

## INVESTIGATION OF ELECTROKINETIC DECONTAMINATION OF CONCRETE

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## ABSTRACT

Experiments have been conducted to investigate the capabilities of electrokinetic decontamination of concrete. Batch equilibration studies have determined that the loading of cesium and strontium on concrete may be decreased using electrolyte solutions containing competing cations, while solubilization of uranium and cobalt, that precipitate at high pH, will require lixiviants containing complexing agents. Dynamic electrokinetic experiments showed greater mobility of cesium than strontium, while some positive results were obtained for the transport of cobalt through concrete using EDTA and for uranium using carbonate.

## INTRODUCTION

Contamination of concrete by radionuclides and metals is a widespread problem throughout the nuclear power industry and the Department of Energy (DOE). Electrokinetic decontamination has been considered for application to this

problem. This technology involves contacting a lixiviant solution with contaminated concrete in combination with an applied electric field. The contaminants mobilized from a bound state in the concrete pores by the lixiviant are transported to the surface by the electric field. The technology is attractive due to several possible advantages: 1) possibility for treatment of contamination existing deep in concrete or in cracks, 2) decontamination without destruction and removal of concrete, 3) significant waste volume reduction, and 4) lower cost.

Electrokinetic decontamination of concrete has many similarities with the widely studied and applied electrokinetic remediation of soils. Both techniques are similar in that each employs oppositely charged electrodes to effect mobilization of contaminants from the medium. In addition, the main transport mechanisms are the same. These transport mechanisms include electromigration, electroosmosis, electrophoresis, sorption, and diffusion. Electrophoresis, electromigration, and electroosmosis involve motion in response to a potential gradient: electrophoresis refers to the motion of charged particles, while the term electromigration is used for the transport of ions, and electroosmosis is movement of liquid relative to a stationary surface such as a capillary pore. Electrophoresis is not expected to be a dominant transport mechanism in most applications of electrokinetics; however, this mechanism could be significant under circumstances in which colloidal precipitates are formed. Electroosmosis is a particularly valuable mechanism for the treatment of tight soils for which the permeability to pressure-driven flow is low; this has been employed widely for dewatering of soils (1) as well as for removal of soluble organic contamination (2). The zeta potential of solid surfaces, a determining factor in the magnitude and direction of electroosmotic flow, is a sensitive function of solution chemistry, including pH and type and concentration of ionic species. Electroosmosis has thus been found to be variable in some applications to soils (e.g., 3). Ionic species may be readily removed by electromigration; this is the dominant mechanism of transport in electrokinetic remediation of metals contamination (4,5).

Several characteristics of concrete differentiate it from soils in general,

including lower permeability, high pH and buffering capacity, higher ionic strength of pore water, usually more complicated chemistry, and greater rigidity. Therefore, electrokinetic decontamination may not necessarily be applied in the same manner or with the same success to concrete as to soils. For instance, higher ionic strength is likely to reduce the magnitude of electroosmotic flow, high pH will affect the mobility and speciation of contaminants, and the rigidity of concrete will limit flexibility in placement of electrodes and electrolyte reservoirs.

Electrokinetics was used as early as the mid-1970's for the removal of chloride ions from concrete structures to decrease corrosion (6). In this application, metal reinforcements within the concrete are connected to the negative lead of a power supply to act as a cathode and an electrode at the surface is connected to the positive lead to act as an anode. Chloride ions within the concrete are transported to the surface by electromigration and either converted to molecular chlorine at the anode or captured by sorbent material. The technique was reported to have successfully reduced chloride concentrations such that active corrosion of the rebar had not resumed in the three months following treatment, with no adverse effect upon the concrete.

Aside from the removal of chloride ions, the application of electrokinetics to in situ decontamination of concrete has been limited to scoping studies within DOE (7,8,9). That work has been aimed mainly at removal of uranium contamination and has included bench-scale experiments and short-term field work. Several positive results were obtained, including reductions in surface radiation readings and coincident increases in solution concentrations. However there are several deficiencies in the previous work that limit the conclusions that can be derived, including: a lack of or inconsistent sampling of concrete cores, no material balances, and no control tests. Further work is necessary to determine whether electrokinetic decontamination is a viable remediation alternative for concrete.

This paper presents some results of experiments aimed at developing an understanding of the capabilities and limitations of electrokinetic decontamination of

concrete. The tests included determination of sorption isotherms of cesium, strontium, cobalt, and uranium on concrete, and experiments involving dynamic loading and desorption of concrete disks with these species by electrokinetics. The results of additional experimentation (10) and modeling (11) are presented elsewhere.

## EXPERIMENTAL

Equilibration tests were conducted to measure the sorption isotherms of cesium, cobalt, strontium, and uranium on concrete. These contaminants were chosen for testing because of their prevalence in contaminated concrete throughout the DOE complex. Equilibration tests were run in 0.016 M NaOH (simulating pH conditions of concrete pore water) or distilled water (for contaminants that precipitate at high pH) and in potential lixiviant solutions.

The tests were conducted by contacting aqueous solutions with crushed concrete. Uncontaminated structural concrete samples were collected from various destruction activities at Oak Ridge National Laboratory. This concrete was crushed and sieved, and the 500- $\mu$ m-to-1-mm fraction was used for all equilibration tests. Weighed concrete samples of approximately 1.0 g were placed in polyethylene centrifuge tubes with 10.0 mL of solution containing a single contaminant. Solution concentrations of 0, 10, 50, 100, 250, and 500 ppm of the contaminant were used, and samples were run in triplicate. Although preliminary tests indicated that equilibration was achieved within approximately 2 hours; tests were conservatively conducted by contacting the solution and concrete for at least 24 hours using a shaker. After equilibration, the tubes were centrifuged and the supernatant was drawn off. This liquid was filtered using 0.2- $\mu$ m cartridge filters to remove any particulates. The samples were diluted, if necessary for analysis, and acidified to pH 2 or lower. Sample analysis was performed using inductively coupled plasma emission spectroscopy (ICP) for cobalt, strontium, and uranium and using atomic absorption

spectroscopy (AA) for cesium.

