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ABSTRACT

A study of the adsorption of HF and UF₆ on NaF pellets in the presence and absence of nitrogen diluent was made. The adsorption reaction was studied in an adiabatic bed by means of thermocouples placed at known heights.

Equations were written for the numerical calculation of compositions and adsorption rates as functions of height and time in the bed. Data from nine experiments on HF is available for numerical analysis.

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TABLE OF CONTENTS

	Page
1.0 Introduction	3
2.0 Mathematical Analysis	3
2.1 HF-N ₂ Mixtures	5
2.2 100% HF	6
2.3 UF ₆ -N ₂ Mixtures	7
3.0 Calculations	7
3.1 Gas and Solid Composition and Adsorption Rate	7
3.2 Vertical Heat Conduction in Bed	8
3.3 Temperature Difference Between Gas and Solid	8
3.4 Adiabatic Reaction Temperature	9
4.0 Operating Techniques	11
4.1 Absorber Bed Preparation	11
4.2 Experimental Procedure	12
4.3 Regeneration of Bed	13
4.4 Miscellaneous Precautions	13
5.0 Experimental Data	13
5.1 NaF Pellets	13
5.2 NaF Bed	13
5.3 Gas Flow Rates	14
5.4 Results	14
6.0 Conclusions	15
7.0 Acknowledgement	15
8.0 References	15
9.0 Appendix	16
9.1 UF ₆ Experimental Apparatus and Operation	16
9.2 Suggested Operational Changes	16

1.0 INTRODUCTION

The volatility process has been proposed for reprocessing molten salt reactor fuels containing uranium. One step in the process involves the adsorption and subsequent desorption of UF_6 from NaF pellets in fixed beds. Rational design of these beds and their cyclic operation will be based on an understanding of the phenomena involved. With these needs in mind, this experiment was set up to obtain fundamental data on the kinetics of HF and UF_6 adsorption on NaF pellets. The results of this experiment will be used in the optimum design and operation of NaF adsorption beds.

2.0 MATHEMATICAL ANALYSIS

The results of the adsorption processes occurring in a NaF bed depend on the chemical characteristics of the adsorption reaction and the physical properties of the bed and gas phase. The separate chemical adsorption processes may be represented:

(a) For HF:

$$\text{with } \log p_{HF} = 8.619 - 3.16 \times 10^3/T$$

where p is in mm Hg and T in $^{\circ}K$

(b) For UF_6 :

$$\text{with } \log p_{UF_6} = 10.88 - 5.09 \times 10^3/T$$

where p is in mm Hg and T in $^{\circ}K$

Rates of these adsorption reactions are high and difficult to measure in the range of 25° to $150^{\circ}C$. Marvin Whatley proposed that the adsorption be carried out in an adiabatic bed and that the adsorption rates and gas and solid compositions in the bed be calculated from a measurement of the temperature gradients in the bed. The development of the necessary equations for the calculations is based on differential material and energy balances as follows.

Consider a cylindrical fixed bed of NaF pellets in which thermocouples are placed at known depths on the center-line of the bed. The center portions of the bed are assumed to operate adiabatically since the thermal conductivity of the bed is low and the bed container is preheated to average bed temperature just before use. Plug flow is assumed for the gas and radial temperature and concentration gradients are neglected. The symbols used are:

A = cross-sectional area of bed, cm^2

C_g = heat capacity of gas, $\text{cal/g mol } ^\circ\text{C}$

C_s = heat capacity of solid

$C_{\text{NaF}}, C_{\text{HF}}, \text{etc.}$ = heat capacity of pure materials

G = gas flow in bed, $\text{g mols mixture/min}$

h = bed depth, cm

ΔH = heat of adsorption reaction, $\text{cal/g mol absorbed}$

I = inert gas flow in bed, g mol/min

k = thermal conductivity of bed, $\text{cal/cm min } ^\circ\text{C}$

S = $\text{g mol NaF/cm bed height}$

t = time, min

x = solid composition, mol HF/mol NaF

y = gas composition, mol fraction

Two independent variables (h, t) are needed to describe the behavior of all points in the bed, hence the material and energy balances result in partial differential equations.

2.1 HF-N₂ Mixtures

The equations are given below for adsorption of HF from HF-N₂ gas mixtures. Nitrogen is referred to as an inert (I).

