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QUARTERLY PROGRESS REPORT
RESEARCH AND DEVELOPMENT ACTIVITIES
FIXATION OF RADIOACTIVE RESIDUES
JULY - SEPTEMBER, 1963

By

Members of the Staff
Chemical Laboratory

Edited by
E. R. Irish

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INTRODUCTION

This progress report is the nineteenth in a series presenting research and development activities in the field of radioactive wastes.

Experimental work charged to programs other than those of the Division of Reactor Development is included for general interest and completeness; such work is identified in the headings.

SUMMARY

The radiant-heat spray calciner in the High-Level Radiochemistry Facility was used for additional experimental studies of full-level Purex 1WW calcination. The unit was operated with ammonia-neutralized Purex waste plus sugar, acidic Purex waste with phosphate added and with sugar added. Behavior of ruthenium especially and the properties of the melt were observed.

A tracer I^{131} run was made in the 8-inch diameter cold calciner to evaluate iodine behavior during calcination.

The large Cold Semiworks spray calciner was operated for further evaluation of the calciner capacity and heat transfer characteristics. Development of "glass" processes was initiated with installation and operation of a continuous melter for calcine powder.

An elaboration on the sugar denitration process for Purex 1WW is provided.

Experimental runs were made with a model annular circulator proposed for in-tank solidification of intermediate-level plant wastes to determine heat transfer characteristics.

Details of the engineering work on dehydration of fission product loaded zeolites were documented.

Equilibrium constants and free-energies were determined for two exchange reactions on six exchangers. Zeolites with low silica to alumina ratios exhibited the highest strontium selectivity.

Breakthrough curves for strontium on Linde 4A zeolite were predicted on the basis of plate theory and compared to experimentally determined curves. The calculated and experimental curves show close agreement.

Work continued on defining flowsheet conditions for removal of cesium from Purex and Redox wastes.

Experimental work is underway to evaluate electrodialysis as a processing technique for treatment of low- and intermediate-level wastes.

Pilot plant solvent extraction studies were continued on the development of CSREX and D2EHPA processes for removal of long-lived heat generating fission products from Purex wastes. Pulse column design details are presented.

CALCINATION AND SOLIDIFICATION STUDIES

(J. M. Atwood, R. L. Moore, A. M. Platt, W. H. Swift)

Waste Calcination Development

Full-Level Pilot Plant Calcination Studies - R. T. Allemann, F. P. Roberts, U. L. Upson

Experimental work continued with calcination of full-level Purex waste in the spray calciner in the High-Level Radiochemistry Facility. Calcination is carried out in the 10-inch diameter by 6-foot high spray calciner with an attached 5-inch diameter, 13-inch high melt pot. Pot calcination is also done in the same size melt pot, but with a feeding head attached. The same off-gas cleanup apparatus is used for both systems.

Further analytical results of last quarter's ammonia neutralized waste runs were obtained, and runs were made in both cold and hot equipment with Purex waste with ammonia plus sugar, with phosphoric acid, with phosphoric acid plus sugar, and with diammonium phosphate as additives.

Spray calcination runs are being made successfully, although the column still has a leak,(1) by drawing as large a gas flow as possible through the off-gas equipment and by using the by-pass filter method of sampling the off-gas dust.

Spray Calcination of Ammonia-Neutralized Purex Waste, Plus Sugar

The analytical results of the spray calciner Run 17 in which ammonia-neutralized 1WW was calcined verified the preliminary ruthenium volatility figures mentioned last quarter.(2) That is, the cumulative ruthenium

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(1) E. R. Irish. Quarterly Progress Report - Research and Development Activities - Fixation of Radioactive Residues - January—March, 1963, HW-77299, p. 9. April 1963.

(2) E. R. Irish. Quarterly Progress Report - Research and Development Activities - Fixation of Radioactive Residues - April—June, 1963, HW-78188, pp. 12-13. July 1963.

collected in the condensate was 4% of that fed to the calciner. Occasional "instantaneous" condensate samples showed losses as low as 1%, but the average was the higher value. Other fission products (Ce, Cs, Zr-Nb) had decontamination factors of about 1000 to the condensate. Off-gas decontamination factors with only the condenser and absolute filters in the line were in the normal range of $\geq 10^8$, which permits safe discharge to the building stack.

A subsequent ammonia-neutralized run (Run 18) was made (after testing in the cold 8-inch diameter calciner) in which 30 grams of sugar were added per liter of 1WW. (With 50 grams or more of sugar per liter of 1WW the powder would not melt at 900 C, although 0.72M sodium bisulfate was used as a fluxing agent.) The sugar was used to provide a reducing atmosphere during calcination and, hopefully, to reduce the ruthenium volatility to the low value (much less than 1%) which is obtained during caustic-neutralized Purex waste calcination with sugar. The total ruthenium found in the condensate under these conditions was 1% of that in the feed. Values for the "instantaneous" condensate samples ranged from 0.6 to 4%. Thus, the sugar addition in this run appears to have helped retain ruthenium somewhat. Decontamination factors to the condensate for the other γ emitting fission products were again over 1000, and decontamination factors to the final off-gas were over 10^7 for ruthenium and over 10^8 for the other fission products.

The ammonium ion analysis of the condensate showed that only 3% of the total ammonium ion added to the feed appeared in the condensate, although 20% recovery was obtained in the cold unit (probably because of somewhat lower operating temperature). Thus, the results indicate that the thermal decomposition of ammonia occurs fast enough in the calciner to limit severely the amount of ammonia available for recycle.

Spray Calcination of Purex Waste Plus Phosphate

Wastes calcined with phosphate added are less soluble in water than straight waste calcines, and the lower solubility is an advantage for safe long-term storage. Cold spray calciner runs were made with simulated Purex waste to which phosphoric acid was added in sufficient amounts to form orthophosphates of the metals present (ca. 0.6$\underline{M}$ H_3PO_4). The calcined powders from these runs did not melt in the pot at 850 C. Melts were obtained at 900 C in a stainless steel crucible, but only a partial sinter resulted in a glazed alumina crucible at this temperature. Since a lower melting calcine is desirable, sodium was added in subsequent runs. The ratio of Na/Fe/Al in the Purex waste used as feed in the hot-cell runs to date is 2.1/1.0/0.4. The radioactive run (Run 19) was made with sodium nitrate added to shift the ratio to 3.2/1.0/0.4 and form a lower melting calcine. Slightly more phosphoric acid was added to match the added sodium.

Calcination of this mixture was uneventful, and a melt was obtained in the pot which was held at 900 C.

Fission product behavior was not strongly affected by the phosphoric acid, as shown in Table I. These values are similar to those obtained with acidic Purex waste, with no additives.

Spray Calcination of Purex Waste Plus Phosphate and Sugar

Run 20 was made in the spray calciner. This run was similar to that described above except that 100 grams sugar per liter of waste were added. As shown in Table I, this sugar addition increased the decontamination of ruthenium to the condensate, although not to the high values obtained with caustic neutralized waste. However, sugar with phopshate did lower the ruthenium volatility more than it did with straight Purex waste (0.8% versus 3% loss). The runs showed a decrease from previous runs in cerium decontamination to the condensate which has not yet been expalined. Possible causes are: (1) a decrease in filter efficiency coupled with a less favorable particle size of cerium-containing powder; (2) cross-contamination with high-cerium solutions which are being studied in the shielded cell adjacent to the calciner cell; and (3) sampling and counting errors.