The loaded concrete samples were subsequently contacted with 2.0-mL samples of lixiviant solutions to determine the capability of the lixiviants for dissolution of the contaminants. The procedure for the desorption tests was otherwise the same as for the loading tests.

Dynamic experiments were performed to determine the mobility of contaminants in concrete with an applied electric field. As shown schematically in Figure 1, these experiments were conducted using a concrete disk held between solutions containing energized electrodes. A known amount of contaminant was initially present in one of the solutions. The voltage was applied such that the contaminant ions would be transported into the concrete toward the solution containing the opposite electrode. The solution concentrations were measured as a function of time in order to determine the rate at which the contaminants are loaded onto the concrete and transported into the opposite solution.

The apparatus used in these experiments was fabricated from glass. Two identical cells 11.5 cm tall were constructed from 57-mm-diameter tubing. The tops of the cells were open, while the bottoms were sealed and rounded. Near the bottom of each cell, one short piece of 48-mm diameter tubing was fused at a 90-degree angle. The end of this tubing was flared to accommodate a concrete disk cut from 46-mm-diameter cores. A concrete disk from the same source as for the equilibration tests was sealed into one of the pieces of glassware using silicone caulk, then the two pieces of glassware were joined using a bead of silicone caulk and electrical tape. The assembly was allowed to sit overnight to allow the caulk to cure, then it was secured above two magnetic stirring plates. Platinum-wire electrodes were placed in each cell. The ends of these electrodes were shaped into approximately 1-inch diameter loops of 20-ga. wire held parallel and in close proximity to the faces of the concrete disk. Teflon-coated magnetic stir bars were placed in each reservoir, the assembly was checked for leaks, and depth scales on each reservoir were calibrated by adding known volumes of water in each side. Prior to conducting several of the dynamic

tests, distilled water was placed in each reservoir, and a large potential difference (several kV) was applied across the concrete. No large electroosmotic flow was noted, with an estimated detection level in these short tests of 5 mL/h. Known volumes of solutions of known concentration were added to each reservoir, and electrical power was applied using a Sigma Aldrich PS 4010-1 power supply. Typical limit settings for an experiment were 500 V, 100 mA, and 100 W. The applied voltage, current, and pH and level of each solution were recorded periodically, and samples of each solution were taken for analysis as each experiment progressed.

## RESULTS

### Equilibration Tests

Results of cesium loading tests are shown in Figure 2. This and the following two figures figure include data from loading tests as solid points and those from desorption tests as hollow points. The loading behavior of cesium on concrete in 0.016 M NaOH is well represented by a Freundlich isotherm,

$$q = 4.07c^{0.668} \quad (1)$$

where  $q$  is the loading of cesium on concrete in milligrams per gram of concrete, and  $c$  is the concentration of cesium in solution in mg/mL. Cesium loading is seen to decrease with increasing sodium concentration in the lixiviant solution. The cesium loading at high sodium concentration is nearly linear, a Freundlich fit of the combined data for 1 M sodium carbonate and 1 M sodium chloride is,

$$q = 2.27c^{1.04} \quad (2)$$

Additional tests indicated that EDTA at low concentration (0.01 M) is relatively ineffective at dissolution of cesium.



Several series of equilibration tests were conducted with strontium, each involving loading from 0.016 M NaOH with subsequent desorption using 0.01 M EDTA, 0.016 M NaOH, 1 M NaCl, or 1 M CaCl<sub>2</sub>. Results from these tests are shown in Figure 3. The loading from 0.016 M NaOH in one set of tests was well described by a Freundlich isotherm,

$$q = 7.77 c^{0.733} \quad (3)$$

which indicates that the loading of strontium is greater than that of cesium. Equilibration data for 0.01 M EDTA was fit well by a linear isotherm with a distribution coefficient of 29.7 mL/g, indicating that this solution was not very effective at dissolving the strontium. This is not surprising, since Sr<sup>2+</sup> forms a weaker complex with EDTA than many other cations, including Ca<sup>2+</sup> (12). A second loading test on a different concrete sample yielded a Freundlich isotherm of

$$q = 2.99 c^{0.423} \quad (4)$$

The fact that tests with two different batches of concrete resulted in two different isotherms indicates that equilibration data will likely be specific to particular concrete samples. The concrete from this test was desorbed by contact with one of three solutions: 0.016 M NaOH, 1 M NaCl, or 1 M CaCl<sub>2</sub>. The isotherm for desorption with 0.016 M NaOH was

$$q = 5.15 c^{0.339} \quad (5)$$

As seen in Figure 4, the equilibrium loading described by this isotherm is greater than that for the initial loading from 0.016 M NaOH. This may be due to a lesser amount of calcium present to compete for sites with strontium during desorption. Competitive sorption is further illustrated by data from desorption with the concentrated solutions. The isotherm for 1 M NaCl was

$$q = 2.68 c^{0.535} \quad (6)$$

resulting in lesser loading of strontium than for the other solutions, and that for 1 M  $\text{CaCl}_2$  was

$$q = 1.15 c^{0.421} \quad (7)$$

resulting in even less loading of strontium on the concrete.