1. Material balance. A material balance on the inert gas gives a relationship between G and y at any point in the bed, thus

$$I = G(1-y) \quad (1)$$

A similar balance on the amount of adsorbate transferred from the gas to the solid in a differential section of the bed may be written,

$$S \frac{\partial x}{\partial t} = - \frac{\partial}{\partial h} (G y) \quad (2)$$

Equation 1 and 2 may be combined to give either

$$-S \frac{\partial x}{\partial t} = \frac{I}{(1-y)^2} \frac{\partial y}{\partial h} \quad (3)$$

or

$$-S \frac{\partial x}{\partial t} = \frac{\partial G}{\partial h} \quad (3a)$$

2. Energy balance. An energy balance on a differential section of the bed may be written as follows assuming adiabatic conditions and that the temperatures of the gas and the solid are identical.

$$\frac{\partial}{\partial t} (SC_s T) = -S \frac{\partial x}{\partial t} \Delta H - \frac{\partial}{\partial h} (GC_g T) - kA \frac{\partial^2 T}{\partial h^2} \quad (4)$$

In the absence of heat capacity data for the solid it is convenient to assume that

$$C_s = C_{NaF} + x C_{HF}$$

from which one finds that

$$\frac{\partial C_s}{\partial t} = C_{HF} \frac{\partial x}{\partial t} \quad (5)$$

Likewise,

$$C_g = C_I (1-y) + C_{HF} y$$

and

$$\partial C_g / \partial h = (C_{HF} - C_I) \partial y / \partial h \quad (6)$$

The heat capacities are assumed to be independent of temperature. Now equation 4 may be expanded as indicated and combined with equations 3, 5, and 6 to give $\partial y / \partial h$ as a function of the heat of adsorption, the bed temperature gradients (which are measurable), and the solid and gas properties, thus

$$\begin{aligned} \partial y / \partial h & \left[- \frac{GT}{S} (C_{HF} - C_I) + \frac{I}{S(1-y)^2} (\Delta H + T(C_{HF} - C_g)) \right] \\ & = C_s \partial T / \partial t + \frac{GC_g}{S} \frac{\partial T}{\partial h} + \frac{kA}{S} \frac{\partial^2 T}{\partial h^2} \end{aligned} \quad (7)$$

Under conditions of the experiment $C_{HF} = C_I = C_g$ and the above equation reduces to a simpler form, either

$$\partial y / \partial h = \frac{(1-y)^2}{I\Delta H} \left[S C_s \frac{\partial T}{\partial t} + \frac{C_g}{(1-y)} \frac{\partial T}{\partial h} + kA \frac{\partial^2 T}{\partial h^2} \right] \quad (8)$$

or

$$\frac{\partial x}{\partial t} = \frac{-1}{S\Delta H} \left[SC_s \frac{\partial T}{\partial t} + \frac{C_g}{(1-y)} \frac{\partial T}{\partial h} + kA \frac{\partial^2 T}{\partial h^2} \right] \quad (9)$$

Now the combination of equation 3 with either equation 8 or 9 is sufficient to determine the concentrations (x,y) in the bed and the adsorption rate, $S\partial x / \partial t$ throughout the bed.

2.2 100% HF

From a theoretical standpoint it may be desirable to study the adsorption reaction using 100% HF gas since this should eliminate the resistance of the gas film to mass transfer. However, in this case the behavior of the N_2 originally in the bed, as it is displaced by the HF, is uncertain. Again equation 3a

holds for the material balance but equation 7 is not well behaved as y approaches unity. But the expansion of equation 4 combined with the assumption of equation 5, and the material balance equation 3a gives the desired result,

$$\partial G / \partial h = \frac{1}{\Delta H} \left[SC_s \frac{\partial T}{\partial t} + GC_{HF} \frac{\partial T}{\partial h} + kA \frac{\partial^2 T}{\partial h^2} \right] \quad (10)$$

Again it is possible to calculate the adsorption rate, the composition x , and flow rate G throughout the bed from the temperature gradients in the bed and the bed properties.

2.3 UF₆-N₂ Mixtures

Similar equations may be written for UF₆-N₂ mixtures but the stoichiometry of the reaction must be observed and the left side of equation 7 will not simplify unless dilute UF₆ feed gases are used as the terms are neglected in comparison to the magnitude of ΔH .