TABLE I

FISSION PRODUCT DECONTAMINATION OF SPRAY CALCINER OFF-GAS

(Run 19: Purex Waste plus Sodium Nitrate and Phosphoric Acid)

	Cumulative Decontamination Factors			
Decontamination Unit	Ce	Ru	Cs	Zr-Nb
Column plus Filters (by Condensate Analysis)	700	8	$>10^3$	$>10^3$
Condenser	$> 10^6$	$>10^5$	$>10^6$	$>10^6$
Cambridge Absolute Filters	$> 10^8$	$>10^8$	$>10^8$	$>10^8$

(Run 20: Purex Waste plus Sodium Nitrate and Phosphoric Acid and Sugar)

	Cumulative Decontamination Factors			
Decontamination Unit	Ce	Ru	Cs	Zr-Nb
Column plus Filters (by Condensate Analysis)	600	125	$>10^4$	$>10^4$
Condenser	10^6	10^5	10^6	10^5
Cambridge Absolute Filters	$>10^8$	$> 10^8$	$>10^8$	$>10^8$

An additional run (Run 21) was made with diammonium phosphate as an additive. The analytical results of this run may help resolve the cerium anomaly. The ruthenium decontamination factor to the condensate was 12 to 16. A melt was obtained, but some difficulty with steam orifice plugging in the nozzle was experienced. When air was used as the spraying fluid, the plugging did not occur.

Iodine Behavior in Spray Calcination

Because the full-level wastes which have been calcined have been aged, radioactive iodine has not been a problem. However, it is desirable to know how iodine (which is present in younger wastes) would behave.

To this end, an iodine spike run was made in the 8-inch diameter cold calciner. Approximately 40 μc of I^{131} as NaI were added to 6.6 liters of simulated sugar-treated 1WW which was then spray calcined. The off-gas train consisted of sintered metallic filters, condenser, de-entrainer, and an 8-inch long by 1-inch diameter tube of charcoal as a backup adsorber. Charcoal samplers were used for sampling the off-gas between the condenser and the de-entrainer. The analytical and counting results are interpreted in Table II.

TABLE II

APPARENT IODINE DISTRIBUTION AFTER SPRAY CALCINATION

Iodine in Calcine	17%
Iodine in Condensate	8%
Iodine in Condenser Wash	13%
Iodine in Charcoal Adsorber	4%
Iodine Unaccounted (by difference)	58%
Iodine DF to Condensate	14
Sodium DF to Condensate	5700

Samples of the charcoal from the backup adsorber showed the same concentration of iodine at both ends. The lack of trail-off in the bed indicates that it was saturated and that some of the iodine escaped. The low adsorptivity may be caused by the high NO_2 or N_2O_4 concentration in the off-gas, which is known to have a detrimental effect on iodine adsorption.(3)

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(3) W. R. Jacobson and L. Jolly, Jr. Measurement of Radioiodine in Purex Stack Gases, DPSPU 63-30-4B. May 1963.

The fact that the backup bed appeared to be saturated indicates that the much smaller charcoal impregnated filters and charcoal vial samplers were probably saturated (sampling time was 15 minutes) and the off-gas decontamination data obtained from them would be erroneous.

Thus, the indications are that 80% of the iodine is volatilized from the calcine and that only part of it is recovered in the condenser system. The low sodium carryover to the condensate infers that the iodine was in a volatile rather than particulate form in going to the condensate.

Cold Semiworks Program

Most of the work during this quarter was devoted to a study of the heat transfer and capacity characteristics of the 18-inch diameter by 10-foot long spray calciner. However, the filter blow-back system, a continuous calcine melter, and a denitrator evaporator were also studied.

Radiant-Heat Spray Calciner - B. M. Johnson, J. D. Kaser

Recirculation Rate

Approximate recirculation rates were calculated from gas temperatures at the top and bottom of the calciner by assuming the total heat required for evaporation of the spray is supplied by the sensible heat of the recirculating gas. Data from four runs made at identical feed rates and atomization pressures (24 gph and 45 psig) but at different furnace temperatures ranging from 650 C to 800 C indicated a nearly constant recirculation rate of 1600 lb/hr with the internal-mix nozzle (Spraying Systems Company Set Up No. 72*). The fact that the recirculation rate is independent of furnace temperature seems to confirm the assumption that direct radiant heat transfer from the draft tube to the spray is not important. Later calculations of the radiant heat transfer situation indicate that about 10% of the heat load to the feed is supplied by direct radiation from the draft tube.

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* Spraying Systems Co., 3201 Randolph, Bellwood, Illinois

A rough estimate of recirculation rate as a function of the steam and liquid rate was obtained from an analysis of temperature data obtained during a series of runs at 750 C furnace temperature. This analysis indicates the recirculation rate varies directly with the steam flowrate but is independent of the liquid rate. At present, data are being obtained with a movable, shielded and aspirated thermocouple to improve the estimates of recirculation rate.

Calculations based on data for the ratio of entrained to original gas in subsonic, single-phase jets(4) indicate the main motive force for recirculating the gas stems from the induction requirements of the spray jet.

Heat Transfer in the Annular Region

As the gas passes up through the annulus it is heated both by radiation and convection from the hot wall and also the draft tube. A differential heat balance then gives the following equation:

$$G C_p W D_m \frac{dt}{dx} = F_{SG} D_o \sigma (T_w^4 - T^4) + h D_o (T_w - T) \quad (1)$$

where G = mass velocity of the recirculating gas

W = width of the annulus

D_m = mean diameter of the annulus

D_o = diameter of the reactor

x = distance up the reactor

T = gas temperature

h = heat transfer coefficient for convective heat transfer from draft tube and reactor wall, based on reactor area alone.

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(4) J. F. Taylor, H. L. Grimmett, E. W. Comings, "Isothermal Free Jets of Air Mixing With Air," Chem. Eng. Prog., vol. 47, pp. 175-180. 1951.

C_p = isobaric heat capacity

t = time

σ = Stefan - Boltzmann Constant

T_w = reactor wall temperature

$$F_{SG} = \frac{\text{(Total net radiative flux to gas)}}{\text{(Area of contact with reactor wall)} \, [\sigma(T_w^4 - T^4)]}$$

Equation (1) may be manipulated and integrated to obtain a relationship requiring estimation of F_{SG} and h to determine net heat duty as a function of reactor geometry. The interchange factor, F_{SG}, was estimated by the method of Hottel,(5) and the convective heat transfer coefficient, h, was calculated by the Sieder-Tate equation. From these numbers the length of column required to raise a fixed quantity of recycle gas from 324 C to 575 C was calculated in an effort to determine the optimum annular spacing. The results are shown in Table III.

TABLE III

COMPARATIVE COLUMN LENGTH FOR FIXED HEAT DUTY AS A FUNCTION OF GAP WIDTH

Gap Width, inches	0.9	1.4	2.4	3.4	4.5
Comparative Column Length, feet	0.81	0.99	1.0	1.01	1.06

With the exception of the narrowest gap width, there is essentially no variation in heat transfer rate versus gap width. The evident advantage of narrow annulus may be offset by an increased pressure drop which may cut down the recycle rate in the narrow annulus and thus reduce the coefficient "h".

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(5) H. C. Hottel, E. S. Cohen. "Radiant Heat Exchange in a Gas-Filled Enclosure," A.I.Ch.E. Journal, vol. 4, p. 3. 1958.

Since conduction-convection is the principal mode of heat-transfer in the narrow annulus, the reactor capacity would be quite sensitive to changes in "h". As a result of this analysis, somewhat narrower annuli and/or extended heat transfer surface will be investigated.

Heat Transfer in the Center Region

The direct transfer of radiant-heat to the spray and gas from the wall of the draft tube is difficult to calculate with precision. The method used is the result of analysis of the work of Hoffman and Gauvin.(6, 7) Cursory use of this technique led to the conclusion that, within the required accuracy of the correlation between theory and measured values of the column capacity, the contribution of radiant transfer to the center section, Q_R, could be approximated as

$$\frac{Q_R}{A} = 0.045\,(0.67\,T_w{}^4 - T_{G_c}{}^4) \qquad (2)$$

T_{G_c} = center region gas temperature (taken as 300 C)

A = area over which radiant heat transfer to center region is significant - 11 ft^2 (top 3 feet).

In the subsequent section on the correlation of results, the contribution of this radiant heat transfer is shown to be less than 15% of the total, which justifies the gross simplification and the probable low precision of the approach.

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(6) T. W. Hoffman, W. H. Gauvin. "An Analysis of Spray Evaporation in High-Temperature Environment," To be published. (Can. J. Chem. Engr.)

