using design equation #1 and assuming no reentrainment.

TABLE 4 - Recent Sodium Studies

Test No.	$\rho(\text{g/cm}^3)$	MMD(μm)	σ_g	$\bar{U}(\frac{\text{cm}}{\text{sec}})$	$D_m(\text{cm})$	$v(\frac{\text{cm}}{\text{sec}})$	$K_{\text{exp}}(\frac{\text{cm}}{\text{sec}})$	$K_{\text{the}}(\frac{\text{cm}}{\text{sec}})$	$K_{D,\text{exp}}(\frac{\text{cm}}{\text{sec}})$	$K_{D,\text{cal}}(\frac{\text{cm}}{\text{sec}})$	$\frac{K_{D,\text{exp}}}{K_{D,\text{cal}}}$
2	0.57	4.12		2740	1.346	0.1682	3.58	0.007	3.57	1.76	2.03
2	0.57	4.12		3720	1.21	0.1682	3.58	0.007	3.57	3.74	0.95
4	0.57	4.12		2740	1.346	0.1682	1.32	0.017	1.30	1.76	0.74

Test No.	$m_{\text{exp}}(\text{g})$	$C_{\text{av}}(\text{g/cm}^3)$	$\Delta t(\text{sec})$	$m_{\text{cal}}(\text{g})$	y_d^+
2	17.6	0.43	4140	9.73	0.382
2	17.6	0.43	4140	18.54	0.426
4	1.8	0.08	5220	2.30	0.382

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PAPER NUMBER TWO

THE EFFECT OF HIGH PRESSURE AND LOW TEMPERATURE
ON THE ADSORPTION OF XENON AND KRYPTON FROM
HELIUM AND ARGON STREAMS

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I. Introduction

Adsorption on charcoal is commonly used to remove rare gas fission products from reactor off-gases and cover gases. Systems now in use operate at a number of different temperatures from as low as -150°C to about 50°C , but most of them are used at or near atmospheric pressure. Information is needed on the adsorption capacity of charcoal at high pressures either for the design of holdup beds or for the evaluation of the safety hazards of pressure buildup in atmospheric pressure systems.

This investigation was initiated to study the simultaneous effect of high pressure and low temperature on the adsorption of krypton and xenon from argon and helium carrier gas streams. Forster⁽¹⁾ has reported that the dynamic adsorption coefficient of krypton from argon streams increases monotonically at temperatures as low as -50°C , but it was thought that competition between carrier and fission gas might result in increased pressure causing a decrease in adsorption capacity at very low temperatures⁽²⁾. Accordingly experiments were done at temperatures extending below the critical point of argon (-122°C) and as shown below, increased pressure does indeed cause decreased adsorption capacity at -140°C .

II. Apparatus

A schematic diagram and photograph of the system are shown in Figures 1 and 2. The adsorption column was a short length of iron pipe containing Pittsburgh PCB charcoal. In most of the experiments, the column was 2" long by 5/8" i.d. (5.08 x 2.19 cm) although smaller or larger columns were used for some experiments. The mesh size was usually 6 x 16 but here too, a few experiments were done with finer material. The results did not seem to be affected by variations in mesh size. The temperature within the bed was measured with a steel clad copper-constantan thermocouple inserted into the center of the column along the axis. The potential produced by the thermocouple was measured with a Leeds and Northrup potentiometer using an ice bath as the reference. The thermocouple voltage was measured several times during a run and the average voltage was converted to a temperature by interpolating in a calibration table. During a run, the thermocouple voltage rarely varied by more than 0.01 mv which corresponds to a temperature variability of about 1°C . After the column had been packed with charcoal, it was baked out at 110°C for several hours to remove moisture.

The constant temperature chamber was a styrofoam box into which liquid nitrogen was sprayed. A solenoid valve on the liquid nitrogen reservoir was turned on and off by a thermostatic controller which sensed the temperature in a chamber with a copper-constantan thermocouple. The temperature in the chamber varied by a few degrees during a run, but the thermal mass of the column smoothed out these variations. Gas being fed to the column was precooled by passage through a ten-foot coil of copper tubing.

FIGURE 1. DIAGRAM OF TEST APPARATUS.

Figure 2. Apparatus for determining adsorption coefficients at elevated pressures and reduced temperatures. *1-nitrogen spray head; *2-adsorption column; *3-carrier gas precooling coil; -- 1-liquid nitrogen source; 2-carrier gas source; 3-pressure gauge; 4-injection system; 5-low temperature chamber; 6-low temperature controller; 7-potentiometer for column thermocouple; 8-ionization chamber; 9-wet test gas meter; 10-electrometer for ionization chamber.

*Close up view.

Carrier gas pressure was controlled with a Matheson high pressure regulator and measured with a Bourdon gauge. No variations in pressure were noted during a run. The accuracy of the pressure gauge was judged to be about 0.5 psi. Gas flow was controlled by a miniature needle valve at the outlet to the column. Total gas volume was read from the wet test meter at intervals and the flow rate was calculated from the time-volume data. The flow rate was found to be constant to within the accuracy of the measuring equipment.

The concentration of radioactive krypton or xenon in the carrier stream was measured by passing it through a 500 ml spherical ionization chamber which had a DC potential of 127 volts across it. The ionization current which is directly proportional to the activity of the gas stream was measured with a Cary vibrating reed electrometer.

In order to avoid injecting samples into the pressurized system, a bypass was set up that allowed carrier gas to continue flowing under pressure while the sample was injected at atmospheric pressure. Then valves were switched to pick up the sample in the pressurized stream.

