

FISSION PRODUCT REMOVAL FROM MOLTEN SALT USING ZEOLITE*

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Abstract

Spent nuclear fuel (SNF) can be treated in a molten salt electrorefiner for conversion into metal and mineral waste forms for geologic disposal. The fuel is dissolved in molten chloride salt. Non-transuranic fission products in the molten salt are ion-exchanged into zeolite A, which is subsequently mixed with glass and consolidated. Zeolite was found to be effective in removing fission product cations from the molten salt. Breakthrough of cesium and the alkaline earths occurred more rapidly than was observed for the rare earths. The effluent composition as a function of time is presented, as well as results for the distribution of fission products along the length of the column. Effects of temperature and salt flow rate are also discussed.

Introduction

Electrochemical treatment is a nonaqueous method developed for treatment of spent nuclear fuel to place fission products and transuranium elements into suitable waste forms for disposal. This treatment includes high temperature electrotransport of uranium from the spent fuel to collector cathodes in an electrorefiner. The electrolyte is a molten eutectic mixture of lithium and potassium chlorides.(1, 2)

After fuel processing, rare earth and active metal fission products and the transuranium (TRU) elements retained in the electrolyte salt are removed by passing the salt over a bed of zeolite A. The fission products and TRU elements remain with the zeolite, and the effluent salt that is fission-product free is recycled to the electrorefiner. The zeolite is ground and blended at high temperature with anhydrous zeolite to fully occlude the salt, mixed with a glass, and hot pressed to form a solid monolith for geologic disposal. This method is now being experimentally evaluated.(3, 4)

Active metal cations are readily ion exchanged into zeolite A in molten salt systems when the zeolite is contacted with an excess of salt in a batch process.(3) Although the fission product cations are effectively concentrated in the zeolite, the fission product concentration in the salt is not reduced sufficiently to recycle the salt unless the weight ratio of salt to zeolite approaches one.(5) Treating larger volumes of salt by a batch process would require repeatedly contacting the salt with small amounts of zeolite. Because a substantial amount of the salt would adhere to the zeolite, recycle would not be possible. Consequently, two options exist for treating electrorefiner salt using zeolites: (1) occluding the waste salt into zeolite in a hot blending operation with no ion-exchange step or (2) concentrating the fission products in the zeolite in an ion-exchange column.

The performance of an ion-exchange column of zeolite for extracting fission products from the electrolyte salt is currently under study. Active metal fission products are readily ion exchanged into zeolite A from molten salt when the zeolite is contacted with an excess of salt.(1, 5, 6) Although high loadings of fission products are obtained, the exchange is slow, and the salt is not sufficiently low in fission products to be recycled(7). The zeolite ion-exchange column should concentrate the fission products more effectively than a simple batch contact, allowing a large fraction of the salt to be recycled and thus reducing the total volume of waste produced by the process. In initial tests, the zeolite column was effective in removing fission products from the salt and concentrating them in the zeolite.

Experimental Setup

Starting Materials

Zeolite A was obtained in pellet form from UOP Corp. The pellets contained between 5 and 10 wt% binder and were formed into 12 × 25 mesh beads. The pellets were dehydrated in

a step-wise fashion to 525°C under a nitrogen flow system and transferred directly to an inert atmosphere glovebox. There was no difference between the X-ray diffraction patterns of the initial and dehydrated materials.

The simulated waste salts were formed from reagent grade components (Johnson Matthey Co.), which were physically mixed, melted, filtered, and coarsely ground. The LiCl and KCl were obtained at the eutectic composition (42.5 wt% LiCl, melting point ~635 K) from APL Engineered Materials, Inc. The composition of the waste salt that was prepared for the batch tests (Salt 1) and initial column tests (Salt 2) is given in Table I. Salt 3 was the composite product of the ion-exchange column tests. This salt has a much lower fission product concentration than Salt 2 and was used in one column test (No. 1110). Salt 2 was used in all of the other column tests.

Table I: Salt Composition (Wt%)

Component	Salt 1	Salt 2	Salt 3
KCl	45.3	49.5	51.6
LiCl	39.6	41.4	39.8
CsCl	3.7	2.3	1.0
BaCl ₂	1.4	1.0	0.6
SrCl ₂	0.6	0.4	0.2
CeCl ₃	0.7	1.9	1.2
LaCl ₃	1.1	0.6	0.4
NdCl ₃	1.0	3.9	2.1
PrCl ₃	0	0.8	0.5
YCl ₃	0.1	0.1	0.1
NaCl	6.0	0	2.6
KI	0.4	ND	ND

ND = Not Determined

Experimental Apparatus

Batch ion exchange tests were done to determine the operating conditions for the column system (Fig. 1). Zeolite pellets were first pre-loaded with LiCl-KCl eutectic salt. The pellets were placed in a metal basket and immersed in a large excess of LiCl-KCl eutectic salt at 725 K for 24 h. Ion-exchange kinetics were determined by then immersing the pre-loaded pellets in a large excess of waste salt 1 at 725 K. The pellets remained immersed for various lengths of time, between 30 min and 72 h, and were then removed from the salt hot. After cooling and in preparation for elemental analysis, the pellets were washed in deionized water, dried, ground, and re-washed. After a second drying, the sample compositions were determined by flame emission spectroscopy for cesium and inductively coupled plasma-atomic emission

spectroscopy for the other elements. Pellets from column tests were prepared for elemental analysis in the same fashion.

