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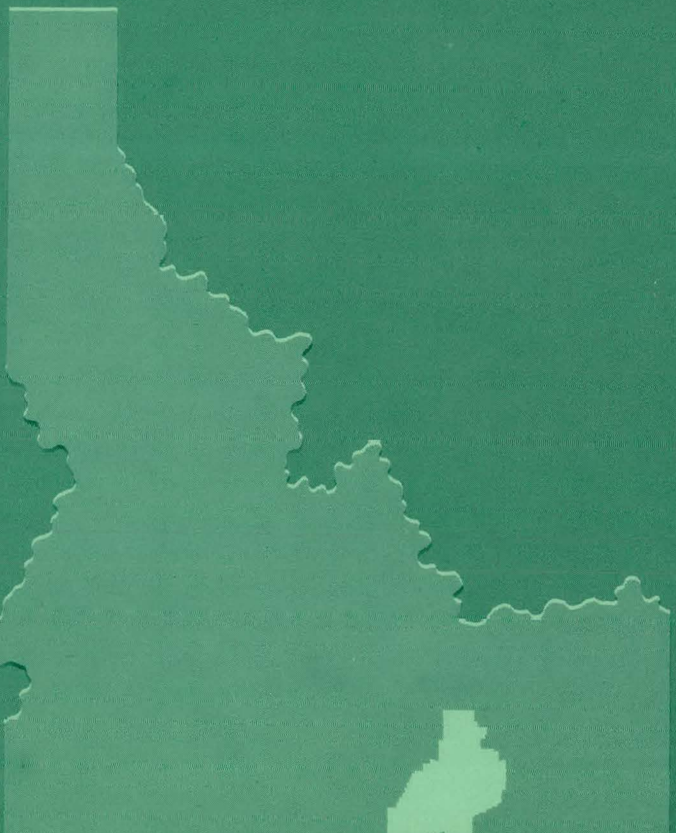
CESIUM REMOVAL FROM ACIDIC RADIOACTIVE WASTE SOLUTIONS

M. W. Wilding

April 14, 1961



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Chemical Development Branch

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CESIUM REMOVAL FROM ACIDIC RADIOACTIVE WASTE SOLUTIONS

M. W. Wilding

ABSTRACT

This report describes laboratory research to develop a method for removing cesium from highly acidic, radioactive, aqueous, fuel-reprocessing waste solutions by ion exchange on ammonium phosphomolybdate (APM). The solubility in acidic solutions and the thermal decomposition of APM were studied. Batch-type equilibrium experiments demonstrated the effect on cesium removal of acid concentration, aluminum nitrate concentration, temperature, sodium and potassium ions, concentration of cesium, and contact time. Ion exchange column experiments using APM on a silica gel carrier indicated that cesium was removed effectively from simulated, highly acidic waste solutions by adsorption on an APM-silica gel column, and that one gram of APM could remove more than 99.9% of the cesium from 0.9 liter of synthetic ICPP waste. While other means of adsorbing cesium from aqueous solutions are known, this is believed to be the first successful adsorption from highly acidic solutions.

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CESIUM REMOVAL FROM ACIDIC RADIOACTIVE WASTE SOLUTIONS

M. W. Wilding

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CESIUM REMOVAL FROM ACIDIC RADIOACTIVE WASTE SOLUTIONS

M. W. Wilding

I. SUMMARY

The removal of cesium from highly acidic, radioactive waste by ammonium phosphomolybdate (APM) was demonstrated by laboratory experiments to be chemically feasible. Distribution coefficients of 1000 or greater were obtained for cesium in batch-type equilibrium experiments from solutions simulating ICPP aluminum nitrate waste as well as dilute solutions of hydrofluoric, sulfuric, and hydrochloric acids.

Batch-equilibrium type experiments indicated that extraneous salts such as aluminum nitrate in the range 0.01 to 1.0$\underline{M}$ and sodium nitrate and potassium nitrate in the range 1.1×10^{-4} to 0.11$\underline{M}$ and 6.7×10^{-5} to 0.067$\underline{M}$ respectively had very little effect on cesium adsorption. Similar equilibrium experiments to determine the effect of cesium concentration resulted in a distribution coefficient of 3600 ± 100 for cesium in the concentration range 1.6×10^{-5} to 0.034 grams of cesium per liter of waste. In the concentration range 0.034 to 68 g Cs/l the distribution coefficient decreased gradually to a value of 3. Ammonium phosphomolybdate had a measured solubility of 0.5 to 0.7 g/l in 0.1$\underline{N}$ nitric, hydrochloric, and sulfuric acids and 4.5 g/l in 0.1$\underline{N}$ hydrofluoric acid. At higher acid concentrations the solubility of APM increased, especially in hydrofluoric acid. The solubility of cesium phosphomolybdate (CPM) was one to two orders of magnitudes less than that of APM in acidic solution. The most important single factor affecting the removal of cesium from solution appeared to be the solubility of APM and CPM in acidic solutions.