III. Procedure

Before injecting a sample for a run, the temperature, pressure, and flow rate were stabilized. The flow rate was adjusted to 0.3-1.5 liters per minute although it has been shown⁽³⁾ that flow rate is not critical so that no attempt was made to set it at a particular value.

Radioactive krypton and xenon were obtained as solution in physiological saline (they are also used as diagnostic reagent in lung disease). 0.1 ml of the solution was drawn into a 1 ml syringe and the needle was then inserted into the injection septum. About 0.9 ml of carrier gas was drawn into the syringe and gas was freed from the solution by agitation. The gas phase was then injected. A drying agent in the gas line removed traces of moisture before the sample reached the adsorption bed.

Every minute or so after the injection, the electrometer current and elapsed time were noted. Soon after injection, the electrometer current rose rapidly to a peak value and then decreased exponentially. Since the exponential decay was so regular, the electrometer current was recorded only until it reached 1-5% of the peak value. It was then extrapolated to the point where it was less than 1 ma.

IV. Data Analysis

All of the data from a run was punched onto IBM cards for analysis by computer. A program was written which calculated the average temperature and flow rate, converted the elapsed time readings to volumes and wrote the entire record of an experiment

on magnetic tape for later analysis. Dynamic adsorption coefficients (4) were calculated from the elution curves by moment analysis.

V. Results

The adsorption of xenon from argon or helium streams was studied only at 38°C. At temperatures below zero, the adsorption coefficients were so high that complete elution curves could not be obtained within one working day. The results are shown in figure 3. The straight lines drawn through the sets of points were placed visually. With krypton, experiments could be run at temperatures as low as -160°C although -140°C was the lowest temperature used in routine experiments. Argon boils at -186°C at one atmosphere and at -160°C at seven atmospheres. In normal operation, the cold chamber was about twenty degrees colder than the adsorption bed, and when the column was operated at much below -140°C, argon would occasionally condense in the precooling coil and cause erratic pressure variations. The lowest temperature used with helium as the carrier gas was -78°C. Below this point, the adsorption coefficient became so large that complete elution curves could not be obtained in a reasonable time.

Plots of the adsorption coefficients vs. the reciprocal of the absolute temperature are shown in figures 4 and 5. The plot shown in figure 4 was obtained by least squares analysis of the results of 116 experiments using argon as the carrier gas. Actually different regression lines should have been obtained for different pressures, but they were indistinguishable within the accuracy of the experiments. With helium as the carrier gas, the adsorption coefficients varied much more with pressure so that separate plots could be constructed for each pressure as shown in figure 5.

Figures 6 and 7 show the effect of pressure on the adsorption of krypton from helium and argon streams at various temperatures. In the case of argon as the carrier gas, the coefficients have been corrected for temperature; with helium, these corrections were found to be unnecessary. Also the results shown for -140°C were obtained with charcoal with a lower adsorptive capacity. The charcoal used for the other runs showed similar behavior but fewer points were obtained. Several differences between the two sets of curves should be noted:

1. The adsorption coefficients are uniformly higher when helium is used as the carrier gas.
2. The effect of pressure is much more marked with helium. For instance with argon at -40°C a pressure change from 20 psig to 120 psig causes a 45% increase in adsorption coefficient while with helium at -47°C a similar pressure change causes a 400% increase.
3. In the higher temperature ranges (above -50°C) where comparable data was obtained for both carriers, the slope of the adsorption coefficient vs. pressure line is a strong function

FIGURE 3. EFFECT OF PRESSURE ON THE DYNAMIC ADSORPTION COEFFICIENT OF KRYPTON ON PCB 6X16 MESH CHARCOAL AT 38°C.

FIGURE 4. EFFECT OF TEMPERATURE ON THE DYNAMIC ADSORPTION OF KRYPTON ON CHARCOAL FROM ARGON STREAMS.

FIGURE 5. EFFECT OF TEMPERATURE ON THE ADSORPTION OF KRYPTON ON CHARCOAL FROM HELIUM STREAMS.

FIGURE 6. DYNAMIC ADSORPTION COEFFICIENT OF KRYPTON ON P.C.B. 6 X 16 MESH CHARCOAL FROM ARGON CARRIER STREAMS (-140°C RUNS DONE ON CHARCOAL WITH LOWER ADSORPTIVE CAPACITY).

FIGURE 7. DYNAMIC ADSORPTION COEFFICIENT OF KRYPTON ON PCB 6 X 16 MESH CHARCOAL FROM HELIUM CARRIER STREAMS.

of temperature with helium whereas temperature changes have little effect with argon.

4. Perhaps the most important observation that can be made is that the adsorption coefficient of krypton from argon streams increases with pressure at temperatures above -120°C ; is almost unaffected by pressure at -120°C , and at -140°C actually decreases markedly over the range of 15-75 psig.

VI. Discussion

The results reported here generally confirm those obtained by Forster⁽¹⁾. He observed the effect of temperature on the dynamic adsorption coefficient and also that temperature had a greater effect with helium as the carrier gas. He also notes that the coefficient increases with pressure although the slope of his curve is higher than that reported here.

It seems significant that the slope of the adsorption coefficient - pressure curve changes from positive to negative around -120°C . The critical temperature of argon is -122°C which means that below this temperature argon is capable of existing as a liquid. According to the BET theory, multilayer adsorption is possible only below the critical temperature and according to Dubinin, the adsorbed phase can be considered to be similar to a liquid below the critical point. Thus above -122°C we are probably observing phenomena associated with the adsorption of krypton and argon on bare carbon surfaces while below this point the situation is more akin to gas-liquid partition. In this region, the parameters that will be needed for a theoretical examination of the data presented here are the solubilities and heats of solution of krypton and xenon in a phase which is similar to, but certainly different from, pure liquid argon.

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