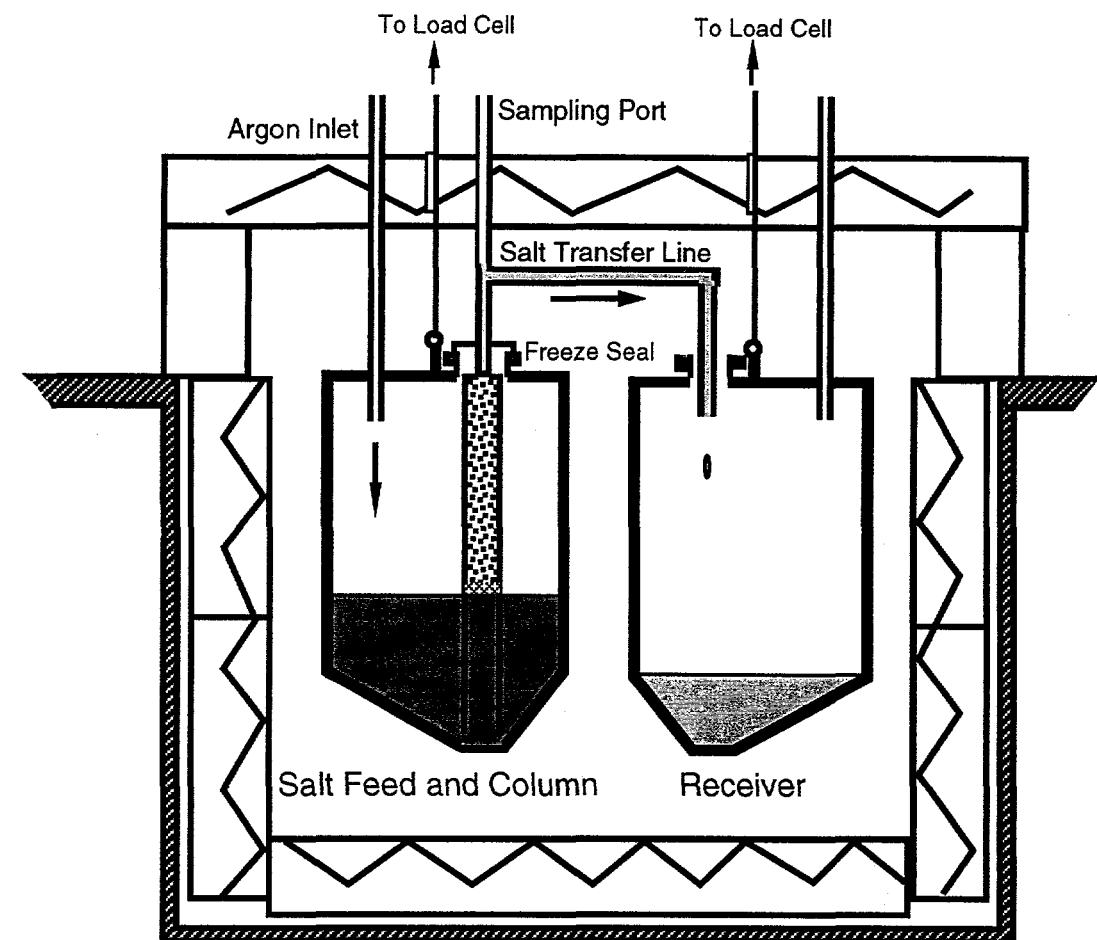


Figure 1. Schematic of Zeolite Column System

As shown in Fig. 1, the ion-exchange column system contains two tanks, the feed and receiver tanks, each of which is suspended from an inconel beam that sits on a pair of load cells. The tanks (16-in. long, 8-in. diameter) are identical so that each may function as the feed or receiver tank. This allows a single batch of salt to be reused several times before it is discarded. Located on top of each tank are a thermocouple well, an argon gas line connection, and an annular freeze seal cup, which contains a braze. A small cylindrical resistance heater used to melt the braze in the freeze seal is placed around each of the cups.

The tanks are located within an internally heated 26-in. diameter well which is accessible through a large glovebox. The exterior walls of the well are water-cooled. A heated shroud lies above the well, covering the transfer line and the top of the tanks. This shroud maintains the transfer line at high temperature and insulates the load cells and glove box from the hot column system. The entire area enclosed by the heaters is maintained above 400°C to prevent freezing of the salt (the eutectic LiCl-KCl salt melts at ~360°C).

The zeolite pellets are held in place in a stainless steel tube by a pair of metal mesh screens, forming the column. At the top of the tube is a Cajon© VCR fitting, which forms a gas-tight seal between the column and the transfer line. Columns with diameters of up to 2 in. may be used interchangeably with the same transfer line and tanks. Columns are removed from the transfer line by breaking the VCR connection. The transfer line contains a portion of the freeze seal and an inverted cup, which mates with the annular cup on the tank to form a gas-tight seal. When in position, the column sits within the salt tank, with the bottom of the zeolite lying just above the salt surface.

Argon gas is slowly added to the feed tank, forcing the salt up through the column through the transfer line to the receiver tank as the pressure increases. The transfer line contains a sampling port through which salt samples may be collected as the salt is transferred. The gas flow rate is controlled by a mass flow controller, while the liquid flow rate is monitored through changes in the masses of the two tanks, as measured by the load cells.

The zeolite pellets are pre-loaded with the eutectic salt by loading the feed tank with the LiCl-KCl eutectic salt and passing the salt through columns containing the dehydrated zeolite. The eutectic salt is pumped out of the tanks and replaced with the simulated waste salt, which is then passed through the pre-loaded zeolite.

Once removed from the heated zone, the zeolite column may be connected to a pressure flow apparatus, shown in Figure 2, so that residual interstitial salt can be removed from the surface of the column zeolite. The column is attached to an argon gas line in the apparatus by a Cajon© VCR fitting identical to that on the transfer line. The argon is pre-heated before it comes into contact with the zeolite column. Just above the fitting are heat shields. Below the column is a crucible, which is used to collect any salt that is removed. The entire apparatus fits in an 8-in. diameter well of the glovebox that is heated to 775 K. The column is pressurized until the glovebox pressure rises, indicating breakthrough of argon.