Precipitate was formed immediately upon contact of cobalt solutions in distilled water with concrete. The precipitate was initially a blue-green color and it changed gradually to brown. The production of precipitate is to be expected, since cobalt is highly insoluble at high pH. No cobalt was measured in solution after loading or after equilibration with 0.01 M EDTA.

Yellow precipitate was formed immediately upon contact of the uranyl nitrate solutions in distilled water with the concrete. The volume of precipitate formed was similar to that for cobalt. Results from the uranium tests are shown in Figure 5. There was considerable scatter in the data, possibly due to suspension of precipitate when taking samples. The loading data for distilled water showed that almost none of the uranium stayed in solution; the distribution coefficient for a linear isotherm of this data is 9410 mL/g. Uranium was only slightly soluble in 0.2 M carbonate, resulting in a linear fit with a distribution coefficient of 26.4 mL/g. By increasing the sodium carbonate concentration to 1 M, a significant increase in uranium solubility was achieved; however, an increased amount of precipitate was formed. A 0.01 M EDTA lixiviant was found to be as effective as the 1 M sodium carbonate solution in solubilizing uranium. A Freundlich fit of the data for 1 M sodium carbonate and 0.01 M EDTA together resulted in

$$q = 55.7 c^{1.40} \quad (8)$$

Figure 6 shows the isotherms fit to the data from the equilibration tests. This plot summarizes the relative solubility of Cs, Sr, and U in contact with concrete and the relative potential of various solutions as lixiviants for concrete decontamination. Of the species tested, Cs is the least tightly bound by concrete. Strontium is more

tightly bound by concrete than cesium, but it still can exist in relatively high concentration in solution. These relatively soluble contaminants can be preferentially desorbed by other cations such as sodium or calcium. Uranium and cobalt are much less soluble. Lixiviants containing complexants such as carbonate or EDTA anions increase the amount of uranium in solution; however, the isotherms for uranium with these complexants are still very steep. Thus, the relative capability for removal of the contaminants tested by chemical means (either by chemical extraction or by electrokinetics) appears to be  $Cs > Sr \gg U, Co$ .

### Dynamic Tests

Two dynamic experiments were conducted with cesium using a 0.95-cm thick concrete disk (mass: 34g). The initial anolyte (i.e., the solution containing the positive electrode) in each test was 200 mL of solution containing 1000 ppm of cesium in 0.016 M NaOH, while 218 mL of 0.016 M NaOH was the initial catholyte (i.e., the solution containing the negative electrode). The solutions were stirred throughout the tests. In the first test, the current was held constant at 100 mA for 7 hours, reduced to 50 mA for 16.8 hours, then held at 100 mA for an additional 6.2 hours. In the second test, the current was maintained at 100 mA for 10 hours. Between tests, an attempt to drive any remaining cesium off the concrete was made by operating the system for 30 minutes with 1 M NaCl solution as the anolyte and distilled water as the catholyte.

During the first test, the voltage required for 100 mA was initially 91V, dropping to approximately 55 V after 1 hour. The conductivity was then steady through 6 hours of operation. Upon resuming operation at 100 mA, the voltage required had risen to a nearly constant 107 V. The conductivity variation during the second test was similar to that of the first test for the first 6 hours, then the conductivity steadily decreased during the next 4 hours to the final conditions of 100 mA at 119 V. The solution volumes decreased significantly during the experiments;

the anolyte volume change was consistent with the removal of 1-mL samples, while the catholyte volume change (after adjustment for samples) amounted to nearly 2 mL/h in the first test and 0.7 mL/h in the second. Accounting for the losses due to electrolysis and assuming equal evaporation in each cell, electroosmotic flow may have been as large as 0.8 mL/h in the first test and 0.2 mL/h in the second. This electroosmotic flow would have been toward the positive electrode, in the opposite direction of the electromigration of cesium ions.

The results from both dynamic tests with cesium are presented in Figure 6. This figure shows the amount of cesium present in the anolyte, in the catholyte, and on the concrete (taken to be the difference between the total amount of cesium present and that in the two solutions) as a fraction of the total amount of cesium present in the system. The experiment is seen to be qualitatively repeatable. The amount of cesium in the anolyte decreased rapidly, while the concentration in the catholyte increased with time, signaling a transport of cesium through the concrete. By the end of the first test, the cesium had essentially been quantitatively transferred from the anolyte to the catholyte, with a material balance of within 1 percent. The amount of cesium loaded on the concrete varied with time, reaching a maximum of 19.4% of the total cesium present in the first test. There was a greater peak loading of cesium on the concrete (25.8%) during the second test that occurred at approximately the same time during the experiment. The difference in loading is likely due to changes in chemistry on the surfaces of the concrete pores as the ions were driven through the concrete over the course of the first test. One interesting potential use for this type of experiment is controlled loading of concrete samples. The test could have been stopped at the point of maximum loading, resulting in a contaminated concrete disk loaded with a known quantity of cesium. In addition, because the cesium was fully driven from the concrete with further operation, it is shown that electrokinetics may be effectively used for removal of cesium contamination.

One dynamic loading/desorption test was conducted with strontium. In this test, strontium at 1000 ppm in 0.016 M NaOH was used as the anolyte, while 0.016

M NaOH was used as the catholyte. A new concrete disk, 8 mm thick, was used for this test. The current was held constant at 100 mA for 8 hours, reduced to 50 mA for 15.2 hour, then held at 100 mA for an additional 3.8 hours. A small amount of white precipitate was noted in the anolyte prior to operation, and additional precipitate formation was noted during the experiment. It is possible that this precipitate was strontium carbonate. The change in solution levels was similar to the cesium tests, with a possible electroosmotic flow of 0 to 0.5 mL/h toward the anolyte (opposite the electromigration of strontium).