(7) W. H. Gauvin, T. W. Hoffman. "Heat Transfer to Particles in High Temperature Surroundings," To be published. (Can. J. Chem. Engr.)

Measured and Predicted Capacity of the Radiant-Heat Spray Calciner

Figures 1 and 2 are curves of the measured capacity of the reactor with water and synthetic waste solution as a function of furnace temperature and atomization pressure for both an internal and external-mix nozzle, Spraying Systems Company Set Up No. 72 and 70, respectively. Figure 2 is a cross-plot showing the variation in capacity with atomization pressure for both types of nozzle.

Reactor capacity with the internal-mix nozzle operating at a given atomization pressure was from 5 to 8% higher than with the external-mix nozzle, and atomization gas requirements were generally about 30% less. However, feed line pressure is higher with the internal-mix nozzle (40 to 80% of the atomization steam pressure versus zero or negative line pressure). At a constant furnace temperature, the reactor capacity increases approximately with the square root of the absolute atomization pressure, which in turn is directly proportional to the atomization flow rate.

Included in Figure 1 is a curve for the reactor capacity when operating with a typical synthetic waste solution. This is approximately 80% of that with water under otherwise identical operating conditions. At least four reasons for this lower capacity can be advanced: (1) the endothermic heat of reaction for evaporation and calcination of the waste is about 7% higher on a feed volume basis, (2) the boiling temperature of the drops of waste is somewhat higher, causing a falling rate period during drying and consequently a slower rate of evaporation, (3) atomization is undoubtedly poorer because of the higher viscosity of the solution, and (4) heat transfer from the walls of the reactor is somewhat impaired by the dust.

The average heat flux obtainable in the shortened reactor (4.75 feet) is more than 30% higher than in the longer reactor, but the variations in capacity with furnace temperature and atomization pressure are similar, as seen in Figures 1 and 2. The higher average flux results from two factors: (1) heat transfer is more rapid in the initial (lower) section of the annulus where

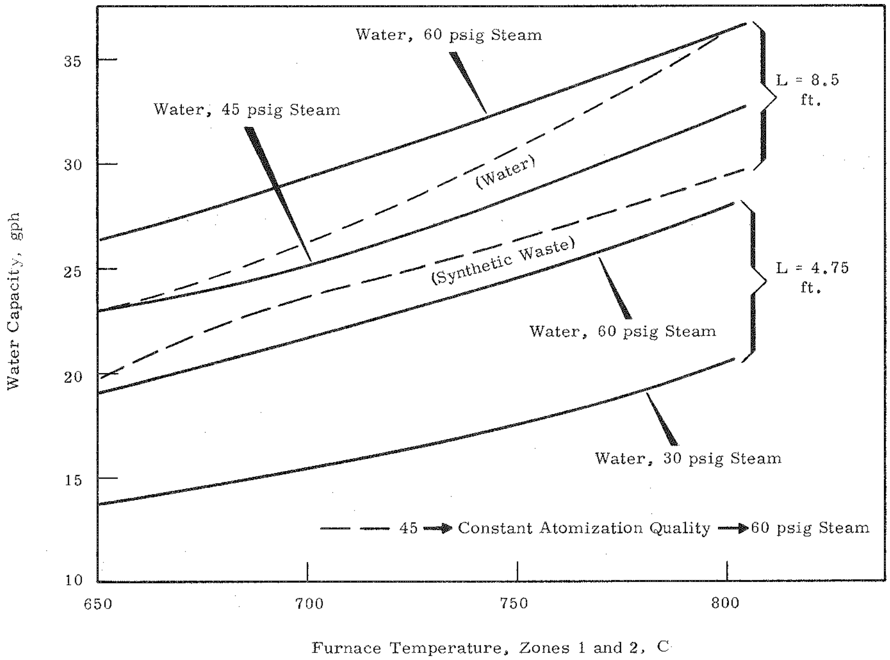

FIGURE 1

Effect of Furnace Temperature
on the Capacity of 18-Inch Spray Calciner

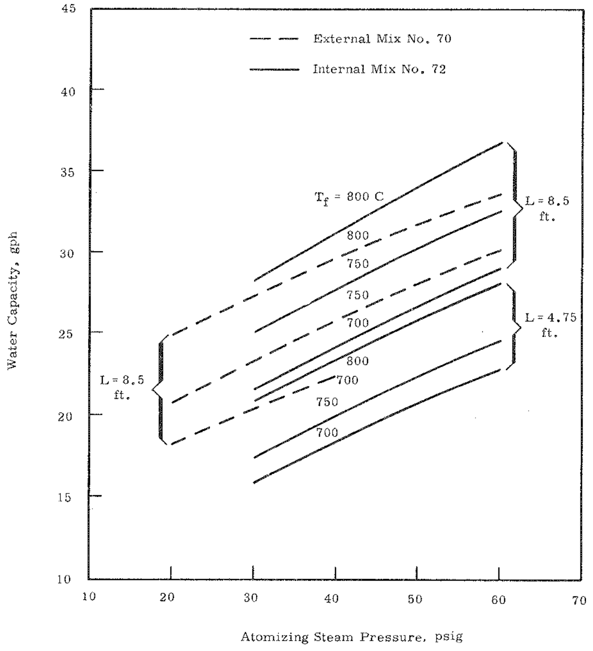

FIGURE 2

Effect of Atomizing Steam Pressure
on the Capacity of 18-Inch Spray Calciner

the temperature difference between the wall and the gas is highest, and (2) the contribution of radiant heat transfer to the center region is significant only near the top of the reactor.

The comparison between the predicted and calculated capacity of the reactor is shown in Table IV.

TABLE IV

PREDICTED AND MEASURED REACTOR CAPACITY WITH VARIOUS WALL TEMPERATURE AND REACTOR LENGTHS

T_f C	(T_w)ave C	Rate gph	Q Btu/hr	Q/A Btu/hr ft^2	Q_R(calc) Btu/hr	Q_A(calc) Btu/hr	Q_T(calc) Btu/hr
Reactor Length = 8.5 feet							
650	610	23	212,000	5,300	15,000	192,000	207,000
700	655	26.4	250,000	6,250	18,000	231,000	249,000
750	705	31	287,000	7,180	25,000	272,000	297,000
800	750	37	334,000	8,350	31,000	305,000	334,000
Reactor Length = 4.75 feet							
650	580	16	147,000	6,580	12,000	128,000	140,000
800	730	26	240,000	10,740	29,000	207,000	237,000

The significance of the various columns is as follows:

T_f = furnace temperature at the control point
T_w = average wall temperature of the reactor
Rate = feed rate at capacity conditions
Q = calculated heat duty at the feed rate
Q/A = average heat flux over the whole length of the reactor
Q_R = calculated contribution of radiant heat transfer to the center region
Q_A = total calculated heat transferred in the annulus
Q_T = sum of Q_R and Q_A.

The values of Q_T agree with the measured values of the heat load well within the limit of precision with which the capacity can be determined. All the runs at the full reactor length were made at what was intended to be a constant atomization quality; i.e., the atomization pressure was varied in such a manner as to give the same predicted mean drop size by the Gretzinger correlation.(8) This condition resulted in a variation in atomization pressure between 45 and 60 psig as the rate varied between 23 and 37 gph.

Filter Blow-Back Study - J. D. Kaser

Initial phases of this study are directed toward determining conditions required to maximize the overpressure using venturi-nozzle blow-back systems. (Over-pressure is the difference in pressure between the upstream and downstream filter faces during blow-back.)

Previous work (9) showed that dust removal increases with the overpressure induced in the filter by the blow-back gas. In this study the overpressure was measured with the same type cylindrical filter and venturi now installed in the Cold Semiworks spray calciner (10, 11), using a strain gage pressure transducer coupled to a conventional oscilloscope. Pressure-time traces were recorded photographically. The blow-back fluid was dry air.

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(8) J. Gretzinger, W. R. Marshall, Jr. "Characteristics of Pneumatic Atomization," A.I.Ch.E. Journal, vol. 7, pp. 312-318. 1961.