3.0 CALCULATIONS

3.1 Gas and Solid Composition and Adsorption Rate

Analytic solutions for equations 3 and 8 would be difficult to obtain and practical solutions are possible only by the use of a computer. For computation purposes the equations may be converted to finite difference equations by taking the first two terms of a Taylor's series to give the partial derivatives, thus,

$$\begin{aligned} \Delta h \partial y / \partial h &= y_{h,t} - y_{h-1,t} \\ \Delta t \partial x / \partial t &= x_{h,t} - x_{h,t-1} \end{aligned} \quad (11)$$

Initial conditions for the adsorption experiments were maintained as (a) y and $x = 0$ for all values of h when $t < 0$, and (b) constant feed gas composition for $t > 0$ at $h = 0$.

A few hand calculations were made from the data of the preliminary adsorption experiments to show the practicality of the calculation method. The calculations of the data from Bed 4 - Run 3 are placed with Notebook A-1388. These calculations showed the feasibility of numerical solution of the equations and the need for defining the finite difference values of the dependent variables as given in equation 11 so that the initial conditions of the experiment are satisfied.

L. E. McNeese is writing a program for this problem so that it can be run on the Oracle. The calculation will give values of x, y, and rate at all values of t and h for the bed.

3.2 Vertical Heat Conduction in Bed

Initially it was proposed to neglect vertical heat conduction in the bed in order to simplify the calculations. However, this is not generally justified because the hand calculations for Bed 4 - Run 3 show that the conduction term ($\partial^2 T / \partial h^2$ term) may be of the same magnitude as the $\partial T / \partial h$ term. This occurs when $\partial T / \partial h$ and $\partial^2 T / \partial h^2$ are the same magnitude since their coefficients are approximately the same, i.e.,

$$C_g / 1-y \approx kA$$

(Whatley had previously found that the thermal diffusivity of the bed, α , was $0.1 \text{ cm}^2/\text{min}$ which gives $k = 0.1 \rho C_s = 0.027 \text{ cal/cm min } ^\circ\text{C}$ and $kA = 0.027(81) = 2.2$.) Therefore, the conduction term should be included in the calculations.

3.3 Temperature Difference Between Gas and Solid

The heat of adsorption is initially released on the pellet surface and is either conducted into the pellet or transferred to the gas. An approximate relationship between the temperature difference of the solid and gas ($T_s - T_g$) and the bed depth, h, can be developed. The rate of heat transfer from the solid (where the heat is developed) to the gas (which removes it) may be equated to the rate of temperature increase of the gas, to give the maximum temperature difference expected at any point in the bed, thus

$$\frac{dq}{dh} = U \left(\frac{dA}{dh} \right) (T_s - T_g) = GC_g \frac{dT_g}{dh}$$

where T_s and T_g are the solid and gas temperatures, °C

U = heat transfer film coefficient taken as 0.04 cal/min cm² °C
(corresponding to 5 BTU/hr ft² °F)

G = gas rate in the bed taken as 1.2 g mol/min

C_g = heat capacity of the gas, 7 cal/g mol °C

dA/dh = interface area per cm bed, 1,000 cm²/cm bed

Integration of this equation gives

$$\frac{T_s - T_g}{T_s - T_{gi}} = \exp - \frac{dA}{dh} \frac{U h}{G C_g} = \exp - \frac{1000 (0.04) h}{1.2 (7.0)}$$

$$= \exp - 4.86 h$$

where T_{gi} is the initial gas temperature.

This indicates that more than 99% of any temperature difference in the bed would vanish in a distance of 1 cm from that point. The calculation appears to justify the assumption that $T_g = T_s$ in the bed.

3.4 Adiabatic Reaction Temperature

Consider runs made with 10 mol % HF. According to the vapor pressure relationship the maximum attainable temperature in the bed is about 192°C. The observed maximum temperature was 191-2°C for Bed 6- Run 7. The maximum adiabatic reaction temperature for this gas mixture is 225°C, therefore the initial adsorption rate is equilibrium controlled since the pellets cannot dissipate the heat of adsorption as rapidly as it is liberated. At later times there is a balance between the rate of adsorption and the rates of HF diffusion into the pellets and the rate of convective heat loss from the pellets.