Results

Kinetics of Ion Exchange from Batch Tests

The time of contact between zeolite A and a large excess of molten salt has a significant effect on the final composition of the zeolite. As can be seen in Figure 3, uptake of cesium at 725 K is very rapid and reaches a nearly constant value after 1 to 2 h of contact. The alkaline earth cations, which are sorbed more slowly, go through a maximum concentration in the zeolite after 4 to 6 h. At longer times, the alkaline earths are displaced by the rare earths. The rare earths are retained most strongly by the zeolite but show the slowest exchange kinetics. At 72 h of contact, the uptake curve for the rare earths has not yet begun to plateau. Based on the results of batch ion exchange tests, low flow rates (on the order of 0.5 to 3 cm/min) were examined in the initial column tests.

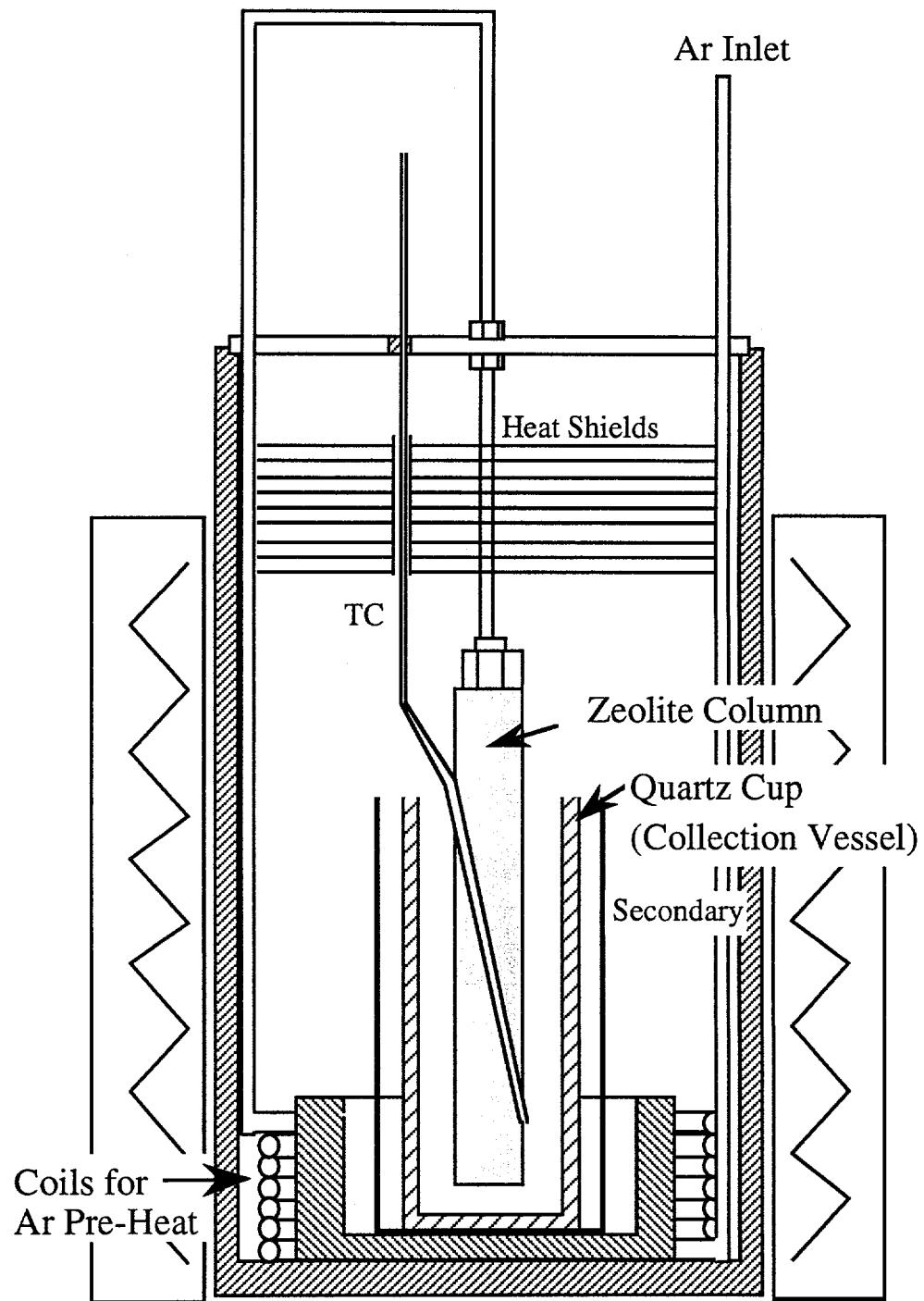


Figure 2. Schematic of Surface Salt Removal Apparatus

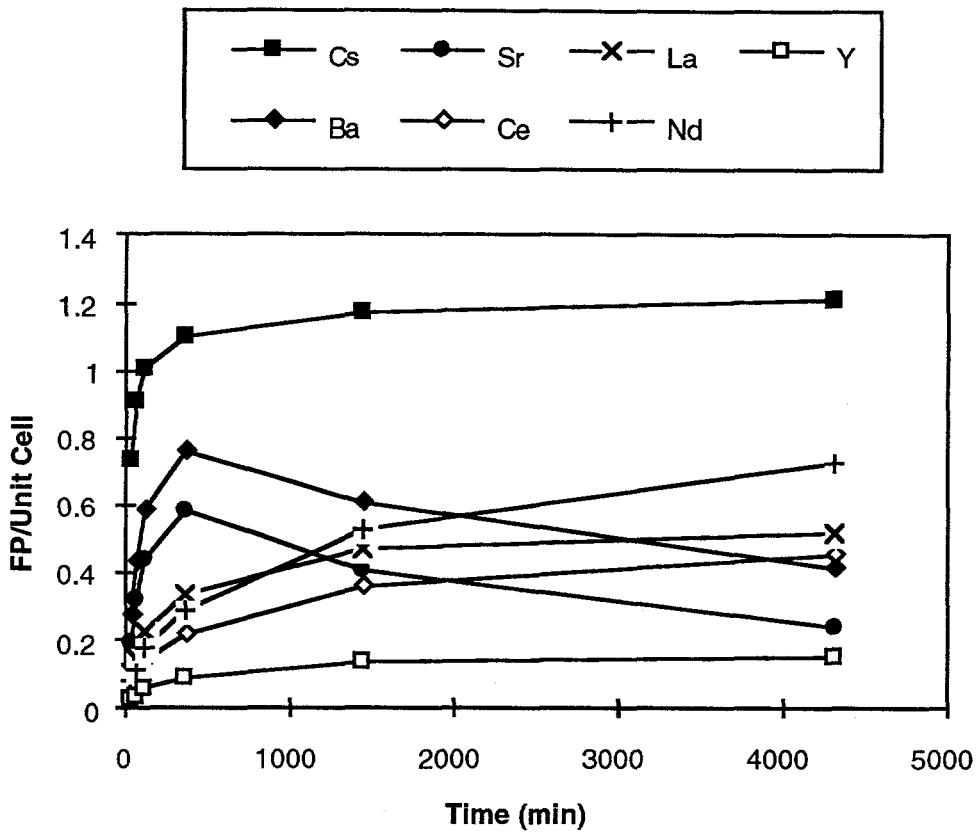


Figure 3. Fission Product (FP) Uptake versus Contact Time between Zeolite A and Waste Salt at 725 K

Ion Exchange with Waste Salt in a Zeolite Column

As part of the overall electrochemical treatment, the effluent salt from the zeolite column will be recycled to the electrorefiner. Consequently, the performance of the column is directly related to its effectiveness in removing fission product cations from the electrolyte salt. The effectiveness of the zeolite, in turn, is a function of both its ion-exchange properties and its fission product capacity. The data presented for the column tests relate the fission product concentration of the column effluent salt to the amount of salt treated by the column zeolite. The composition of the column zeolite gives the fission product loading.

The fission product concentration in the effluent salt is normalized to the concentration in the inlet salt, C/C_0 . The amount of salt that is treated is given as the number of grams of salt treated per gram of dehydrated bed zeolite. Breakthrough occurs at the point where fission products are observed in the effluent salt. As long as the aspect ratio (ratio of height to diameter) of the column is not too low, the breakthrough essentially defines the amount of salt that can be treated by the zeolite under a particular set of operating conditions. The relationship between aspect ratio and breakthrough has not yet been investigated.

Before the zeolite is contacted with waste salt, it is contacted with a LiCl-KCl eutectic salt to pre-load the zeolite with chloride. Pre-loading the zeolite with chloride effectively wets and infuses the zeolite with salt, improving the initial rate of ion exchange. The surface of the