Silica gel was used effectively as a binder in column experiments to hold the APM <u>in situ</u> at flow rates up to 70 gal/(ft^2)(hr). A decontamination factor of 5.0×10^3 ± 50% (the limit of detection for the concentration of Cs^{137} used) was obtained for the first 25 column volumes using 0.18 to 1.98 mm diameter particles at a flow rate of 70 gal/(ft^2)(hr) with a synthetic waste solution containing 0.1$\underline{N}$ nitric acid, 0.06 g/l cesium, and 2$\underline{M}$ aluminum nitrate. The laboratory data indicated that the column capacity was markedly influenced by the flow rate of the feed solution through the column due to the solubility of

APM in the acid solutions. Optimum conditions would utilize the fastest flow rate and the smallest particle size consistent with adequate cesium removal and equipment performance.

II. INTRODUCTION

One of the major problems in the field of atomic energy today is the disposition of highly radioactive wastes. These wastes are presently stored in underground tanks and present a potential hazard in that corrosion, seismic disturbances, mechanical failures in the equipment, explosions, etc., may result in the spread of radioactive material to the environs. Attempts are being made throughout the atomic energy industry to remove economically the long-lived radioisotopes from the bulk of the waste material and store these radioisotopes as a solid in safe, compact storage. The removal of cesium-137--one of the dominant, long-lived (30 year half-life), radioactive nuclides--would eliminate during storage a potentially volatile fission product (above 650°C) and long-lived heat producer and simplify, to some extent, disposal of the remaining waste constituents.

This report presents the results of laboratory studies to develop an ion exchange method for removing cesium from acidic wastes with ammonium phosphomolybdate (APM). Ammonium phosphomolybdate was chosen for this investigation because of its high selectivity for cesium in an acidic medium.

III. LITERATURE REVIEW

The ion exchange property of APM is described by Buchwald and Thistlethwaite(1) as an isomorphous exchange of cesium ions for ammonium ions in the crystal lattice. According to Smit(5), "the phosphomolybdate complex ion, $(PMo_{12}O_{40})^{-3}$, consists of a hollow sphere formed by the twelve MoO_6 octahedra with the PO_4 group in the center of the crystal structure of the ammonium salt of this ion. The ammonium ions with associated water molecules are probably fitted in between these spheres of negative ions thus accounting for the cohesion of these ions". The large spaces that probably exist between these spheres permit ions such as cesium to enter and replace ammonium ions. The monovalent ions (Na^{+}, K^{+}, Rb^{+}, and Cs^{+}) will exchange for the ammonium ions on APM, as shown by Smit(5), with cesium being exchanged more easily than the others. The specificity of APM for cesium is probably due to the small size of the hydrated cesium ion which permits the cesium ions to approach the exchange sites more easily than the other monovalent ions. The exchange is very rapid and obeys the mass action law. The adsorbability of monovalent ions is usually little affected by the presence in solution of large quantities of nonexchangeable or weakly exchangeable ions.

In the chromatographic experiments of Smit, et al.(3) one gram of asbestos per gram of APM was used as a binding material to hold the APM in situ. Efforts made to convert APM into large stable crystals without the aid of a binding material met with little success. The formation of the very fine crystals (usually smaller than 200 mesh) may be attributed to the weak crystal faces resulting from the peculiar structure of the APM crystals.

Smit, et al.(4) indicated that small samples of APM irradiated for one week in the core of the BEPO pile at Harwell exhibited a slight photochemical reduction of the APM when the sample was mixed with a small quantity of water. A dry sample and samples wetted with 2N nitric acid and 2N hydrochloric acid showed no sign of reduction. However, the ion-exchange properties of the samples were not examined after irradiation.

There are several advantages in using APM as an inorganic ion exchanger; some of which have been pointed out by Smit, et al.(4). The principal advantage is the high selectivity of APM for cesium in acidic solution. Ammonium phosphomolybdate is one of the best known ammonium salts of the 12-heteropolyacids as well as one of the easiest to prepare in large quantities. It is also one of the cheapest in price ($1.75/lb)(2).

IV EXPERIMENTAL

The APM that was used in the laboratory experiments was prepared by precipitation in the laboratory or obtained commercially. Samples of APM from both sources were examined by X-ray diffraction and were determined to be $(NH_4)_3P(Mo_3O_{10})_4 \cdot 3H_2O$ by comparison with the ASTM standard card file. No differences in the chemical and physical properties of the APM from the two sources were observed.

Two principal methods used to study the ion exchange reaction of cesium with APM were batch equilibrium experiments and fixed-bed column experiments. High pressure drops prevented the use of the fine particles of APM without a carrier in the column experiments. One means of overcoming these high pressure drops is by the use of silica gel as a carrier for the APM.

A. Exchange Capacity of APM

The total exchange capacity of APM was determined as follows: First, the APM was saturated with successive washings of a concentrated solution of cesium nitrate traced with cesium-137. The cesium phosphomolybdate (CPM) formed during these washings by the exchange of cesium ions for ammonium ions was then washed with ethyl alcohol to remove the excess cesium nitrate solution. The cesium was then replaced with ammonium ions by using an ammonium nitrate solution. The exchange capacity was determined by measuring the amount of cesium in the ammonium nitrate wash solution. The total exchange capacity of APM was determined by this method to be 1.6 milliequivalents (211 mg) of cesium per gram of APM, which corresponds to replacement of essentially all the ammonium ions in the APM.