(9) R. Dennis, E. Kristal, G. A. Peters, L. Silverman. Laboratory Performance of the Mikro-Pulsaire Collector, NYO-4816, pp. 34-35. June 1962.

(10) E. R. Irish. Quarterly Progress Report - Research and Development Activities - Fixation of Radioactive Residues - January—March, 1963, HW-77299, p. 20. April 1963.

(11) E. R. Irish. Quarterly Progress Report - Research and Development Activities - Fixation of Radioactive Residues - April—June, 1963, HW-78188, p. 14. July 1963.

In the first runs the effects of distance from the blow-back nozzle to the venturi throat and nozzle size were investigated. Two nozzles (0.41 and 0.16 in. ID) were compared at constant upstream air delivery pressures and at constant air flow rates. In all cases the overpressure attained a steady value 0.2 sec after a solenoid valve opened, allowing air to flow through the nozzle. Overpressure was independent of position on the filter face, except within two inches of the top of the filter where the extension of the venturi into the filter probably interfered with air flow to the filter face. Steady state overpressure data measured at the midpoint of the filter are summarized in Table V.

TABLE V

STEADY STATE FILTER OVERPRESSURES

Distance from Nozzle to Throat, Inches	Overpressure, Inches of Water: 0.41 Inch ID Nozzle	Overpressure, Inches of Water: 0.16 Inch ID Nozzle
A. Air Delivery Pressure Constant at 60 psig		
-1	3.1	1.5
0.5	3.8	1.8
1	3.9	2.2
2	4.0	2.2
3	3.9	2.3
4	3.8	2.4
B. Air Flow Rate Constant at 24 scfm		
-1	0.9	1.5
0	0.9	2.2
2	1.1	2.5
4	1.3	2.6
6	1.1	2.5
10	0.8	1.8

Within experimental error, the data indicate little effect of nozzle to venturi distance when the nozzle is from 2 to 6 inches from the venturi throat. At the same flow rate, the smaller nozzle produced greater overpressures than did the larger nozzle. However, the gas velocity in the smaller nozzle was sonic, while that in the larger was subsonic.

Calcine Powder Melter - J. D. Moore

As part of the accelerated development program in support of the Waste Solidification Engineering Prototypes, a continuous melter for calcine powder was installed and successfully operated in the Cold Semiworks. The melter is designed to produce a phosphate "glass" product at an instantaneous rate equivalent to processing waste from a separations plant handling 2 tons of uranium per day. Later, the melter will be coupled directly to the 18-inch radiant-heat spray calciner.

The melter consists of an induction heated Inconel 702 pot, 8 inches in diameter and 11 inches high (including the conical lower section), with provisions for off-gas removal and molten "glass" product collection. Powder is fed to the melt pot by a screw conveyor and an air vibrator. Melt may be discharged from the pot by a bellows-actuated valve seated in the bottom of the pot or by a siphon which controls the melt level at a depth of 3 inches. At present, the phosphate "glass" product is collected in a mild steel container which is enclosed in a stainless steel hood coupled to the melt pot. A 1-1/2 inch stainless steel pipe at the top of the melter is connected to a water-cooled condenser and caustic scrubber to provide for the removal of off-gases. Provisions are included for injection of steam into the off-gas stream to expedite the removal of any sulfur dioxide evolved from the melt.

To date, four experimental runs ranging from 2 to 4 hours in duration have been completed. The melter was operated at 900 C under a slight vacuum (2 to 6 inches of water). Continuous feeding of the powder was not attained, but estimated overall powder flow rates were 3 to 4 lb/hr. No efforts were made to reach the capacity of the melter. Feed to the melter was a calcine powder with a bulk density of 30 lb/ft^3. The feed was prepared as a synthetic Purex waste with lithium (1.26M), calcium (0.23M), and phosphate (1.26M), added before calcination in the 18-inch radiant-heat spray calciner. The powder on melting formed a free-flowing liquid with little foaming or spattering. The melting process as observed visually through a window in the melt pot was rapid and took place with no noticeable off-gassing and with very little solid phase present.

Intermittent removal of the molten product was made satisfactorily with either the bellows valve or the siphon systems.

Upon cooling, the product formed a hard, dark-brown, seemingly deliquescent solid with a density of 2.7 g/cm^3. The product fractured into pieces during rapid cooling.

Future operation of the calcine powder melter will be aimed at determining capacity and measuring the extent of sulfate volatilization from the feed.

Denitrator Evaporator - J. L. McElroy, G. Rey

The concept of the continuous phosphate glass process, under development by Brookhaven National Laboratories, incorporates a denitration evaporation step prior to a high temperature melter. Studies in support of design were initiated to determine the requirements for a denitration evaporator.

A small evaporator made of 6-inch Pyrex glass with a 304-L stainless steel steam-heated bayonet was constructed. Heat transfer studies on simulated Purex type wastes were conducted. Purex 1965, Type A waste with 0.005 $\underline{M}$ Zr^{+4}, 0.005 $\underline{M}$ rare earths, 1.3 $\underline{M}$ PO_4^{-3} (added for glass formation) and 1.25 $\underline{N}$ in total metal ions was concentrated in the evaporator by a factor of seven to a boiling point of 135 C. At this temperature the concentrate had a viscosity of 300 centipoises and a specific gravity of 1.85. During concentration, foaming occurred at high boilup rates. However, a sodium analysis of the distillate showed very little entrainment (DF = 4×10^4). Initial data, at a ΔT of 40 F°, gave an overall heat transfer coefficient of 190 Btu/hr-ft^2 - F. This coefficient steadily climbed to a peak value of 225 after boiling 45 hr, at a constant temperature of 135 C, at which time it began to decrease. After a down period of 3 days, solids had settled to about 50% of the total volume. These solids restricted natural circulation and reduced the rate of heat transfer to 140 Btu/hr-ft^2-F in subsequent boiling tests. Extensive boiling and mild agitation did not mix these solids back into solution. Inspection of the bayonet heater revealed some pitting of the surface and an absence of scale. The heat transfer coefficient for water before and after the Purex waste test increased from 300 to 330, respectively.

Attempts were made at concentrating a second waste (Purex Type B) having $0.1\underline{M}$ Zr^{+4}, $0.2\underline{M}$ rare earths, $1.7\underline{M}$ PO_4^{-3} (added for glass formation), and $1.64\underline{N}$ total metal ions. This type waste is conceivably attainable from a 10,000 Mwd/ton fuel processing plant which would segregate first cycle Purex waste. It is also possible to generate it inadvertently in plant operations through a process of concentrating by settling in fluid transfers. This mixture contained about 40% settled solids before concentration and was concentrated by a factor of two to a boiling point of 112 C. At this temperature the mixture was a gel and could not be boiled in the experimental evaporator. One run was made with Type B waste boiling at 110 C. The measured heat transfer coefficient was 80 Btu/hr-ft^2 - F. On a second run at a 110 C the waste began to dry out near the heater and solids were baked onto the bayonet.

Both types of waste (A and B) studies represent worst-case compositions in terms of calcium and zirconium content since calcium was used to represent the total anticipated alkali earths (Ca-Sr-Ba) and additional zirconium was used to simulate molybdenum.

The stainless heater was replaced by one made of Nionel to determine the suitability of Nionel as a heat transfer surface under the conditions encountered in evaporating acidic Purex waste.

High-Level Waste Treatment

(E. A. Coppinger)

(Not Charged to Division of Reactor Development)

Denitration of Purex Waste with Sugar

A sugar denitration process for Purex 1WW was reported last quarter. A batch method of operation, which was employed in the pilot plant studies and adopted for testing in the Purex Pant, was only briefly mentioned and is described more fully here. In the batch method, the desired volume of Purex waste (typically 1500 to 2500 gallons) is transferred to the reaction vessel and heated to about 100 C. A concentrated sugar solution sufficient to accomplish the desired degree of denitration is slowly added to the hot waste over a period of up to 12 hours. A major portion of the nitric acid is destroyed during the sugar addition period. After sugar addition is complete, the 100 C temperature is maintained and the solution is digested for up to 12 hours to complete the reaction. The rate at which sugar can be added is determined by the capacity of the off-gas system, the dimensions of the reaction vessel, and the tendency of the waste to foam. Limitations on the sugar addition rate are discussed in detail.(12)

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(12) E. A. Coppinger. <u>Pilot-Plant Denitration of Purex Waste with Sugar</u>, HW-77080. March 29, 1963.