The results of the strontium dynamic test are shown in Figure 7. These results show that loss of strontium from the anolyte was approximately half as fast as that for cesium. Because the samples were taken from a well-agitated solution and acidified without filtration, it is unlikely that a significant portion of the strontium loss from the anolyte is due to precipitate formation. The transfer of strontium through the concrete to the catholyte was significantly slower than that for cesium, and strontium in the catholyte amounted to only 17% of the total strontium present at the end of the test. Factors that may have contributed to slower transport of strontium are the formation of larger, monovalent  $\text{Sr}(\text{OH})^+$  ions, and the formation of insoluble carbonate. At the end of the test, 79% of the strontium was not present in either solution, and therefore assumed to be on the concrete. This value appears to be near the peak of the loading curve. The greater uptake of strontium than cesium illustrates the effect of the difference in the loading isotherms. It is of interest to conduct additional dynamic experiments with strontium. Extended operation would be useful to determine if strontium can be fully transferred across the concrete, and how long the transfer would take. Another recommended experiment would start with a run similar to this one for loading of the concrete, followed by a change to an anolyte of high ionic strength, such as 1 M  $\text{CaCl}_2$  to see if competitive sorption/ion exchange could be exploited to force the strontium off the concrete more completely and at a faster rate.

A series of dynamic tests were conducted with cobalt that provide some

insight into techniques that may be effective at mobilizing species that precipitate at high pH. Operation with 218 mL of 1080 ppm cobalt in distilled water as the anolyte and distilled water as the catholyte with an applied current of 100 mA resulted in a steady decrease in concentration of cobalt in the anolyte for approximately 3 hours, followed by a nearly steady concentration. A dark brown precipitate formed on the surface of the concrete exposed to the anolyte. The precipitate persisted throughout the operation, even through the pH of the anolyte was as low as 2.3 (due to the water electrolysis reaction at the anode). The near-steady cobalt concentration in the anolyte may be a balance between dissolution of cobalt from the concrete face and electromigration into the concrete (followed by precipitation). After operation for 7 hours, 63% of the cobalt had precipitated on the concrete. No cobalt was detected in the catholyte.

Following the loading of the concrete disk with cobalt, the solutions were replaced with distilled water anolyte and 0.025 M EDTA catholyte, and electrical power was applied for 7 hours at 100 mA. After 90 minutes, the anolyte was a noticeable pink color, indicating that some EDTA had been transferred through the concrete and had formed a soluble complex with cobalt from the precipitate. By the end of the test, the anolyte was a dark purple. Analysis of anolyte samples by ICP shows an increase in cobalt concentration corresponding to the color change; at run times of 2, 4, and 7 hours, the cobalt concentration was 15 ppm, 130 ppm, and 320 ppm, respectively. The final anolyte volume was 173 mL. Thus, 38% of the cobalt that had been precipitated in the loading phase was dissolved by the electrokinetic transport of EDTA through the concrete. Since the cobalt concentration had not leveled off, it is likely that significantly more cobalt could have been dissolved with further operation.

An additional dynamic test was conducted with this disk. The purple solution containing the EDTA and cobalt was retained, while 0.016 M NaOH was added to the opposite reservoir. The polarity was changed such that the Co-EDTA solution was the catholyte and the NaOH solution was the anolyte. Because the Co-EDTA

complex is negatively charged, there was a driving force for electromigration of the complex to the anolyte. After 5 hours of operation, the initially purple catholyte had changed to a suspension of muddy-brown precipitate, while the anolyte had a slight pink tint. The pink tint became darker with time, while the precipitate persisted in the catholyte. The progress of this test was followed qualitatively using a spectrophotometer at 537 nm. After 7 hours, there was nearly no Co-EDTA complex remaining in the catholyte and a small amount of the complex was present in the anolyte. The formation of precipitate in the high-pH catholyte indicated that a portion of the Co present was not transferred by the EDTA.

Three dynamic tests were conducted with uranium. As with cobalt, these tests were conducted to see if this precipitating contaminant can be mobilized through concrete by electrokinetic means. One test was conducted using a distilled water anolyte and a catholyte consisting of 218 mL of 250 ppm U (from uranyl nitrate) in 0.5 M sodium carbonate/bicarbonate, pH adjusted to 9.8. This configuration was used such that there would be a driving force for electromigration of the uranyl tricarbonatate ion,  $\text{UO}_2(\text{CO}_3)_3^{4-}$ , through the concrete toward the anolyte. The test was run for a total of 14 hours with voltage and current limits of 500 V and 100 mA. The catholyte was initially yellow; this coloration continued throughout the test, and no color change was noted in the anolyte. The volume change for each reservoir was nearly equal, indicating that there was no electroosmotic flow. The amount of uranium in the catholyte was nearly constant, and no transport of uranium into the anolyte was measured. The most striking feature of this test is low conductivity. Initially, 367 V was required for a current of 100 mA, and the conductivity continued to decrease throughout the test to final conditions of 500 V and 11 mA. The low conductivity, even with a 1 M carbonate solution in one reservoir, is likely due to the use of carbonate as the complexing ion for uranium. Carbonate forms insoluble compounds with several cations, including calcium and magnesium that are prevalent in concrete; in addition, any carbonate transferred to the anolyte would form weakly dissociated carbonic acid. The formation of carbonate-containing precipitates within

the concrete would also cause precipitation of non-complexed uranium, and it is possible that some pores could be obstructed by precipitate.