B. Solubility of APM

Solubility measurements were made of APM in nitric, sulfuric, hydrofluoric, and hydrochloric acids of various concentrations by employing a radiochemical technique. The phosphate in the APM was labeled with P^{32} by dissolving APM in ammonium hydroxide, adding P^{32} tracer, and reprecipitating the original compound with nitric acid. Excess solution was washed off, and the APM was dried for immediate use. One gram of

APM was mixed with 20 milliliters of acid, and the solubility was determined by measuring the concentration of P^{32} in the acid solution.

Table 1 shows the measured solubilities of APM in various acid solutions of different normalities after 24 hours and 120 hours contact.

Table 1

SOLUBILITY OF APM IN ACIDS AT 25°C

Acid	Acid Normality	Concentration of APM in Solution at: 24 Hrs. (g/l)	120 Hrs. (g/l)
HNO_3	1	3.1	3.4
	0.1	0.36	0.58
	0.01	0.33	0.58
H_2SO_4	1	6.8	7.2
	0.1	0.55	0.68
	0.01	0.49	0.62
HCl	1	2.5	2.5
	0.1	0.75	0.52
	0.01	0.48	0.55
HF	1	>43.0	>43.0
	0.1	3.6	4.6
	0.01	0.78	2.6

The data in Table 1 indicate that APM has a low solubility in 0.1 and 0.01N nitric, hydrochloric, and sulfuric acids, and in 0.01N hydrofluoric acid, and moderate solubility in 1N nitric, hydrochloric, and sulfuric acids and in 0.1N hydrofluoric acid. In all acids studied, the solubility of APM increased with increasing acid concentration.

C. Solubility of Cesium Phosphomolybdate

The solubility of cesium phosphomolybdate (CPM) was measured by batch equilibrium experiments using CPM on a silica gel carrier. All measurements were made by determining the concentration of cesium-137 in the supernatant liquid. Two grams of CPM-silica gel were placed in 100 milliliters each of water, 0.1N and 1.0N nitric acid, and in similar solutions saturated with APM. Each sample was agitated once a day, and aliquots were taken at intervals over a period of 168 hours to determine the solubility of CPM as a function of time. The results of the solubility experiments are shown in Table 2.

Table 2

SOLUBILITY OF CESIUM PHOSPHOMOLYBDATE AT 25°C

		Concentration of CPM in Solution, g/l				
Hrs. of Contact	H_2O	APM Sat'd H_2O	0.1N HNO_3	APM Sat'd 0.1N HNO_3	1.0N HNO_3	APM Sat'd 1.0N HNO_3
24	0.0050		0.0113		0.0229	
48	0.0105	0.0076	0.0152	0.0069	0.0296	0.0243
72	0.0161	0.0090	0.0161	0.0081	0.0295	0.0242
168	0.0258	0.0096	0.0160	0.0086	0.0350	0.0276

A comparison of the data in Table 2 with the data in Table 1 shows that the solubility of CPM, as indicated by the concentration of cesium in solution, is one to two orders of magnitude less than the solubility of APM.

D. Adsorption of Cesium in Equilibrium-Type Experiments by APM

Batch equilibrium-type experiments were used to study the ion-exchange reaction of cesium with APM. These equilibrium-type experiments were designed to study the effects of acidity, cesium concentration, solution composition, concentration of sodium and potassium ions, and temperature upon cesium adsorption.

The extent of the cesium adsorption was expressed as a distribution coefficient (Kd) where

$$\text{Kd} = \frac{\text{Grams of cesium/gram of solid}}{\text{Grams of cesium/milliliter of solution}}$$

The ratio of solid to liquid in all experiments was one gram per 20 milliliters.

1. Effect of Time on Cesium Adsorption by APM

Solutions containing 2M aluminum nitrate, 1.5N nitric acid, and tracer levels of cesium-137 were equilibrated with APM-silica gel and samples were taken periodically to determine the effect of contact time on the adsorption of cesium.

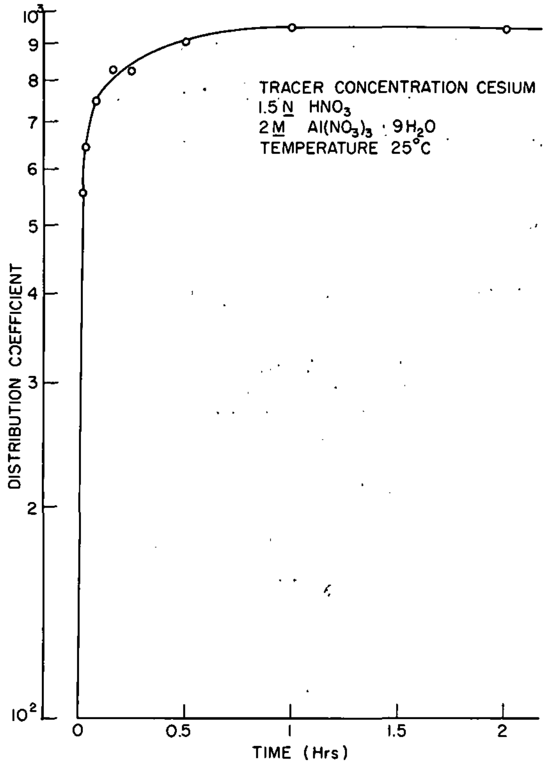

Fig. 1 - Effect of Time on Cesium Adsorption by APM

Fig. 1, which shows the distribution coefficients as a function of time, indicates that after a one-hour period, equilibrium, for all practical purposes, had been reached. For this reason, most of the subsequent batch equilibrium experiments used a one hour or greater equilibration time.