In-Tank Solidification

(J. Dunn, A. J. McElfresh)

(Not Charged to Division of Reactor Development)

Present plans for in-tank solidification of intermediate-level plant wastes call for evaporation of the waste solutions by introducing 1200 F air near the bottom of the underground tanks. The air, acting as an air-lift circulator, flows downward through an annulus, transfers heat to the solution, and induces circulation in the tank. A pilot scale mock-up of the proposed in-tank solidification system was completed and placed in operation. The purpose of the experimental unit is to provide information needed for design of the full-scale system to be installed in a plant tank.

Calculations indicated that the heat transfer coefficient attainable from a straight annular circulator as currently proposed is approximately 8 $Btu/hr\text{-}ft^2\text{-}F$, which is insufficient to cool the hot gas to the approximately 700 F required to avoid entrainment problems. Experimental runs conducted in a model annular circulator yielded a heat transfer coefficient value in the range 10 to 12 $Btu/hr\text{-}ft^2\text{-}F$. It is concluded that a finned annular surface is needed to reduce the inlet gas temperature to the desired value. Calculations are now essentially complete for the optimum fin design.

Fission Products and Waste Packaging

(W. V. DeMier)

(Not Charged to Division of Reactor Development)

Dehydration of Zeolite Beds

The majority of the engineering work on the dehydration of fission product loaded zeolite has been completed. Details of this work have recently been documented.(13)

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(13) W. V. DeMier. Dehydration of Inorganic Ion Exchangers, HW-78893. September 13, 1963.

SORPTION AND EXTRACTION STUDIES

(G. J. Alkire, C. E. Linderoth, W. H. Swift)

Fundamental Sorption Research

(L. L. Ames)

(In Part Charged to Division of Reactor Development)

Rational thermodynamic equilibrium constants and standard Gibbs free-energies were determined for two exchange reactions with the zeolites Linde 4AXW, 13X, AW-400, AW-500, Norton Zeolon and clinoptilolite. The initial step was computation of a mass action quotient for the given reaction, uncorrected for solution or zeolite activity coefficients. The computation of activity coefficients for correction of cation concentrations in mixed solutions was accomplished through the use of Glueckauf's equation.(14) The expression for determination of concentration corrections in a sodium-strontium system is:

$$\log \frac{\begin{bmatrix} f^{x_1 y} \\ (x_2 y) \end{bmatrix}^4}{\begin{bmatrix} f^{x_2 y} \\ (x_1 y) \end{bmatrix}^3} = \frac{r+2}{r+3} \left\{ 2 \log f^{x_1 y} - 9/4 \log f^{x_2 y} \right\} - \frac{1/4\,(r-2)}{I(1+I^{-1/2})} (x_2)$$

where x_1 = sodium concentration in molarity

x_2 = strontium concentration in molarity

r = ${}^{x_1}/{}_{x_2}$

$\begin{matrix} f^{x_1 y} \\ (x_2 y) \end{matrix}$ = the mean activity coefficient for sodium chloride in the presence of strontium chloride

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(14) E. Glueckauf. "Activity Coefficients in Concentrated Solutions Containing Several Electrolytes," Nature, vol. 163, pp. 414-415. 1949.

$f^{x_2y}_{(x_1y)}$ = the mean activity coefficient for strontium chloride in the presence of sodium chloride

f^{x_1y} = the mean activity coefficient for sodium chloride.

f^{x_2y} = the mean activity coefficient for strontium chloride

and I = the total ionic strength of the equilibrium solution.

It should be noted that the above are rational, and not molal, mean activity coefficients.

The Glueckauf equation reduces to:

$$\frac{f^{x_1y}_{(x_2y)}}{f^{x_2y}_{(x_1y)}} = \left(\frac{f^{x_1y}}{f^{x_2y}}\right)^{3/2}$$

, for activity coefficients in a calcium-strontium system.

Activities in the zeolite phase were determined by a cumulative integration of the area under the line resulting when the log of the mass action quotients, corrected with solution activity coefficients, was plotted versus the fraction of the traced cation on the zeolite at equilibrium. The derived thermodynamic data are given in Tables VI and VII. A general trend may be noted for both reactions, with the greater strontium selectivities for the zeolites with the lower silica to alumina ratios (4AXW and 13X).

By completing a third associated exchange reaction,

$$Ca_{zeolite} + 2Na^{+}_{solution} \rightleftharpoons 2Na_{zeolite} + Ca^{+2}_{solution}$$

the accuracy of the experimental results will be ascertained with the "triangle rule".

TABLE VI

THERMODYNAMIC DATA FOR THE EXCHANGE REACTION AT 25 C

$$2Na_{zeolite} + Sr^{+2}_{solution} \rightleftharpoons Sr_{zeolite} + 2Na^{+}_{solution}$$

Zeolite	K	ΔG^{o}, Cal/mole
Linde 4AXW	42.33	- 2200
Linde 13X	13.03	- 1500
Linde AW-400	0.339	+ 600
Linde AW-500	0.295	+ 700
Norton Zeolon	0.220	+ 900
Clinoptilolite	1.093	- 100

K = a rational thermodynamic equilibrium constant

ΔG^{o} = the standard Gibbs free-energy rounded to the nearest 100 Cal/mole

TABLE VII

THERMODYNAMIC DATA FOR THE EXCHANGE REACTION AT 25 C

$$Ca_{zeolite} + Sr^{+2}_{solution} \rightleftharpoons Sr_{zeolite} + Ca^{+2}_{solution}$$

Zeolite	K	ΔG^{o}, Cal/mole
Linde 4AXW	3.61	- 800
Linde 13X	2.95	- 600
Linde AW-400	0.351	+ 600
Linde AW-500	0.478	+ 400
Norton Zeolon	0.376	+ 600
Clinoptilolite	1.70	- 300

K = a rational thermodynamic equilibrium constant

ΔG^{o} = the standard Gibbs free-energy rounded to the nearest 100 Cal/mole

Low- and Intermediate-Level Waste Treatment

(L. F. Coleman, B. W. Mercer, W. F. Unzicker)

Batch equilibrium experiments were conducted to determine the uptake of trace ruthenium from steam stripped high-level alkaline waste condensate by various anion exchange resins. The resins were based with nitrite ion, the predominant anion (5-10 ppm) in the condensate, in order to obtain the relative selectivities of the exchanger for ruthenium in the presence of nitrite. The highest ruthenium distribution coefficients (K_d's = 120 - 130) were obtained with Duolite A-14, Amberlite IR-400, and Dowex 1 x 8. Ruthenium adsorption by Dowex 1 x 4 was less than that by Dowex 1 x 8 which indicates that increased cross linkage of the resin (nitrite form) results in greater ruthenium adsorption from the condensate.

Although the resins listed above have the highest selectivity of those tested for ruthenium in the presence of nitrite, they do not necessarily have the highest capacity for ruthenium in the condensate waste. Past experience with condensate waste has shown that ruthenium breakthrough occurs along with the nitrite breakthrough on hydroxyl-based resins. Therefore, in selection of an exchanger in the hydroxyl form for maximum ruthenium loading, both rutheniun selectivity and the total capacity of the exchanger for nitrite must be considered.

Electrodialysis is being investigated as a possible process step for the treatment of intermediate-level wastes. The main applicability appears to be in the pretreatment of wastes to extend the bed life of adsorbent columns; however, it may have added benefits with respect to the removal of radio-ruthenium.