pellets becomes coated with salt. The pre-loaded zeolite in the column prior to a test contains ~0.8 g of salt per gram of zeolite. Pre-loading also allows the chloride balance in the overall process to be maintained or nearly maintained; because the zeolite would already contain 10 to 12 chlorides per unit cell prior to contacting with waste salt, the overall amount of salt in the process is constant.

Tests were conducted at two different temperatures and at several different flow rates (Table II). Because of the slow kinetics observed in batch tests, salt flow rates were very low, between 0.5 and 3.3 cm/min. All of these tests with waste salt were conducted with 12-in. long, 1-in. diameter columns.

Table II. Summary of Ion-Exchange Column Tests

Column Test No.	Temp. (K)	Salt Flow Rate (cm/min)
1024	725	0.7
1027	725	3.3
1031	725	0.5
1108	815	1.3
1110	815	1.0

In most column tests, the fission product concentration in the effluent salt rose rapidly once breakthrough was observed. Figure 4 shows the uptake of the various fission products versus the bed volume of salt for tests run at 725 K and three different flow rates. The salt flow rate has a strong effect on fission product breakthrough. At a flow rate of 3.3 cm/min, breakthrough occurs between 0.5 and 0.9 g salt/g zeolite. Since this corresponds approximately to the initial salt-to-zeolite ratio in the pre-loaded zeolite in the column, breakthrough occurs when the salt initially has been displaced by waste salt. A larger volume of the effluent salt is effectively cleaned of fission products as the salt flow rate is reduced to 0.7 and 0.5 cm/min.

At 0.7 cm/min, breakthrough of cesium, barium, and strontium occurs nearly simultaneously after 2.2 g of salt has been treated per gram of zeolite; the rare earth cations lag slightly. At a salt flow rate of 0.5 cm/min, breakthrough of cesium and the alkaline earths occurs after 3 g of salt has been treated per gram of zeolite. Breakthrough of the rare earths is not observed until 4 g of salt has been treated. In addition, the overall rare earth concentration in the salt increases much more slowly than observed at 0.7 cm/min. The breakthrough curves for cesium and the alkaline earths begin to deviate at the point where rare earth breakthrough occurs.

Breakthrough curves for the individual ions are given in Figure 5 for a flow rate of 0.5 cm/min. The same-charge ions generally show similar behavior. The one exception appears to be strontium, whose concentration in the effluent salt does not rise rapidly at breakthrough as barium does.