2. Effect of Temperature on Cesium Adsorption by APM

All solutions used in this experiment contained 0.06 g/l cesium as cesium nitrate in 1M aluminum nitrate and 1.0N nitric acid; the contact time was one hour.

The results of this experiment are shown in Table 3.

Table 3

EFFECT OF TEMPERATURE ON CESIUM REMOVAL BY APM

Temperature °C	Distribution Coefficient
2	1.2×10^3
20	1.1×10^3
50	1.5×10^3
83	1.2×10^3

There was no difference not attributable to experimental error in the distribution coefficients at the temperatures studied. This indicates that under these conditions temperature probably has little, if any, effect on cesium adsorption. However, in fixed-bed columns with high flow rates, the temperature effect may be important.

3. Effect of Acids on Cesium Adsorption by APM

The effect of acids on cesium adsorption was determined by using tracer concentrations of cesium-137 and a one hour equilibrium contact. The calculated distribution coefficients are shown in Table 4.

Table 4

EFFECT OF ACID ON CESIUM ADSORPTION BY APM AT 25°C

Acid	Acid Normality	Distribution Coefficient
HNO_3	0.001	2.0×10^4
	0.01	1.7×10^4
	0.1	3.7×10^4
	1.0	2.0×10^4
	1.5	5.0×10^3
	7.5	8.4×10^2
	12.0	3.9×10^2
H_2SO_4	3.0	1.1×10^3
HCl	0.1	3.5×10^3
	1.0	2.0×10^3
HF	1.0	2.4×10^3

The distribution coefficients for cesium were greater than 1000 for all of the acids at the different normalities tested except nitric acid at 7.5 and 12N. A distribution coefficient was obtained in 1N hydrofluoric acid only because the short contact time was insufficient to dissolve the APM.

4. Effect of Aluminum Nitrate on Cesium Adsorption by APM

The effect of aluminum nitrate on cesium adsorption was studied using aluminum nitrate solutions and aluminum nitrate-nitric acid solutions of various concentrations. Tracer concentrations of cesium and a 24 hour contact were used. The results are shown in Table 5.

Table 5

EFFECT OF ALUMINUM NITRATE ON CESIUM ADSORPTION BY APM AT 25°C

Concentration		Distribution Coefficient
$Al(NO_3)_3$		
1M		1.71×10^4
0.1M		2.27×10^4
0.01M		2.87×10^4
$Al(NO_3)_3$	+ HNO_3	
1.0M	1N	1.05×10^4
0.1M	1N	1.34×10^4
0.01M	1N	1.65×10^4

The data in Table 5 indicate that there was an apparent slight decrease in cesium adsorption with increasing concentration of aluminum nitrate. The addition of 1N nitric acid to 1.0M aluminum nitrate solution resulted in a further decrease of cesium adsorption. However, decreasing the aluminum nitrate concentration had less effect on the adsorption of cesium when nitric acid was present in the system.

5. Effect of Sodium and Potassium on Cesium Adsorption by APM

The solutions used for these experiments all contained 0.06 g/l cesium in 1N nitric acid. The sodium and potassium ions were supplied as the nitrate salts. The contact time was one hour.

Table 6 shows the distribution coefficients at each concentration of sodium and potassium ions.

Table 6

EFFECT OF SODIUM AND POTASSIUM NITRATES ON CESIUM ADSORPTION BY APM AT 25°C

Ion	g/l	Distribution Coefficients
Sodium	0.105	1.85×10^3
	1.05	1.41×10^3
	10.5	1.36×10^3
Potassium	0.103	1.09×10^3
	1.03	0.98×10^3
	10.3	0.59×10^3

The highest potassium and sodium concentration (>10.0 g/l) indicated a trend toward slight potassium and sodium interference with the adsorption of cesium. This may be due to the nearly equal hydrated radius of the potassium (5.32Å), sodium (7.90Å), and cesium (5.05Å) ions. The sodium ion (7.90Å) is larger than either the potassium or cesium ion; for this reason, it would not be expected to interfere as much as the potassium ion in the exchange reaction. This was indicated by the higher distribution coefficients for cesium in the solutions containing sodium ions.

6. Effect of Cesium Concentration on Cesium Adsorption by APM

The concentration of cesium as cesium nitrate was varied from tracer levels to 68.0 g/l in 2M aluminum nitrate and 1N nitric acid solution. The equilibration period for this experiment was 36 hours.

Fig. 2 shows the effect of cesium concentration on the distribution of cesium between the solid and the liquid. The distribution coefficients calculated from these data were essentially constant at 3600 (± 100) from tracer concentrations to 0.034 g/l of cesium and decreased gradually to a value of about three at 68 g/l of cesium. At the highest concentration of cesium (68 g/l) the amount of cesium removed from solution was equivalent to approximately 90% of the exchange capacity of the APM. However, the shape of the curve suggests that at these higher concentrations some other mechanism such as precipitation was also effective in removing cesium from solution.