The laboratory-scale electrodialysis unit being used has 20 concentration-dilution cell pairs, each with an effective area of 220 cm^2. Preliminary desalting experiments using feed solutions of dilute sodium hydroxide demonstrated the effects of salt concentration, feed rate and current density on desalting rates, current efficiency and power consumption. Conductivity of the dilution stream largely controls the amount of current which can be applied to the unit; thus, it also controls the desalting rate. Feed rate is important due to less polarization of the membranes at higher flow rates. Current density obtainable is related to solution and membrane resistances and is important in relation to desalting rates versus power consumption. Operation at too high a current density results in power losses due to the limiting current-polarization effect.

Optimum operating conditions for desalting specific waste streams to a given limit are being evaluated. In conjunction with this research a small 6-membrane electrodialysis unit was constructed to determine radio-ruthenium transport with respect to bulk salt transport. The unit is also being used to determine relative concentrations of cationic and anionic ruthenium species present. The membranes used in the cell are Nepton* type ion exchange membranes or resin impregnated filter papers. Operation of the cell was such that anionic and cationic ruthenium species were separated in two waste streams which were isolated from the electrode compartments to avoid adsorption effects. Preliminary data on nitrate-carrier free Ru^{106} transfer rates in $0.016\underline{N}$ HNO_3 show a slower transfer rate for Ru^{106} than for nitrate ion. At the time 20, 50 and 95% of the nitrate ion had transferred about 4, 17, and 90% of the Ru^{106} had transferred. It is not known whether the observed increase in ruthenium/nitrate transfer ratio with time is associated with concentration effects, ionic species mobilities or pH changes.

Equipment installation was completed for demonstrating ion exchange treatment of Purex Tank Farm condensate on a pilot plant scale. The process(15) is based on data reported in previous progress reports and consists of ammonia and organic removal by steam stripping, followed by ion exchange. Startup of hot operation is planned for early October.

Specifications were written for a horizontal plate pressure filter to be used in thin bed ion exchange studies.

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* Trademark. Ionics, Inc.

(15) J. M. Skarpelos. Progress Report - Treatment of a Radioactive Condensate Waste, HW-79174. October 8, 1963.

High-Level Waste Treatment, Sorption

(B. W. Mercer, J. L. Nelson)

(Not Charged to Division of Reactor Development)

Cesium Separation

Development of ion exchange processes for the removal and purification of cesium from high-level Purex and Redox wastes was continued. Three additional synthetic waste solutions were tested for cesium removal. The first of these was an "aqueous sludge leach" solution simulating water wash of the sludge in Purex waste tanks following supernatant solution removal. This aqueous solution is expected to contain most of the cesium from the sludge as well as about 2 - 3$\underline{M}$ Na^+. Linde AW-400 zeolite proved to be the best exchanger, yielding a 1% cesium breakthrough at 120 column volumes and a 50% breakthrough at 300 column volumes, using a flow rate of 4 column volumes per hour. Duolite C-3 resin performed much less favorably both with and without a caustic addition to the waste.

Another synthetic waste tested for cesium removal by ion exchange was a "1965 FTW" which had been neutralized to pH 10 and filtered or centrifuged. This "1965 FTW" is Purex high-level acid waste that has been treated with formaldehyde for partial acid destruction and contains about 1.5$\underline{M}$ Na^+. Such a waste may be processed following strontium removal and neutralization in preparation for tank storage or after interim storage and before in-tank solidification. With Linde AW-400 zeolite a 1% cesium breakthrough was obtained at 130 column volumes and 50% breakthrough at 170 column volumes at a flow rate of 4 column volumes per hour. This indicates very favorable capacity as well as kinetics.

The third synthetic waste used was an acid Redox high-level waste which contains about 2$\underline{M}$ Na^+ and 2$\underline{M}$ Al^{+3}. Clinoptilolite was used as the exchanger and a poor cesium loading of less than 10 column volumes was obtained.

Since kinetics are limiting in the removal of cesium from Redox alkaline supernatant waste with Duolite C-3 resin, pressure-drop measurements were made with various resin particle sizes to determine the practicability of using finer mesh resin. Pressure-drop data were obtained from 20-40, 40-60, and 20-60 mesh granular resin, both for water and for synthetic "SX" tank farm waste, and are shown in Figure 3. The results indicate that the 40-60 mesh resin has a high pressure drop with SX waste, but that the other particle sizes have satisfactorily low pressure drops at the 2 to 4 column volumes per hour flow rate of interest. Cesium breakthrough curves with the same particle sizes and waste indicate that kinetics are poor for the 20-40 mesh but satisfactory for the other particle sizes; thus, 20-60 mesh resin appears to be satisfactory from both standpoints.

Recent experiments showed that the flow sheet for removal of cesium from high-level wastes can be altered to use water rather than dilute ammonium carbonate for scrubbing sodium ions from the loaded column. The Na/Cs ratio in the eluate is about three times that where an ammonium carbonate scrub is used but is still very satisfactory for zeolite packaging if two ion exchange cycles are used.

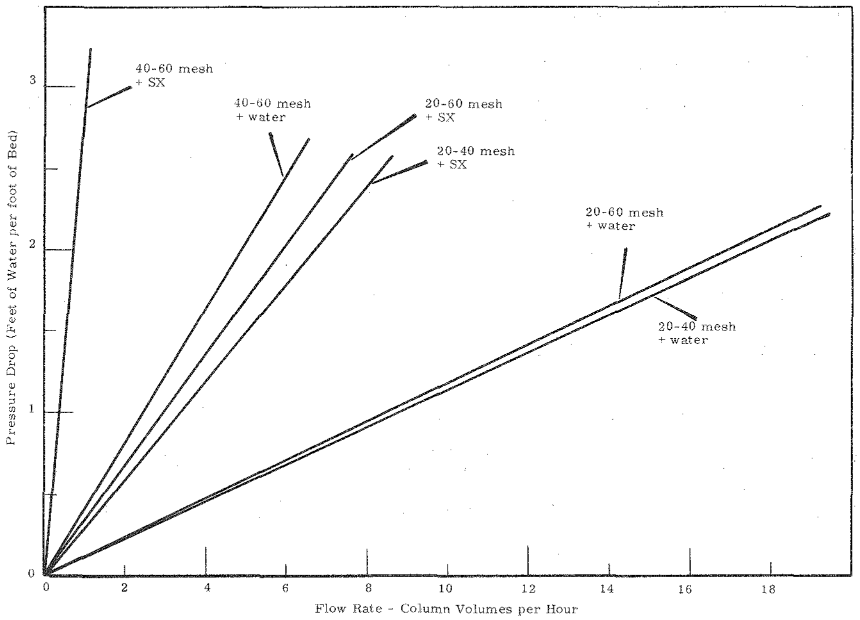

FIGURE 3

Pressure-Drop Data for Columns of Duolite C-3 Resin with Water and with Synthetic Redox Alkaline Supernatant (SX) Waste

Loading of Strontium on Type A Zeolite

The application of Glueckauf's plate theory (16, 17) to strontium ion exchange in columns of Linde 4A zeolite was studied. Liquid diffusion coefficients for Sr^{+2} were calculated for a simulated D2EHPA 1BP solution ($0.20\underline{M}$ $NaNO_3$, $0.035\underline{M}$ $Sr(NO_3)_2$, $0.010\underline{M}$ $Ca(NO_3)_2$) from electrical conductance data at different temperatures.(18) Counter-diffusion of Sr^{+2} and Ca^{+2} with Na^+ was assumed. The liquid diffusion coefficients for Sr^{+2} were used with other parameters to calculate particle diffusion coefficients from column data with the following equations:

$$\text{Number of Theoretical Plates (N)} = \frac{\text{Column Height}}{\Delta} = \frac{(V_{0.5})\ (V_{0.159})}{(V_{0.5} - V_{0.159})^2}$$

where Δ = effective height of the theoretical plate (cm)

$V_{0.5}$ = column volumes to $C/C_o = 0.5$

$V_{0.159}$ = column volumes to $C/C_o = 0.159$

and

$$\Delta = 1.64\ r + \frac{\delta}{\delta + \beta}\ 2 \cdot \frac{0.142\ r^2\ \overline{F}}{D_S} + {\frac{\delta}{\delta + \beta}}^{2} \cdot \frac{0.266\ r^2\ \overline{F}}{D_L(1 + 70\ r\ \overline{F})}$$

where δ = amount of adsorbed cation per cm^3 of zeolite divided by amount of cation per ml of solution in equilibrium with the zeolite

β = void fraction of the column of zeolite

---

(16) E. Glueckauf. "Principles of Operation of Ion Exchange Columns," Ion Exchange and Its Applications, pp. 34-46. 1955. London. Soc. of Chem. Industry.