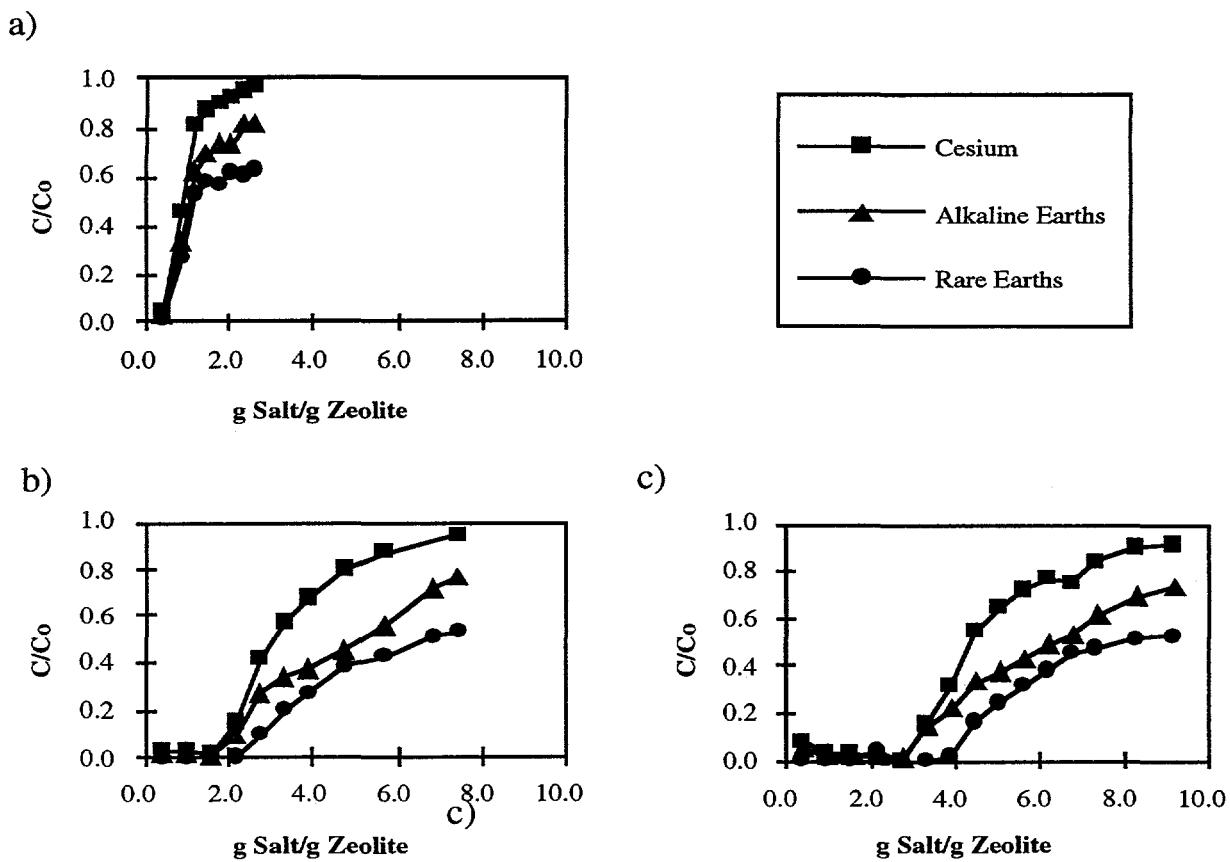


Figure 4. Breakthrough Curves for Columns Tests at 725 K and (a) 3.3 cm/min, (b) 0.7 cm/min, and (c) 0.5 cm/min

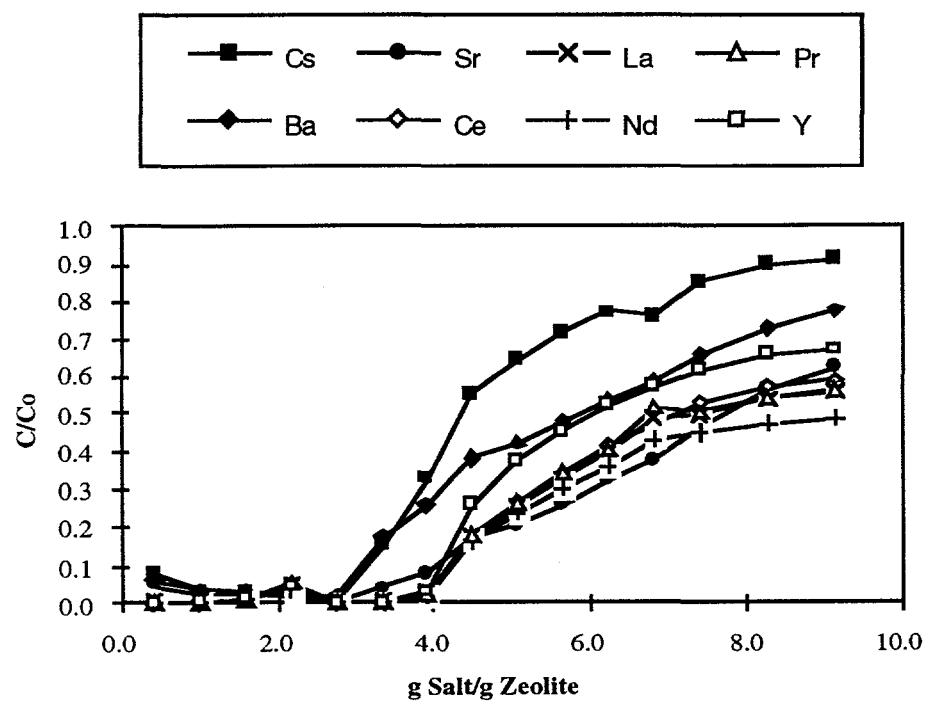


Figure 5. Breakthrough Curves for Fission Products in Test 1031 at 725 K and 0.5 cm/min

Breakthrough curves for two tests conducted at 815 K are shown in Figure 6. Test 1108 was run with salt 2, which was also used in the tests at 725 K, and Test 1110 with salt 3, which has a lower fission product concentration. Breakthrough occurs after similar amounts of salt have been treated in both tests. Breakthrough of cesium and the alkaline earths is observed first after 5 g of salt has been treated per gram of zeolite. Rare earths breakthrough after ~6 g of salt has been treated per gram of zeolite. The initial rise in concentration for cesium and the alkaline earths in the effluent salt is steeper for test 1110 with salt 3; however, the rare earths show a much more gradual increase in concentration for this salt. Based on the results for the tests run at 725 K, the small difference in flow rates would affect the location of breakthrough but not the overall shape of the curves. As can be seen in Figure 7, the curves for the individual species in Test 1108 show similar shapes for the same-valence cations, as is the case at 725 K in Figure 5. Again, the breakthrough curves show a sharp rise in the cesium and alkaline earth concentrations and a much more gradual rise in rare earth concentration.