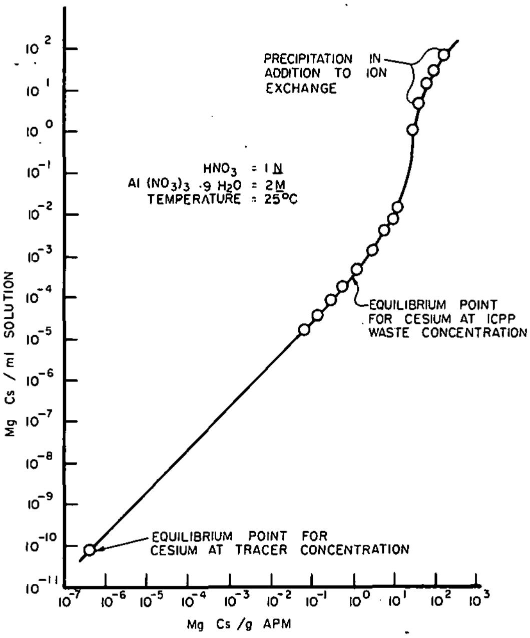

Fig. 2 - Equilibrium Distribution for Cesium in Synthetic ICPP Waste

E. Ion Exchange Column Experiments

The APM in these experiments was first dissolved in ammonium hydroxide; then a highly absorbent, decrepitation resistant, granular silica gel was soaked in the ammonium hydroxide-APM solution until the silica gel was saturated. The excess ammonium hydroxide-APM solution was drained off, and nitric acid was added to precipitate the APM within the pore spaces of the silica gel to hold it uniformly distributed in situ. By this carrier technique, high flow rates were obtained in a fixed-bed column without appreciable movement of solids and without high pressure drops.

Four column experiments were conducted using 300 cc columns, 76 cm long and 2.2 cm in diameter, each containing 120 g of silica gel carrier and 30 g of APM. The columns were connected to a SIGMA-motor pump to give flow rates of 5 to 70 gal/(ft^2)(hr) through the column. Two different particle sizes were used in the column--0.18 to 1.98 mm diameter and 2.0 to 6.4 mm diameter. The feed solutions for all column experiments contained 2M aluminum nitrate, 0.06 g/l of cesium traced with cesium-137 and either 1N or 0.1N nitric acid. Aliquots of each column volume of the effluent were collected with an automatic fraction-collector. The residence time of an individual increment of feed solution in the column varied from approximately two hours for a flow rate of 5-6 gal/(ft^2)(hr) to 12 minutes for a flow rate of 70 gal/(ft^2)(hr).

The results of the first two experiments are shown in Fig. 3 and 4. The nitric acid feed solutions were passed through APM-silica gel columns containing 2.0 to 6.4 mm diameter particles at a flow rate of 5-6 gal/(ft^2)(hr). The ratio of the cesium concentration in the column effluent to the cesium concentration in the column influent (C/C_0) was plotted against column volumes of effluent (total volume of APM-silica gel bed).

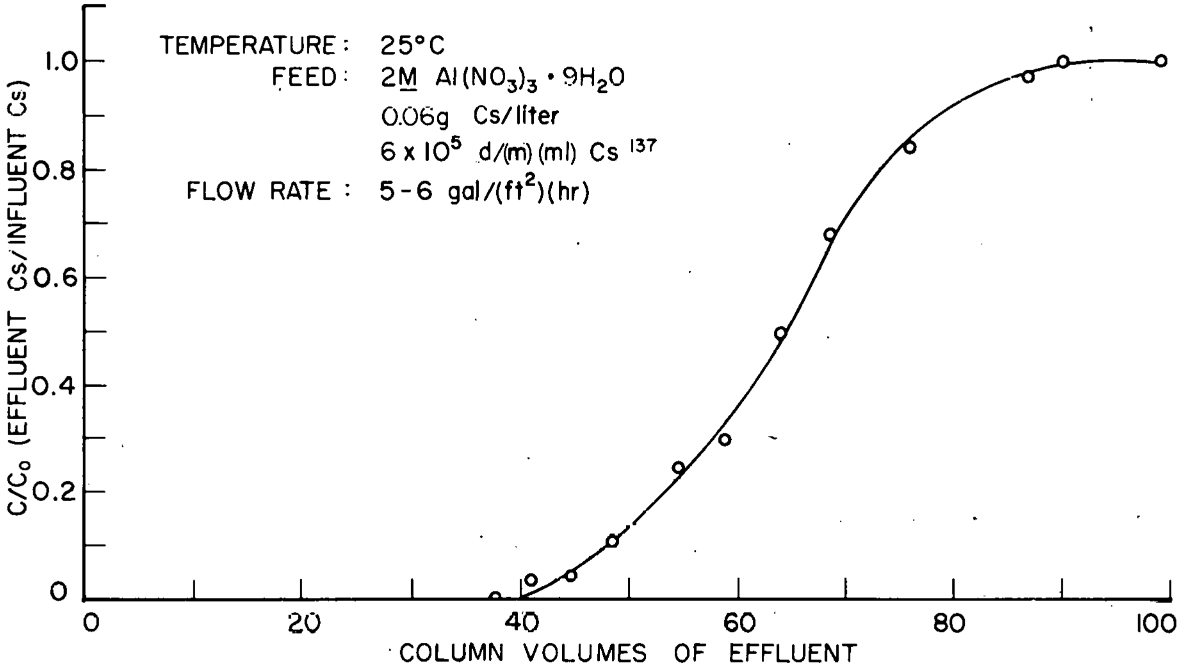

Fig. 3 - Breakthrough Curve for Cesium in an APM-SiO_2 Column--0.1N Nitric Acid Feed, >2 mm Diameter Particles