The composition of the solid when it initially reaches maximum temperature may be estimated from an energy balance, i.e., the heat liberated by the adsorption reaction must be absorbed by the gas and the solid, thus for a 10% HF gas with complete adsorption of the HF,

$$x(16,400) = x(9)(7)(192 - 125) + (12 + 7x)(192 - 25)$$

$$x = 0.17 \text{ mols HF/mol NaF}$$

where the initial gas temperature is 125°C and solid temperature of 25°C. The first term on the right is the heat absorbed by the inert gas (assuming all HF was adsorbed) and the second term is that absorbed by the solid. From this calculation, one would expect the solid composition to rapidly reach 0.17 mols HF/mol NaF and then gradually increase to higher values eventually reaching unity if the run continues. Such behavior is typical of the two runs for which bed solids were analyzed (see pp. 56 and 72 in Notebook A-1388). Both beds gave x vs h curves in which x rapidly reached a plateau value of 0.06 to 0.07 and then it gradually increased to higher values (≈ 0.5) at the top of the bed. In these beds and others considered, the maximum temperature developed corresponds to the vapor pressure relationship for NaF·HF as tabulated below:

Table I

Maximum Temperature Developed in NaF Beds

Run	G_{N_2}	y	Temperature	
	g mol/min	mol % HF	Observed	Eq. Vapor Pressure
Bed 6 - Run 5	1.2	6.8%	175-8°C	185°C
Bed 6 - Run 7	1.2	10	191-2	192
Bed 7 - Run 1	1.1	20	213	216
Bed 6 - Runs 1 and 2	1.0	25	215-217	220

If a value of $x = 0.06$ corresponds to a monomolecular layer of HF on the NaF, the molecular area of HF would be 0.2 sq Ångstroms as shown

$$\text{Molecular area} = \left(\frac{42}{0.98} \right) \frac{1.3 (10^{10})^2}{0.06 (6.02 \times 10^{23})} = 0.2 \text{ sq Ångstroms}$$

The surface area of the pellets is 1.3 sq meters per gram and the pellets contain 98 wt % NaF. It is likely that the molecular area of HF is more than 100 times this value (data not available) so it is concluded that HF rapidly saturates, and maintains in, the surface region perhaps 100 multimolecular layers as the diffusion process attempts to distribute the HF in the bulk of the NaF pellet. As the pellets cool from the maximum temperature value the magnitude of x increases and should approach unity as a limit.

4.0 OPERATING TECHNIQUES

Much of the adsorber operation has been described in previous sections. Some repetition will occur but mostly the purpose here is to give necessary details for duplication of any HF adsorption experiment.

4.1 Adsorber Bed Preparation

The bare thermocouples are brought out the bottom of the bed and they must be insulated from the wire screen which supports the bed. NaF pellets are fused into the wire screen and two holes, each sufficient to pass a No. 30 wire, are drilled through the NaF which acts as an insulator. The NaF insulators are arranged around the edge of the screen.

Thermocouples pass out of the adsorber through a beeswax filled tube. Whenever thermocouples are to be replaced, the old couples are cut off and a fresh length of wire is pulled through the wax tube (while heating it) and stripped of insulation for a length of about 20 cm. The bare wires are threaded through the NaF insulators, cut to the proper length, twisted together for a 1/4" length at the ends, and silver soldered using a small wire of silver solder. During the soldering operation the tip of the thermocouple should point down so that any excess solder will flow to the tip. Finally the thermocouple tip is cut off leaving a soldered junction about 0.1" long having a small mass. The lengths of the thermocouples should be judged according to their position in the bed so that there is little excess wire in the bed to cause pellet dislocations which would interfere with the normal gas flow pattern in the bed.

The thermocouples should be placed in order (1, 2, 3, etc.) from the top or bottom of the bed to minimize time lost by the recorder in finding the temperature of each thermocouple. In forming the bed, the thermocouples are brought up vertically from their point of entry through the supporting screen to the elevation at which they are to be located, bent at right angles,

and the junction is placed in the center of the bed. NaF pellets are poured into the bed in a random manner and settled around and underneath the thermocouples. The thermocouples should have at least 1" of horizontal travel at their elevation point to minimize heat conduction from the junction along the wires. A thermocouple locator made up of a base ring, scale, and pointer is used to measure the elevation of any couple and its relation to the center of the bed. The base ring sets atop the adsorber and with the pointer and scale, the position of the thermocouple relative to the base ring may be measured. The complete bed is built up in this manner, one thermocouple placed about 1 cm above another, and the last thermocouple projects above the bed to measure the entering gas temperature.

Two trays of 1/2" monel Raschig rings are stacked above the pellet bed to increase turbulence in the entering gas. The bed is closed, carefully installed in the apparatus, and leak tested before making a run.