To further investigate the use of carbonate lixiviant, a second dynamic test was conducted with the same disk as the first test. The anolyte and catholyte solutions used in this test were both 1 M carbonate/bicarbonate solution, pH adjusted to 9.64. The catholyte also contained 500 ppm U. The catholyte was initially yellow, while the anolyte was clear. Some precipitate from the first experiment remained in the catholyte reservoir. The conductivity was initially relatively low, but it rose throughout the first 5 hours of the test and remained high for the next 21 hours of operation. The precipitate initially in the catholyte appeared to dissolve after approximately 90 minutes, near the time when a sharp rise in conductivity was noted. No color change in either solution was observed, and pH values remained close to the initial value of 9.6. This test indicated that the conductivity may be adequate when carbonate solution lixiviants of high conductivity are used at both electrodes. In addition, as shown in Figure 8, the test was successful in transporting uranium slowly into the anolyte. Because of the relative success of this test it is recommended that further study be made of this system.

A third dynamic test was conducted with uranium using EDTA as the complexant rather than carbonate. The initial anolyte was tap water, while 400 ppm U in 0.025 M EDTA was used as the catholyte solution. Yellow precipitate was noted in the catholyte. Electrical power was maintained for 54 hours, and the conductivity of the system decreased throughout the test from initial conditions of 221 V and 100 mA to final conditions of 500 V and 33 mA. With the exception of a relatively large amount of evaporation, there was no noticeable difference was noted in either solution, and no uranium was detected in the anolyte.



## CONCLUSIONS

These experiments have collected valuable information regarding the capability for removal of contaminants from concrete using electrokinetics. The results indicate that contaminants may be broken down into two general types: those species that do not precipitate at high pH and those that do. The non-precipitating species, such as cesium and strontium, may be solubilized using high concentrations of competing cations. It appears to be possible to transport these contaminants through concrete by electrokinetics using electrolyte solutions as lixiviants. Solubilization of precipitating species, such as cobalt and uranium, will require complexing agents such as carbonate or EDTA. Tests that displayed the movement of EDTA and the cobalt-EDTA complex through concrete indicate that decontamination of concrete containing precipitating species may be possible through the use of a proper complexant. In addition, some success was achieved in transport of uranium using a carbonate lixiviant. However, at this point, the decontamination of concrete containing precipitating species using EK appears to be slow and troublesome. Further testing is needed to devise lixiviant solutions that are successful at solubilizing these species and do not interact greatly with concrete. In addition, demonstrations are necessary to provide information on achievable cleanup levels and required treatment times.

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## FIGURES

1. Schematic of dynamic electrokinetic experiment.
2. Results of cesium equilibration tests.
3. Results of strontium equilibration tests.
4. Results of uranium equilibration tests.
5. Summary of equilibration test results.
6. Results of cesium dynamic tests, indicating the variation with time of the distribution of cesium between the anolyte, catholyte, and concrete.
7. Results of strontium dynamic tests, indicating the variation with time of the distribution of strontium between the anolyte, catholyte, and concrete.
8. Variation of uranium concentration in anolyte and catholyte during dynamic test using 1 M carbonate solutions as initial anolyte and catholyte.