The 0.1N nitric acid feed resulted in 50% breakthrough of cesium ($C/C_o = 0.5$) at 65 column volumes. This was considered to be the column capacity for this particular type of feed solution. The average decontamination factor (D.F.) for the first 15 column volumes was $5.0 \times 10^3 \pm 50\%$. The 1.0N nitric acid feed resulted in 50% breakthrough at 36 column volumes and an average D.F. of $5.0 \times 10^3 \pm 50\%$ was obtained for 9 column volumes. Higher D.F.'s were not measurable with the concentration of cesium-137 used as a tracer.

An experiment using a column containing 2.0 to 6.4 mm diameter particles and the feed solution containing 0.1N nitric acid at a flow rate of 70 gal/(ft^2)(hr) resulted in a cesium D.F. of <5 in the first column volume. The D.F. remained at about the same value for at least 30 column volumes indicating that the interstices between the particles were so large that the cesium ions were not approaching near enough to the exchange sites at the faster flow rate for exchange to occur.

Fig. 5 shows the results of an experiment using the same feed solution at the same flow rate through an APM-silica gel column of smaller particle size (0.18 to 1.98 mm). Under these conditions, the D.F. was $5.0 \times 10^3 \pm 50\%$ for the first 24 column volumes, and 50% breakthrough occurred

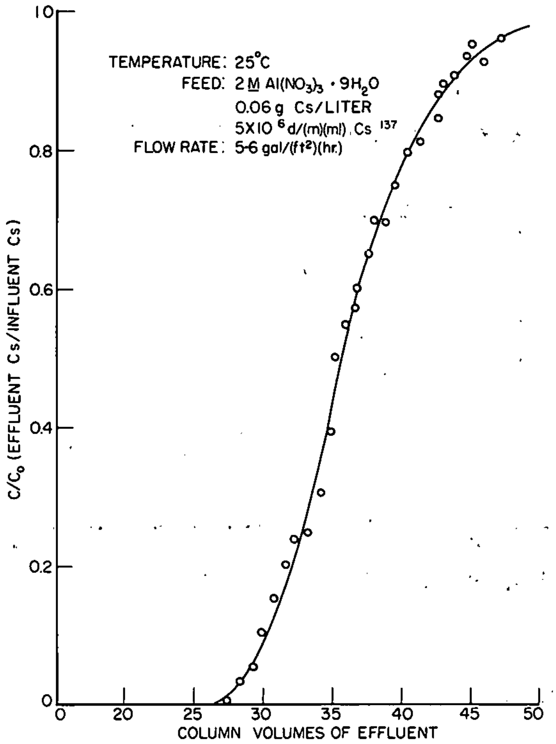

Fig. 4 - Breakthrough Curve for Cesium in an APM-SiO$_2$ Column--1.0N Nitric Acid Feed, >2 mm Diameter Particles

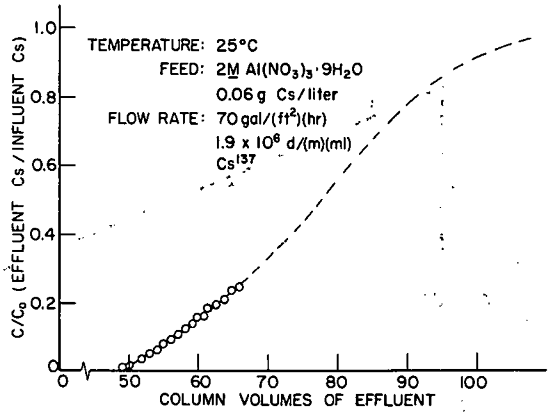

Fig. 5 - Breakthrough Curve for Cesium in an APM-SiO$_2$ Column--0.1N Nitric Acid Feed, <2 mm Diameter Particles

at 78 column volumes (an increase in capacity of 20% over that obtained with the large particles at the slower feed rate as shown in Fig. 3).

The equilibrium solubility data (Table 1) indicated that large volumes of acidic waste solution passing through a column should dissolve appreciable amounts of the APM. Consequently, the concentration of APM in the column effluents was determined by X-ray fluorescence. Fig. 6 shows the concentration of APM in the column effluent plotted as a function of column volumes of effluent for the feed solution containing 1.0N nitric acid. Fig. 7 shows similar data for the feed solution containing 0.1N nitric acid at two different flow rates and two different particle sizes.