(17) E. Glueckauf. "Theory of Chromatography, Part 9, The Theoretical Plate Concept in Column Separations," Trans. Faraday Soc., vol. 51, pp. 34-44. 1955.

(18) J. R. Vinograd, J. W. McBain. "Diffusion of Electrolytes and of the Ions in Mixtures," J. Am. Chem. Soc., vol. 63, pp. 2008-2015. 1941.

r = radius of particle in cm

$\overline{F}$ = linear flow velocity of the feed solution above the bed (cm/sec)

D_S = particle diffusion coefficient (cm^2/sec)

D_L = liquid diffusion coefficient (cm^2/sec).

Curves of D_L and D_S for Sr^{+2} as a function of temperature with simulated D2EHPA 1BP solution and 4A zeolite are shown in Figure 4. The D_S curve is plotted through D_S values calculated from experimental column data using well defined particle sizes. Reducing the flow rate and particle size appeared to increase the D_S values. Considerable variation in duplicate results probably reflects deviations in column operation (e.g., channeling).

Figures 5 and 6 show calculated and experimental breakthrough curves with 4A and simulated D2EHPA 1BP feed. The particle size was not well defined in these experiments so a weighted average particle size was used for the calculated curves. Column volumes to $C/C_o = 0.5$ were determined from estimates of zeolite strontium loading calculated from binary exchange data.(19)

High-Level Waste Treatment, Solvent Extraction

(G. L. Richardson)

(Not Charged to Division of Reactor Development)

CSREX Waste Extraction Process Development

Pilot plant solvent extraction studies continued on the development of the CSREX and D2EHPA processes for removal of long-lived heat generating fission products from Purex wastes. Development studies resulted in firming

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(19) E. R. Irish. Quarterly Progress Report - Research and Development Activities - Fixation of Radioactive Residues - April–June, 1963, HW-78188. July 1963.

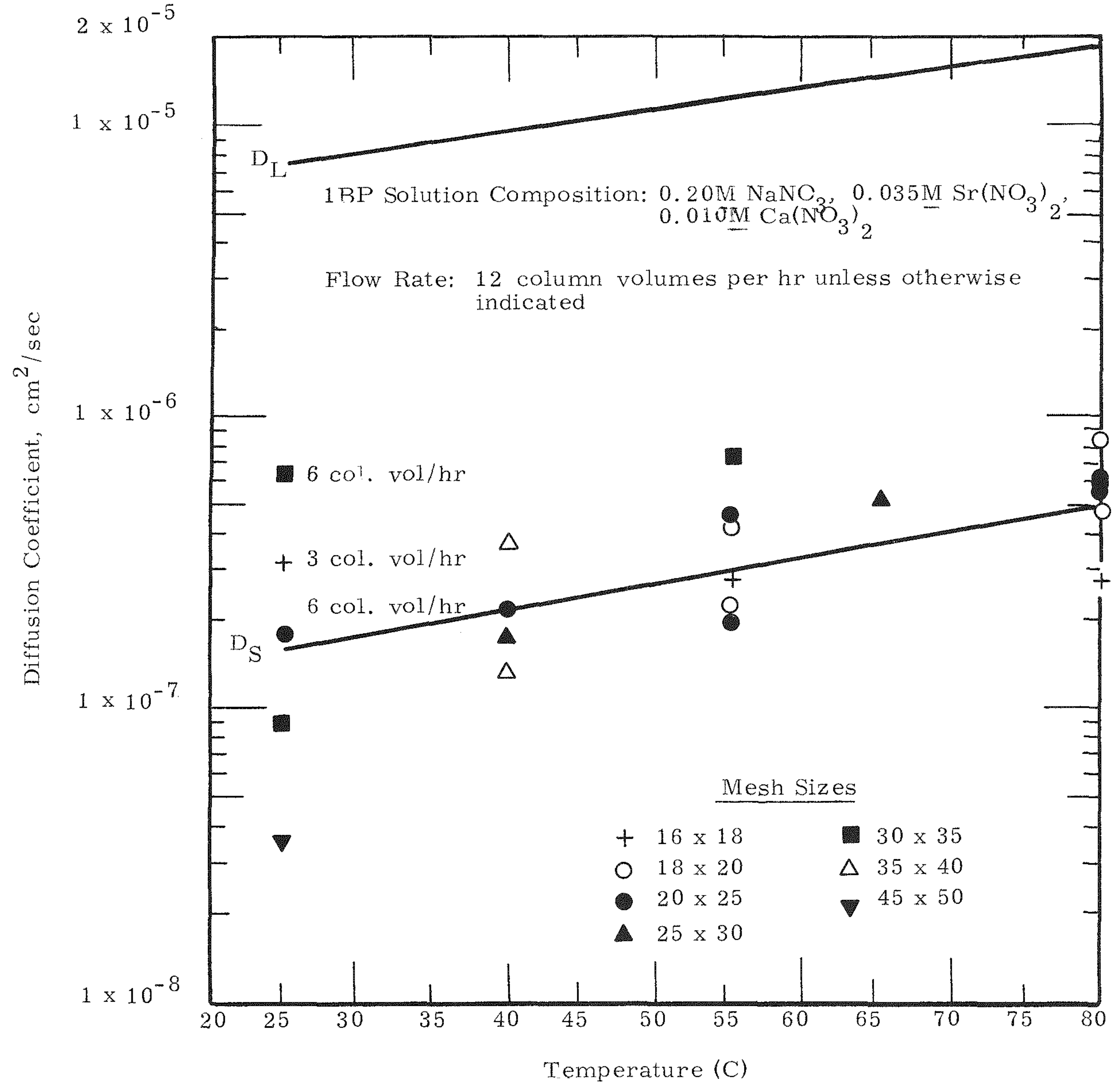
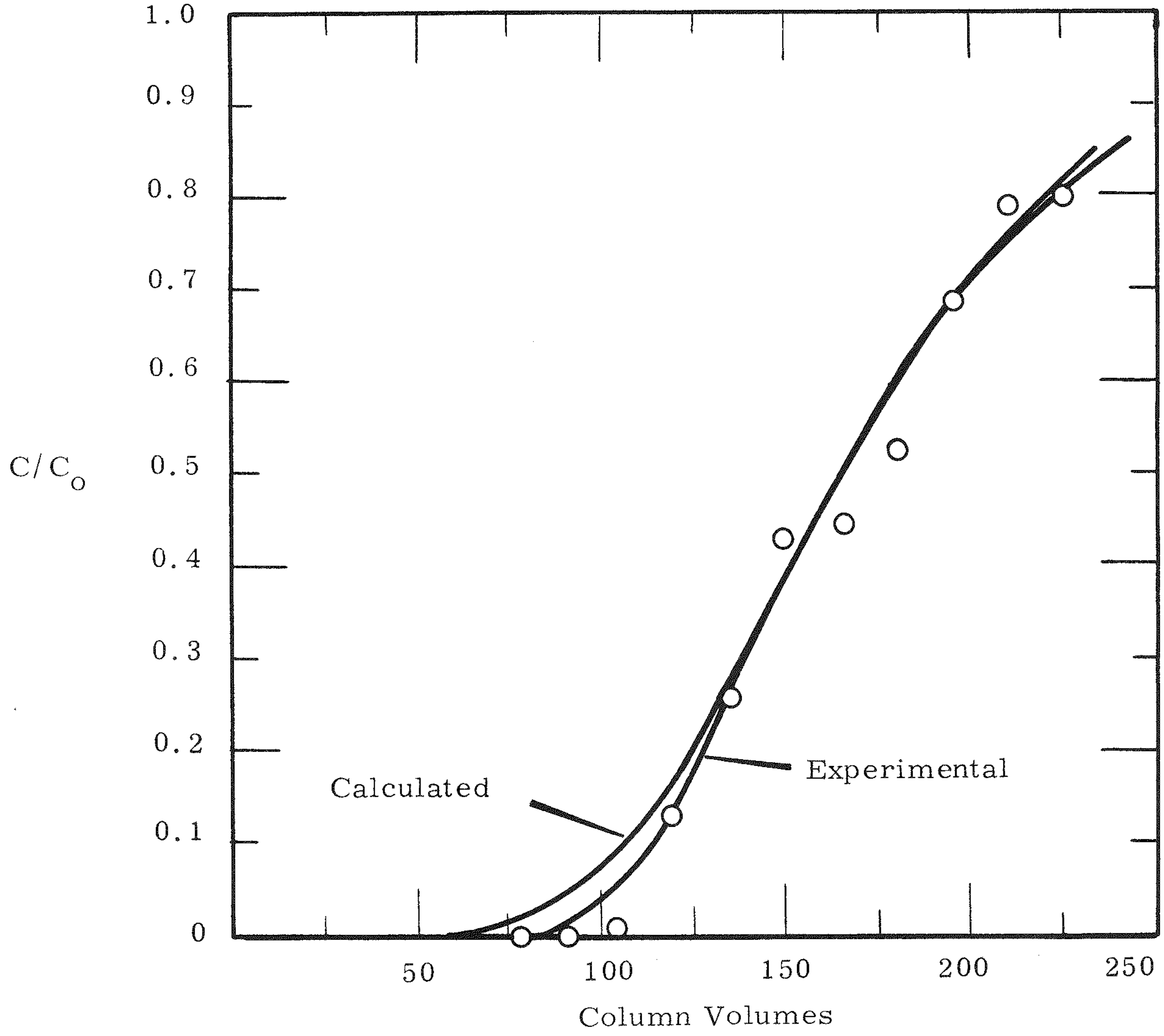