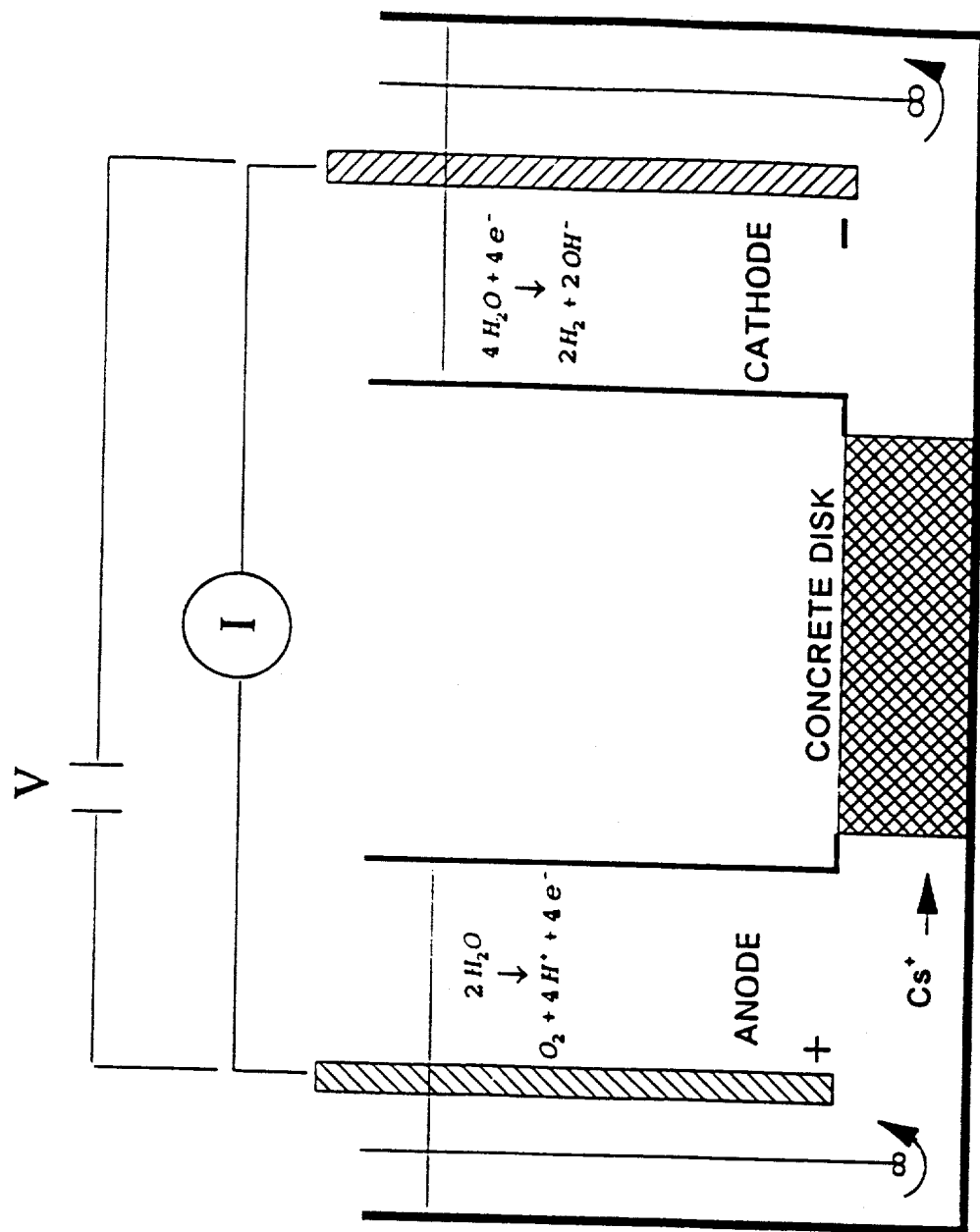
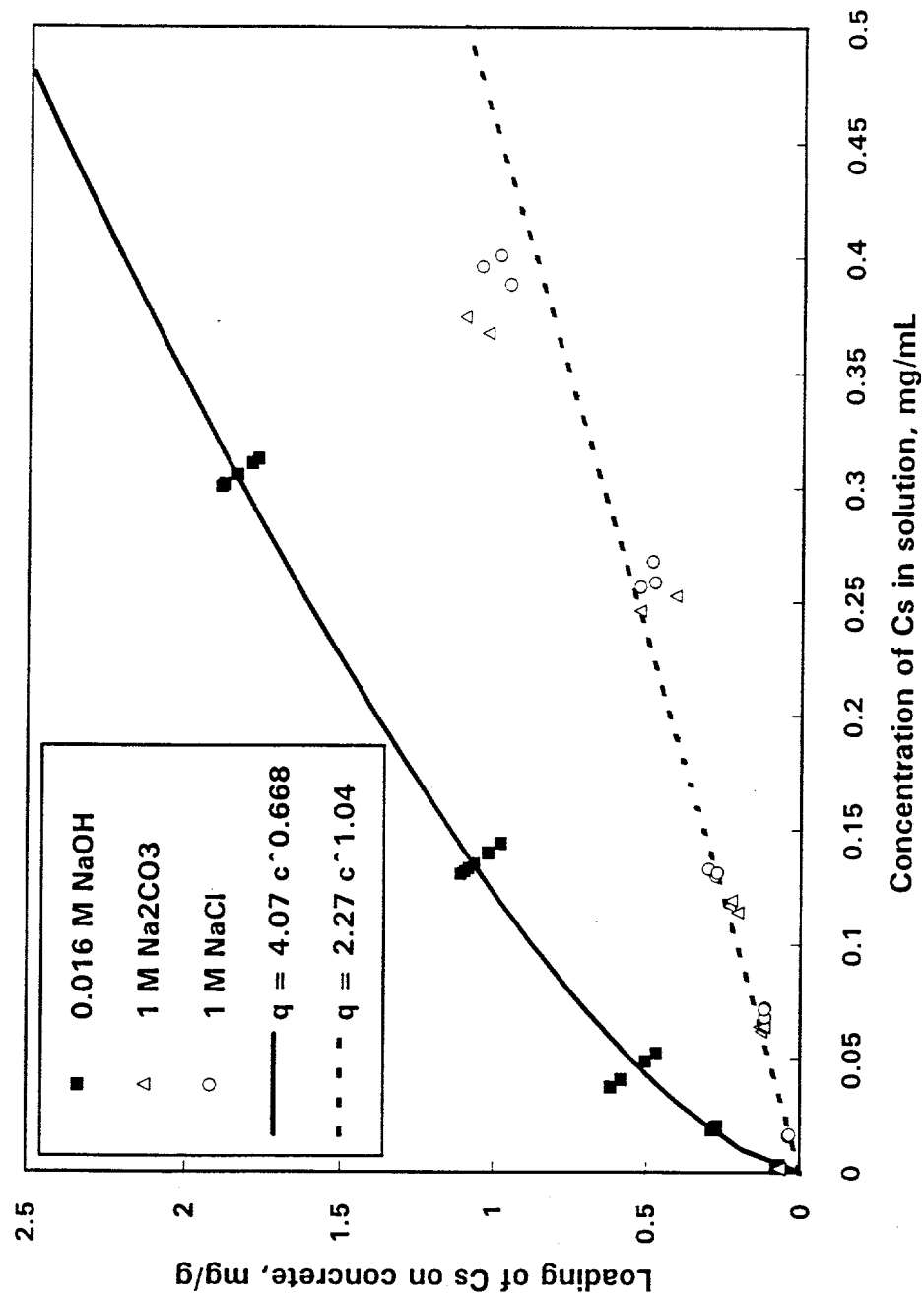
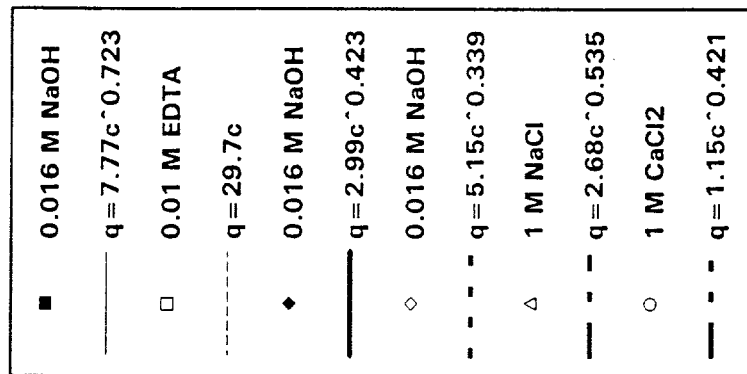