The data in Fig. 5 and Fig. 7 demonstrate that the amount of APM dissolved from a fixed-bed column can be decreased markedly without reducing the removal efficiency for cesium by increasing the flow rate of the feed solution through the column and simultaneously decreasing the particle size of the APM-silica gel granules. Obviously, the optimum conditions should include the fastest flow rate and smallest particle size compatible with adequate cesium removal. Additional experimental work would be required to establish these optimum conditions.

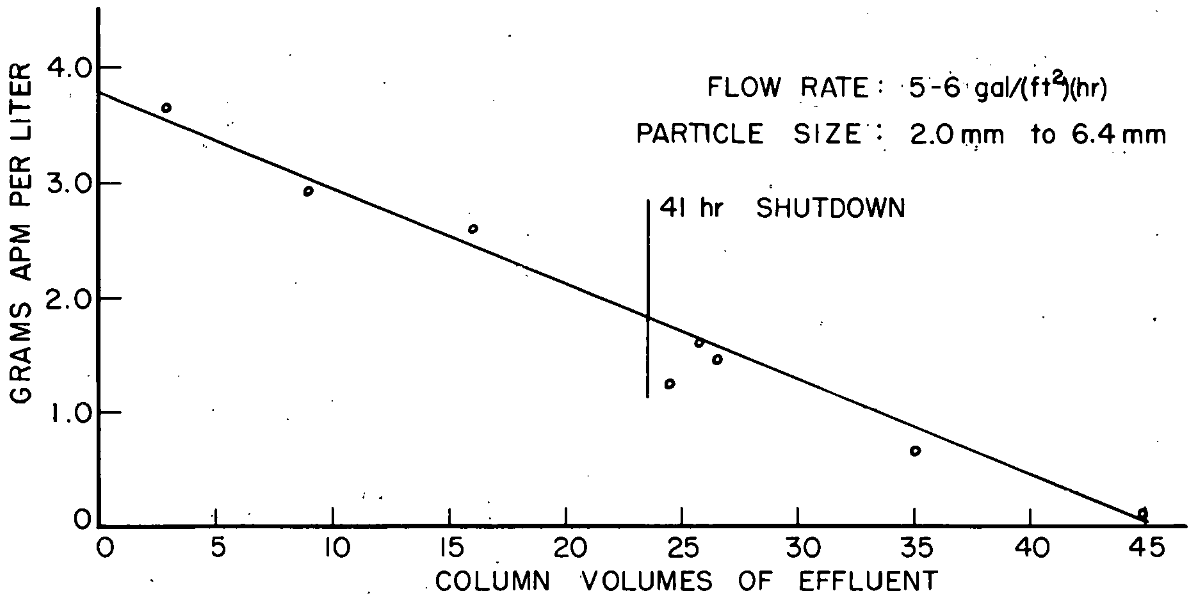

Fig. 6 - Concentration of APM in Column Effluent (1.0N HNO_3) at 25°C

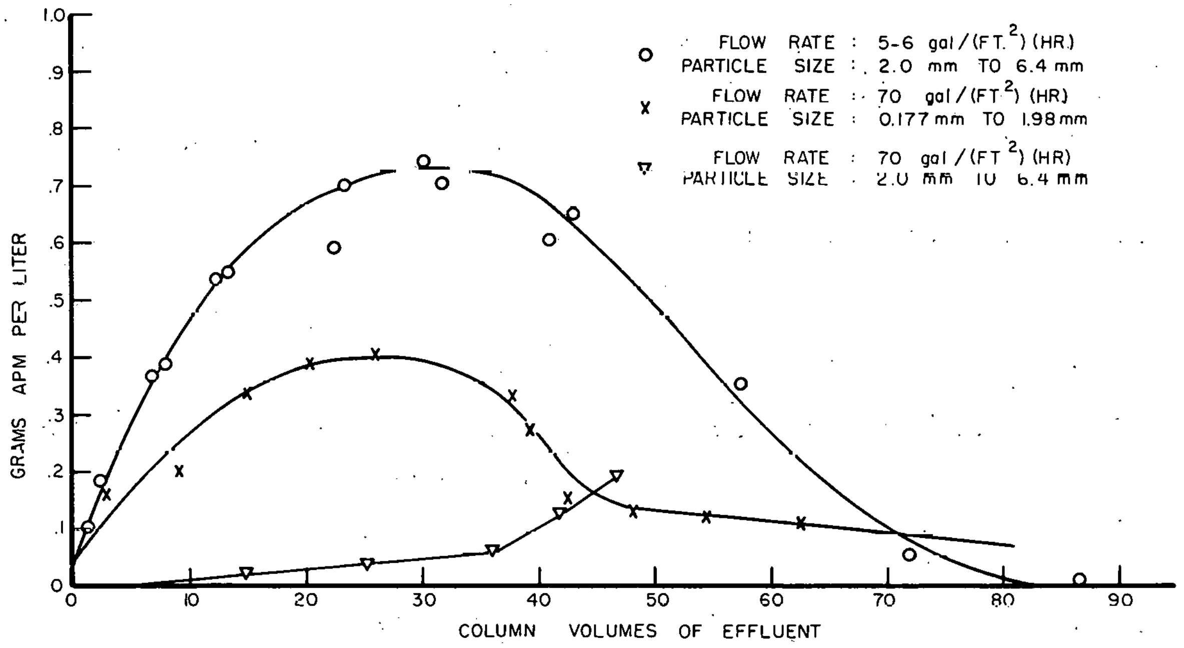

Fig. 7 - Concentration of APM in Column Effluent (0.1N HNO_3) at 25°C

F. Decomposition of Ammonium Phosphomolybdate

The storage of APM solid containing high concentrations of fission product cesium could result in the generation of appreciable heat by fission product decay. For this reason, the thermal decomposition of APM was studied in the laboratory.