FIGURE 4

Variation of Liquid (D_L) and Particle (D_S) Strontium Diffusion Coefficients with Temperature for 4A and a Simulated D2EHPA 1BP Solution

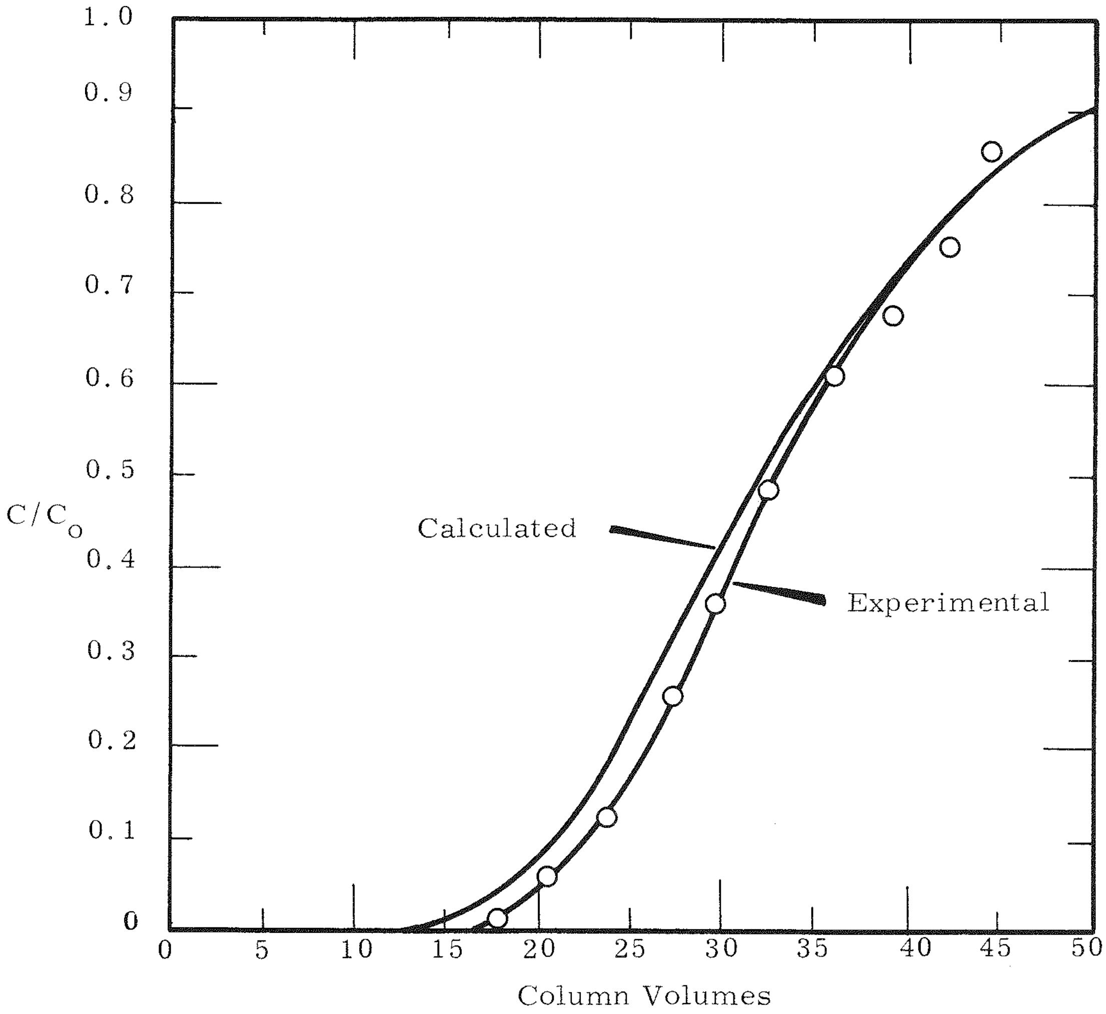


Conditions:

Feed	0.04$\underline{M}$ $NaNO_3$, 0.007$\underline{M}$ $Sr(NO_3)_2$, 0.002$\underline{M}$ $Ca(NO_3)_2$
Flow Rate	60 Column Volumes/Hour
Column Size	1.9 cm Diameter, 21 cm Height
Exchanger Weight	50 g
Temperature	80 C

FIGURE 5

Calculated and Experimental Strontium Breakthrough Curves for 20 x 50 mesh 4A Zeolite and Dilute Feed



Conditions:

Feed	0.20$\underline{M}$ $NaNO_3$, 0.035$\underline{M}$ $Sr(NO_3)_2$, 0.010$\underline{M}$ $Ca(NO_3)_2$
Flow Rate	12 Column Volumes / Hour
Column Size	1.9 cm Diameter, 21 cm Height
Exchanger Weight	50 g
Temperature	80 C

FIGURE 6

Calculated and Experimental Strontium Breakthrough Curves for 14 x 30 mesh 4A Zeolite and Simulated D2EHPA 1BP Solution up pulse column design details which are interchangeable between the CSREX and the D2EHPA solvent systems. These details are summarized in Table IX.

TABLE IX

PULSE COLUMN SPECIFICATIONS FOR CSREX AND D2EHPA PROCESSES

Column	Nozzle Plate Dimensions Cartridge Height, feet	Plate Spacing, inches	Hole Diameter, inch	% Free Area	Nozzles Point	Amplitude Frequency Range (in) (cycle)/min	Nominal Volume Velocity GPH/ft^2
1A	14	2	3/16	23	Down	30 - 110	400
1S	10	2	1/8	10	Up	30 - 110	280 - 330
1B	14	2	1/8	10	Up	30 - 110	410 - 475
1C	10	2	1/8	10	Up	30 - 110	270 - 320

A separate sodium scrub column (1S) is required because of the wide variety of flowsheets that might be used and the difficulty of guaranteeing a suitable match between the optimum operating conditions in both the scrub and extraction sections of a single dual-purpose column. The sodium contamination in the strontium product of the pilot plant columns was reduced about 10-fold with this flowsheet modification, yielding a product containing slightly less than a 2 to 1 mole ratio of sodium to strontium.

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