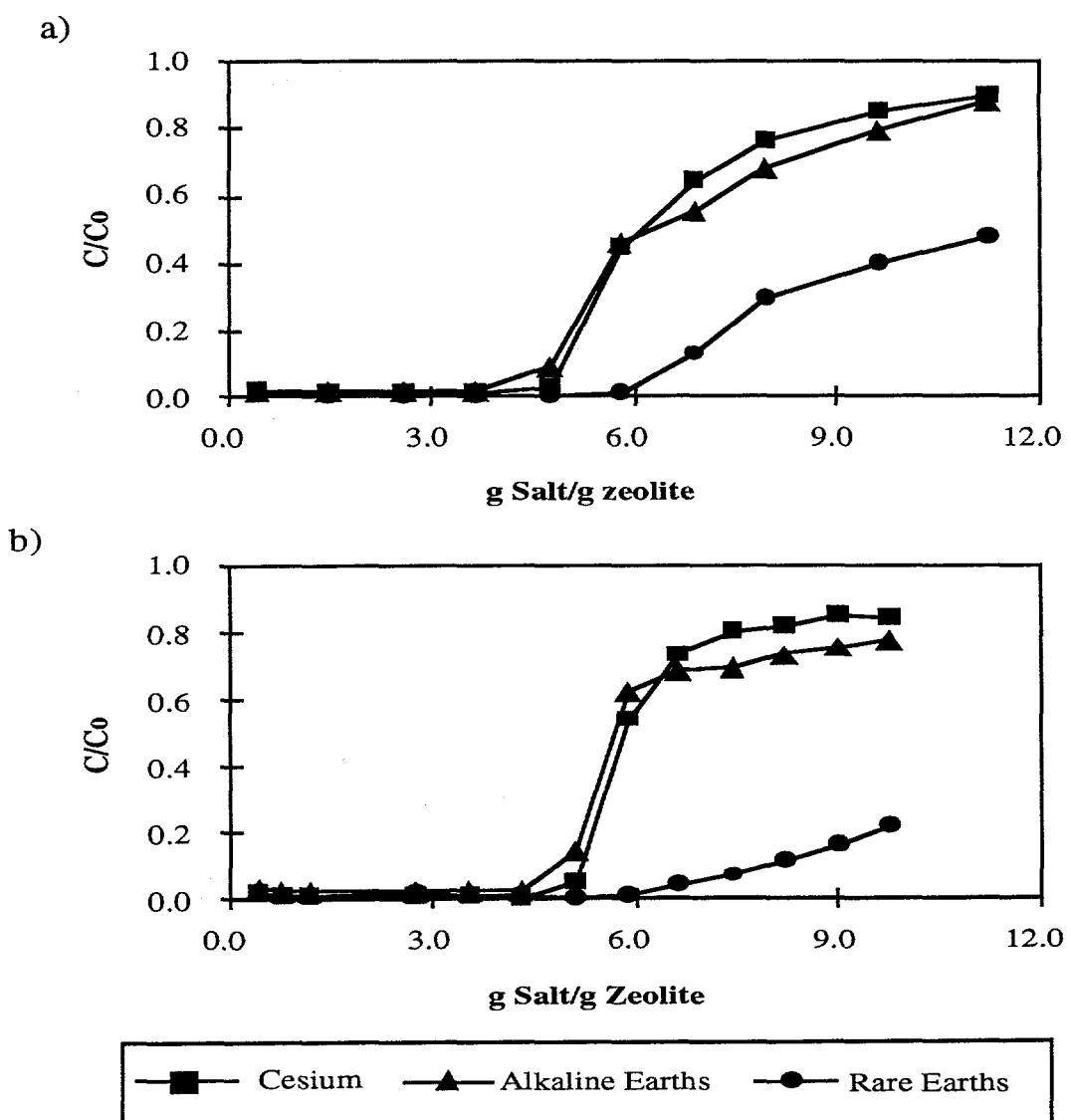


Figure 6. Breakthrough Curves for Column Tests at 815 K: (a) Test 1108, 1.3 cm/min, Salt 2, and (b) Test 1100, 1.0 cm/min, Salt 3