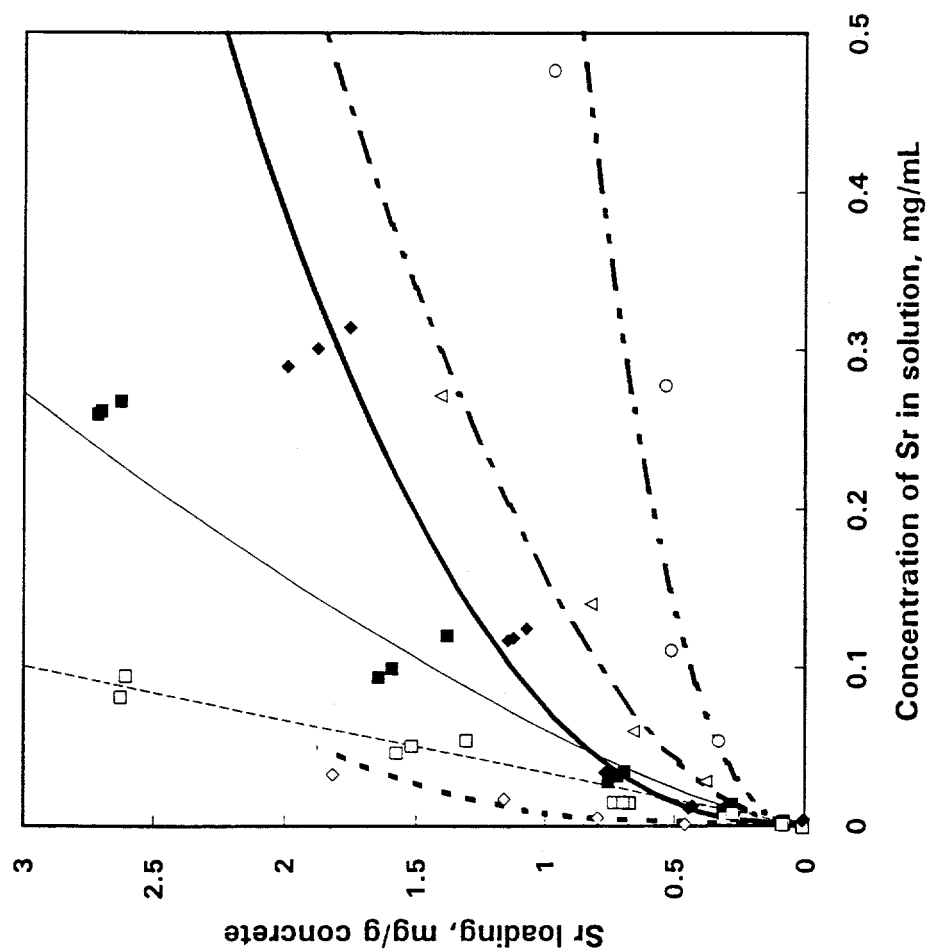
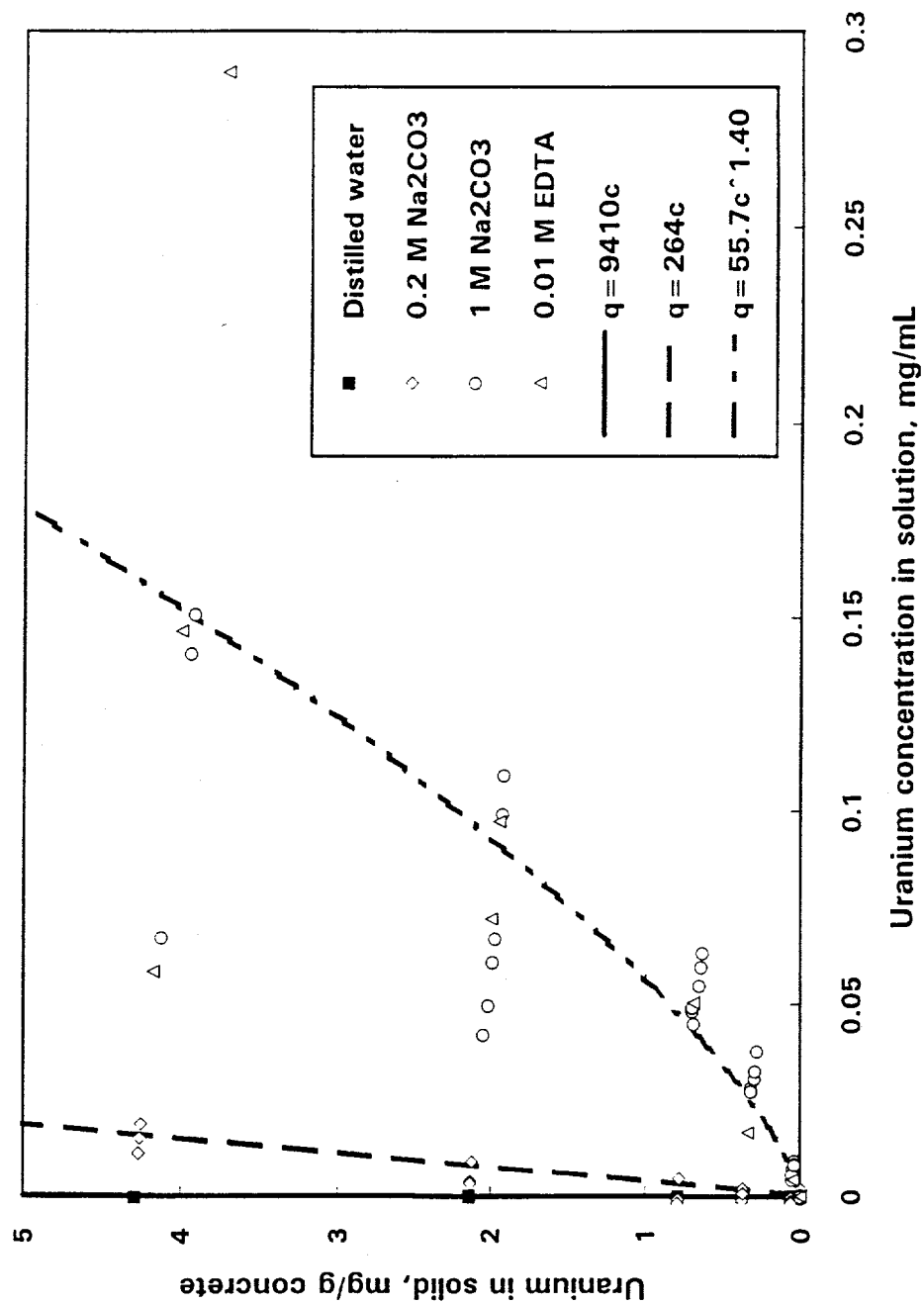