4.2 Experimental Procedure

In operating the facility, the following procedure was followed:

(a) Turn on the steam heater, the electric tapes and preheater and allow them to reach steady-state. Be sure that thermocouples brazed onto tubes read the tube temperature and not that of a contact heating tape. HF should be above 60-75°C when metered so as to avoid any uncertainty in its molecular weight. About 15 minutes before run time turn on the adsorber furnace set for 100°C. This value was selected as an average for any run and the furnace is used to decrease heat losses from the system.

(b) Decide on the HF and N₂ flow rate and adjust the N₂ pressure to 25 psig and open the HF lines to the rotameter value. The rotameter value cannot control the HF flow rate unless the HF tank pressure is appreciably greater than the gas manifold pressure.

(c) When everything is ready, start the bed temperature recorder and turn on the HF and N₂ at the proper rates. If desired the gases may be first by-passed to the clean-up trap while their composition is adjusted to the proper value and later turned on the bed. Due to the time-lag created by the volume of the lines and adsorber it is necessary to fix zero time for the adsorption run from the bed temperature record.

(d) If a thermocouple fails it must be temporarily "shorted out" so that the recorder will continue and read the other thermocouples on the instrument. For a few runs the initial temperature of the bed changed so rapidly that the recorder had difficulty measuring it.

4.3 Regeneration of Bed

The bed was regenerated by the following procedure:

(a) Turn the adsorber furnace up to 400°C and pass a stream of N₂ through the bed at a rate of 3-4 liters/min.

(b) Watch the temperatures in the bed and in Trap 1. Trap 1 picks up the HF driven from the bed and a decrease in the temperature of Trap 1 is an indication that HF has stopped coming into the bed. Regeneration may not be complete for HF unless the temperature of the bed goes above 270°C. The amount of HF caught in Trap 1 (found by weighing the NaF pellets before and after a run) may be used to check the calibration of the HF rotameter.

4.4 Miscellaneous Precautions

(a) Always check the adsorber and any new joints for leaks with soap solution.

(b) A portable millivoltmeter may be used to read adsorption bed temperatures above 300°C.

(c) A wet test meter should be used to calibrate rotameters. Both the HF and N₂ rotameters were newly calibrated for the reported experiments.

5.0 EXPERIMENTAL DATA

5.1 NaF Pellets

The pellets are 1/8" right cylinders having a bulk density of 1.02 gm/cm³ as placed in the bed ($S = 1.96 \text{ g mol NaF/cm bed}$). Their heat capacity is $10.4 + 0.00289 T^{\circ}\text{K}$ in cal/g mol °C (Perry). The surface area of the pellets by N₂ adsorption is 1.3 sq meters per gram. One hundred pellets weigh 3.91 gm. The pellets were obtained from the Harshaw Chemical Company.

5.2 NaF Beds

The bed is formed in a 4" i.d. cylinder with pellets in a random arrangement. Twelve thermocouples made of No. 30 B & S copper-constantan wire are placed in the center of the bed at various depths ranging from the gas above the

pellets to about 12 cm below the bed surface. The thermocouples are brought out the bottom of the bed through NaF insulators fused in the supporting wire screen. During a run the thermocouples are read about four times a minute by a fast electronic recorder. Immediately after a run, the bed is regenerated by heating at about 300°C while dry N₂ is passed through it.

5.3 Gas Flow Rates

Gas flow conditions are varied from 0.95 to 1.39 g-mol mixture per min to the bed (superficial gas velocity of 13 to 19 ft/min) with initial gas compositions that vary from 6.96 to 25.0 mol % HF. Some runs were made with 100% HF feed gas. The best runs are summarized on page 73 in Notebook A-1388. Inlet gas temperature to the adsorber was 125 to 130°C and the instantaneous gas temperature to the pellet bed is recorded on the recorder chart for the run. The rotameter gas pressure varied from zero to 3 psig.

5.4 Results

Each run is a record of the thermal response of the bed to a step concentration forcing function feeding the bed. The feed rate and adsorbate concentration in the feed are constant during a run. A run is stopped when the temperature increase is fully developed in the last thermocouple in the bed. Some runs were terminated sooner and samples of the bed pellets were analyzed for composition x so as to check the computer result for that run.