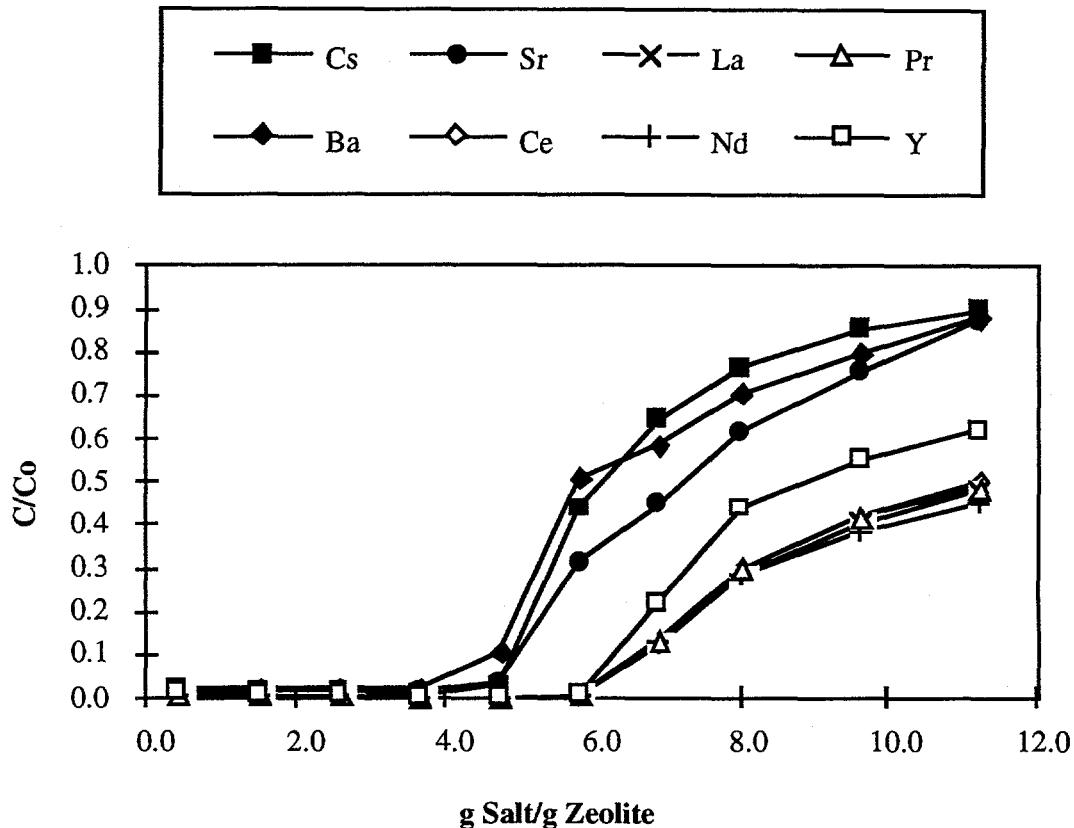


Figure 7. Breakthrough Curves for the Fission Products in Test 1108 at 815 K and 1.3 cm/min

A comparison of breakthrough data for the two test temperatures shows that the column temperature has a strong effect on performance. A much greater quantity of salt is treated at 815 K than at 725 K. At 815 K for a salt flow rate of 1.3 cm/min, breakthrough is observed after 5 g of salt is treated, compared with 2.2 g at 725 K and 0.7 cm/min. Since lower flow rates increase the amount of salt treated prior to breakthrough, the effect of temperature difference is greater than the difference in flow rates.

Table III gives the average chemical composition of the zeolite packing from the two column tests at 815 K. The zeolite samples were obtained well after breakthrough; 10 and 11 g of salt were treated per gram of zeolite for Tests 1108 and 1110, respectively. Some segregation is observed along the length of the column. Cesium and the rare earths are slightly more concentrated near the head of the column in both runs. Barium and strontium appear to be slightly more concentrated at the tail end of the column after Test 1108. Table IV illustrates the effect of salt concentration on fission product uptake in the zeolite. The rare earth concentration in the zeolite relative to that in the inlet salt is much higher for salt 3. The zeolite sorbs rare earths, and possibly strontium, more effectively when their concentrations in the salt are lower. The concentration of cesium and barium in the inlet salt does not affect relative uptakes in the zeolite.

Table III. Composition of Zeolite Pellets from Column Tests at 815 K

	Weight Percent							
	Cs	Ba	Sr	Ce	La	Nd	Pr	Y
Test 1108, 1.3 cm/min								
Head	0.54	0.14	0.087	0.60	0.22	1.1	0.19	0.028
Middle	0.51	0.15	0.10	0.52	0.17	1.1	0.18	0.029
Tail	0.48	0.16	0.12	0.36	0.11	0.89	0.13	0.027
Test 1110, 1.0 cm/min								
Head	0.30	0.073	0.045	0.41	0.14	0.85	0.14	0.022
Tail	0.25	0.073	0.047	0.31	0.098	0.71	0.11	0.023

Table IV. Composition of Zeolite Pellets from Column Tests at 815 K

	(FP per Unit Cell of Zeolite)/(Molar FP Concentration in Salt)							
	Cs	Ba	Sr	Ce	La	Nd	Pr	Y
Salt 2	3.9	3.1	4.3	6.6	7.1	4.4	5.4	4.1
Salt 3	3.7	3.2	5.0	8.6	9.1	8.2	8.4	8.3

Free Salt Determination

The salt-loaded pellets always contain a large excess of free salt that is not fully occluded by the zeolite. This salt may reside in the interstitial sites between the column pellets or within the porous binder phase. To determine the approximate location and the amount of free salt, the pellets were washed twice with deionized water (initially as whole pellets), and were prewashed a second time after the pellets had been ground. The first wash was intended to remove predominantly bulk surface salt remaining on the pellets from the interstitial sites in the column. The second wash removed salt trapped within the binder.

Washing indicated that 20 to 25 wt% of the pellets was free salt. In Table V, samples that were washed twice are designated by a W. Results are reported for the calculated chloride concentrations and the total fission products (Cs, Ba, Sr, Y, La, Ce, Nd, and Pr). No adjustment was made for the presence of binder, which comprised between 5 and 10 wt% of the pellets. The values are given in terms of ions per unit cell, assuming 12 aluminum atoms per zeolite unit cell. The overall chloride concentration was determined by summing the total cation charge per unit cell and subtracting 12 (the framework anionic charge per unit cell was assumed to be identical to the aluminum content). Since the chloride concentration in fully loaded zeolite A is approximately 11 to 13 per unit cell,(3) any value significantly above this indicates that some salt is not contained within the zeolite structure.