Fig. 1







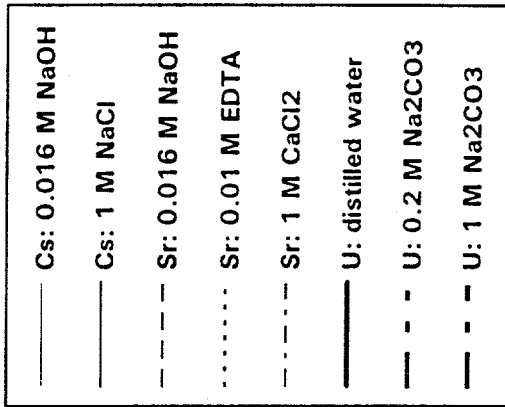
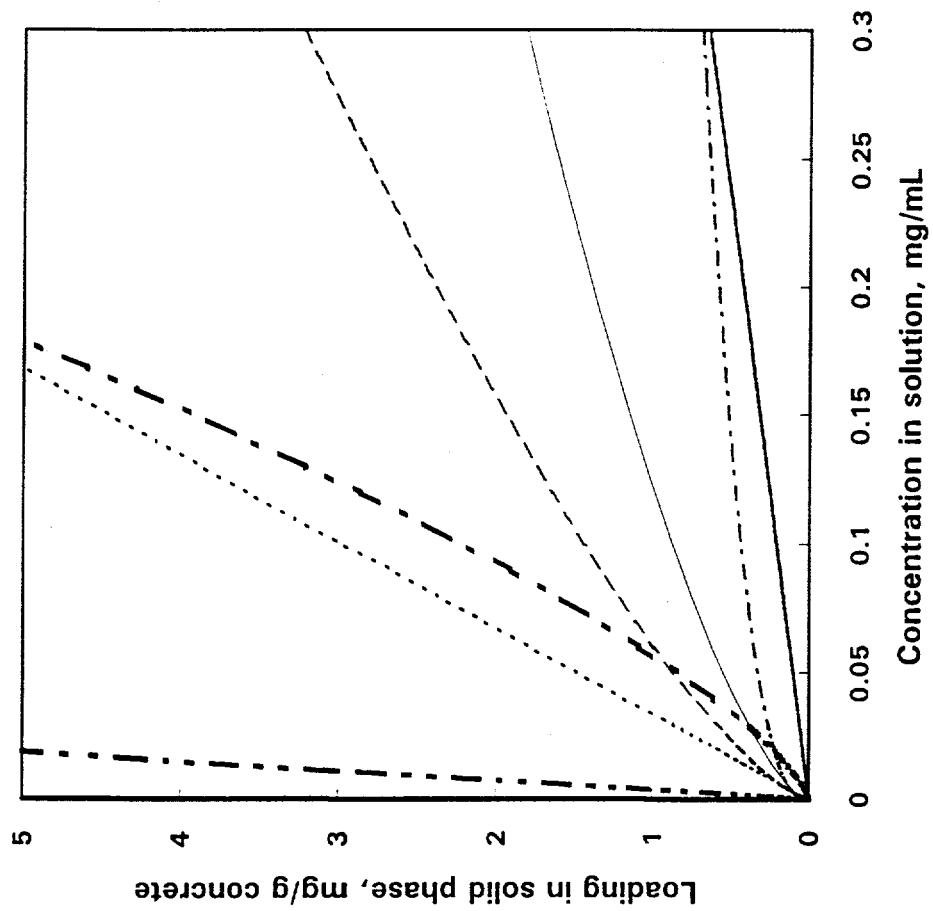


Fig. 5