Ammonium phosphomolybdate was heated at various temperatures to a maximum temperature of 800°C in an alundum boat in the combustion tube of a high-temperature furnace, and the chemical and physical changes were observed as a function of temperature. The solid was heated at various temperatures for one hour and then examined by X-ray diffraction to identify the product.

Ammonium phosphomolybdate was stable from 20°C to about 425°C as indicated by lack of a color change and by X-ray diffraction. A gradual color change occurred during heating from 425°C (yellow) to 550°C (black) and then to 750°C (gray). In the range of 425°C to 750°C ammonia and phosphorus were volatilized; a residue of molybdenum oxide remained. Above 750°C the molybdenum oxide sublimed and formed greenish-white, needle-like crystals on the wall of the combustion tube. These crystals were identified as molybdenum trioxide by X-ray diffraction.

No experimental work was done in this study to determine the radiation stability of APM. It is expected that the radiation stability of APM will be adequate based on preliminary studies made by Smit, et.al.(4) at Harwell. However, the ion exchange properties of APM under irradiation should be verified experimentally.

V. CONCLUSIONS

The results of the laboratory experiments indicate that the inorganic compound APM can be used to remove cesium from highly acidic, aqueous radioactive wastes. Cesium was removed effectively by APM from waste containing 1N nitric, hydrochloric, sulfuric, and hydrofluoric acids. However, the solubility of APM is too great in 1N hydrofluoric acid to consider removing cesium without first complexing the fluoride ions. Better cesium removal can be obtained in the presence of all of the acids studied by lowering the acid concentration to 0.1N or less.

The removal of cesium by APM was not appreciably affected by the presence of up to 2M concentrations of aluminum nitrate. Monovalent cations in concentrations likely to occur in acidic wastes caused very little interference with the exchange reaction. Cesium ions have the smallest hydrated radius of the monovalent ions that are normally found in acidic, radioactive waste solutions and are preferentially exchanged for the ammonium ions. The total exchange capacity of APM in neutral solution with respect to cesium was determined by equilibrium saturation to be 1.6 milliequivalents (211 mg) of cesium per gram of APM compared to a calculated value of 213 mg Cs/g APM for $Cs_3 P(Mo_3O_{10})_4 \cdot 3H_2O$. The solubility of APM is increased by the presence of 1N HNO_3. Since the volume of an acidic waste containing a given quantity of cesium is large, the available APM on the column is reduced by solubility. The final exchange capacity in 1.0N nitric acid, 0.06 g/l cesium solution is 10.8% of the theoretical value calculated on the basis of the initial charge of APM. The use of synthetic waste containing 0.1N nitric acid under similar conditions increased the utilization of the exchange capacity to 18.6%. The same waste solutions in a column containing smaller APM particles (0.18 to 1.98 mm) and with an increased flow rate (increased from 5 to 70 gal/(ft^2)(hr)) increased the utilization of the exchange capacity of APM to 24%. Still smaller particles at higher flow rates could possibly be used in a pulsed bed, liquid-solid contactor to increase further the utilization of the exchange capacity.(6)

Assuming only 25% utilization of the original exchange capacity of the column as charged, one gram of APM would be capable of removing all of the cesium from about 0.9 liter of ICPP first extraction cycle waste from the aluminum-uranium fuel recovery process. The cost of chemicals for such a recovery process will be small and negligible compared to investment and operating costs.

The silica gel used in the column experiments to hold the APM uniformly distributed in situ showed no visible evidence of decrepitation in the column, and high flow rates were possible using 0.18 to 1.98 mm diameter APM-silica gel particles. Based on the solubility data for APM and CPM in acid solutions and the adsorption data for cesium, the flow rate through the column and particle size of the APM-silica gel particles will be the most important factors in obtaining maximum removal of cesium in fixed-bed columns.

Additional laboratory experiments will be required to determine the optimum particle size and flow rate for removing cesium from a specific waste solution for a given piece of equipment. Also the effect of a high radiation field on the stability of APM will be determined before it can be considered for a waste treatment process.

Only one brief attempt was made to elute adsorbed cesium from the APM-silica gel system with ammonium nitrate solution. The removal of an appreciable amount of cesium by eluting with ammonium nitrate solution was unsuccessful. However, greater than 90% of the cesium was removed by dissolving the APM with ammonium hydroxide. In general, the low cost of APM and silica gel materials and difficulty of eluting cesium indicated that the more feasible approach would be to store the APM-SiO_2-Cs as a solid. If recovery of cesium for industrial purposes were to be considered, further research to develop methods for separating cesium from the APM-SiO_2 should be undertaken.

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