Construction details for each bed and the runs made on those beds are given in Notebook A-1388. A summary of the best runs is given on page 73 of the notebook. The temperature records for each run are with the notebook. In general, the temperature gradients in the bed are expected to be in this range, depending on the flow rate and the HF concentration:

$$\begin{aligned}\partial T / \partial t &= 0 \text{ to } 200^\circ\text{C/min} \\ \partial T / \partial h &= 100 \text{ to } -250^\circ\text{C/cm} \\ \partial^2 T / \partial h^2 &= 400 \text{ to } -400^\circ\text{C/cm}^2\end{aligned}$$

The calculated results for each run will give a spread of adsorption rate values for various temperatures, solid compositions, and gas compositions; which can be used for studying the adsorption mechanism in greater detail.

Three UF₆ runs were attempted on this apparatus without satisfactory results. In the first run some UF₆ leaked into the top of the adsorber bed; in the second, it was found that the bed from the previous run had not been fully regenerated; and in the third run, the soda-lime clean-up trap into which UF₆ was bypassed during start-up of the meter was plugged due to a larger pressure surge from the UF₆ meter. Now the whole apparatus is contaminated with UF₆.

6.0 CONCLUSIONS

1. The adsorber apparatus and operating techniques appear to be adequate for studying the adsorption of HF on the NaF pellets. This is shown by duplication of runs at different times.
2. The data from a total of nine good runs made on three different beds are available for numerical calculation.
3. Numerical solution of the system equations was demonstrated by hand calculation. Work is in progress on a program for the system equations which will be run on the Oracle.
4. General behavior of the adsorption process appears to be a very rapid adsorption of HF in NaF with the corresponding development of a sustained maximum bed temperature. As this condition moves down the bed, the upstream portion of the bed continues to adsorb HF but at a slower rate depending on the heat removal from the pellet and the diffusion of HF into the pellet.
5. Three runs using UF_6 as the adsorbate were attempted with unsatisfactory results. General indications were that a similar behavior exists between UF_6 and HF although smaller temperature gradients were observed when using UF_6 due, probably, to a lower adsorption rate.

7.0 ACKNOWLEDGEMENT

It is a pleasure to acknowledge the guidance and advice of Marvin Whatley, who originated and analyzed the problem, and designed and built the experimental equipment; C. C. Nance, who showed me many practical laboratory techniques; and others of the UNOP Section, who answered my many questions and requests for help.

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9.0 APPENDIX

The system equations for UF_6 adsorption on NaF are similar to the HF system equations and are not developed here.

9.1 UF_6 Experimental Apparatus and Operation

A line was brought from the UF_6 meter in cell 3B through the wall into cell 4A and connected to the adsorber apparatus ahead of the preheater. All the apparatus lines which might contain UF_6 were traced with electric heaters and asbestos tape to keep them above the gas dew point.

9.2 Suggested Operational Changes

1. Perhaps the main problem is to start-up the UF_6 meter without producing pressure surges that might plug the adsorber equipment. A study of the operating principles of the meter will reveal methods of starting up the meter without producing large pressure surges. Another approach might be to manually throttle the UF_6 line (downstream from the meter) independently of the meter until the desired flow is obtained and then let the meter take over the flow.

2. A gas distribution pipe could be installed in the clean-up trap to allow the trap to take a bigger UF_6 flow without plugging.

3. Regeneration of the adsorber bed. The $\text{UF}_6 \cdot 3\text{NaF}$ complex requires higher temperatures for decomposition than does the HF complex; moreover, it slowly decomposes to UF_5 and UF_4 complexes at regeneration temperatures. With the present furnace set at 400°C the maximum temperature obtainable in the bed is about 340°C when asbestos cloth insulation is placed on the top and bottom of the adsorber chamber. It is recommended that heat insulation be placed on the projecting ends of the adsorber chamber flush with the furnace insulation so that the pellet bed may be heated to about 400°C . The aluminum base on the adsorber should be removed to help minimize heat losses from the system. It is desirable to use some fluorine at some time during regeneration to be sure that any UF_5 or UF_4 complex formed by decomposition is removed from the bed.

It is desirable to have a handy means of checking bed temperatures above 300°C . A manual millivoltmeter was used.

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23. Mack Tyner, Jr., U. of Fla., Gainesville, Fla.
24. J. W. Ullmann
25. C. D. Watson
26. M. E. Whatley
27. Central Research Library
28. Document Reference Section
29. Laboratory Records (RC)
- 30-31. Laboratory Records
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