Table V. Composition of Zeolite Pellets after One and Two Washings

Zeolite	Cl/Unit Cell	FP*/Unit Cell
Test 1027: 725 K, 3.3 cm/min (1 washing)		
Head	21.4	2.5
Middle	20.7	2.5
Tail	24.7	2.5
Test 1027W: 725 K, 3.3 cm/min (2 washings)		
Head	11.3	2.6
Middle	11.4	2.5
Tail	11.6	2.3
Test 1031: 725 K, 0.4 cm/min (1 washing)		
Head	21.0	2.7
Middle	20.3	2.5
Tail	20.3	2.5
Test 1031W: 725 K, 0.4 cm/min (2 washings)		
Head	11.5	2.8
Middle	11.3	2.5
Tail	11.3	2.5

*FP = Cs, Sr, Ba, Y, La, Ce, Nd, and Pr.

While determining free salt by a water wash is inexact, the chloride levels calculated for the two zeolite samples are very consistent. After the second washing, the chloride concentration was between 11 and 12 Cl/unit cell, consistent with fully salt-loaded zeolite A. This indicates that most of the free salt was removed by washing. Weight measurements made after each washing step show that approximately half of the free chloride was removed in the initial washing, consistent with a substantial fraction of the free salt residing in the binder. The fission product concentration was not significantly altered by the washing, indicating that the binder contained relatively few fission products. This result suggests that ion exchange occurs between the salt in the binder and the zeolite phase in the column once salt flow has stopped.

Removal of Surface Salt from Column Zeolite

Zeolite from the column tests contains up to 25 wt% surface salt, in addition to salt that is occluded within the zeolite structure. As discussed above, the surface salt is located within the pores of the binder and on the external surface of the pellets. A fraction of the latter free salt may be removed by applying gas pressure to a column of salt-loaded zeolite.

Salt-removal tests were run with several test columns to determine the effectiveness of the pressure flow apparatus (Figure 2) in removing free salt. In these tests, the pressure drop across the column increases as the amount of surface salt on the particles increases. In tests with the columns loaded with either eutectic salt or simulated waste salt, the bed length, the temperature, and the size of the zeolite pellets were identical. Therefore, any differences in the pressure drops across the column between the two sets of tests can be attributed to the amount of surface salt left on the zeolite. In the tests with the zeolite particles used in the ion-exchange

column, an argon pressure of only 1.3 to 1.4 psi was required to produce breakthrough in all of the columns that were examined.

A much higher gas pressure was required to remove free salt from the columns that were pre-loaded with the LiCl-KCl eutectic salt than was required for the columns that were treated with the waste salt. Pressures of 10 psi were required for the pre-loaded material. These results are again consistent for all of the columns that were tested. A greater amount of salt was removed from the pre-loaded zeolite, as would be expected based on the greater pressure required.

Unlike the salt within the binder, the interstitial salt removed by application of argon pressure to the column contained a high concentration of fission products. Table VI shows compositions for two salts collected after application of argon pressure to the columns. The compositions of these salts are very similar to that of the inlet salt (salt 2).

Table VI. Composition of Salt Removed By Argon Pressure

	Inlet Salt	Test 1103	Test 1108
Cs	17.84	15.1	12.8
Ba	6.9	5.8	4.9
Sr	2.2	1.9	1.6
Ce	11.3	9.6	8.1
La	3.5	3.0	2.5
Nd	23	19.5	16.4
Pr	4.7	4.0	3.4
Y	0.6	0.5	0.4
Na	0.001	0.0	0.0
Li	7	5.9	5.0
K	26.8	22.7	19.2

Discussion

Two options exist for treating electrorefiner salt using zeolites: (1) occluding the waste salt into zeolite in a hot blending operation with no ion-exchange step or (2) concentrating the fission products in the zeolite in an ion-exchange column. For salt 2 given in Table I, the fission product weight fraction is 7.0%. Hot blending this salt with zeolite A to 10 chloride ions per unit cell yields a fission product concentration of 1.75 wt% in the zeolite.

An ion exchange system would utilize a series of columns in which the initial column in contact with the salt becomes highly loaded with fission products quickly. Once saturated, this column is removed and processed. In such an operation, a column yielding salt-loaded zeolite A at a fission product loading of 15 wt%, with an additional 25 wt% free salt at the initial composition, generates a final zeolite product containing approximately 8 wt% fission products at a chloride loading of 10 per unit cell. The chloride level is lowered by a hot blending step, which is used to fully occlude the free salt remaining in the column.

Although the salt occluded in the zeolite has a significantly higher fission product concentration than the inlet salt, if the effluent salt is to be recycled to the electrorefiner, the

column must remove a significant fraction of the fission products. This indicates that a column operating at 815 K or higher would allow a greater amount of salt to be treated. Operating at temperatures much above 825 K for prolonged periods is not desirable, however, because the zeolite structure begins to show significant degradation at temperatures above 875 K.(8) Therefore, any improvements in the column performance must be obtained by reducing the salt flow rate, reducing the particle size, changing the binder composition, or possibly adjusting the salt composition. In scoping experiments, zeolite A was found to be the most effective molecular sieve for sorbing and retaining all of the fission products present in the waste salt.(3)

Based on the tests conducted at 815 K described above, changes in salt composition would have a small effect on the point at which breakthrough is observed, even though the zeolite sorbs a greater fraction of the fission products in the salt. Lowering the salt flow rate does improve ion exchange, but backmixing may begin to affect column performance if flow rates are reduced further. The binder does appear to play a role in ion exchange in the zeolite column. Reducing the pellet size or adjusting the binder composition may improve contact between the zeolite and the bulk salt, which should improve the ion-exchange properties of the column packing.

Conclusion

The initial tests with the zeolite ion-exchange column show that zeolite A is effective in removing fission products from molten salt, particularly at temperatures above 800 K. Low flow rates also improve the extent of ion exchange, as would be expected based on kinetics measurements in batch tests. Surface salt contributes greatly to the volume of waste that is produced by the exchange process but can be reduced by forcing flow of salt by application of argon pressure. Although conditions have not been optimized, these initial tests indicate that an ion exchange column may be effective in reducing the amount of waste that is generated by the electrochemical treatment of spent nuclear fuel.

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