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ELECTROKINETICS

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Transport of Radioactive Ions in Soil by Electrokinetics

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Abstract

An electrokinetic approach is being evaluated for *in situ* soil remediation at the Hanford Site in Richland, Washington. This approach uses an applied electric field to induce transport of both radioactive and hazardous waste ions in soil. The work discussed in this paper involves the development of a new method to monitor the movement of the radioactive ions within the soil during the electrokinetic process. A closed cell and a gamma counter were used to provide *in situ* measurements of ^{137}Cs and ^{60}Co movement in Hanford soil. Preliminary results show that for an applied potential of 200 V over approximately 200 hr, ^{137}Cs and ^{60}Co were transported a distance of 4 to 5 in. The monitoring technique demonstrated the feasibility of using electrokinetics for soil separation applications.

Introduction

Soil contaminated with heavy metals and/or radionuclides is a widespread problem at many government and industrial sites, particularly regarding the movement of the toxic metals by diffusion or leaching into underground water tables. Because these sites are subject to regulatory cleanup and disposal requirements, more effective remediation methods are being developed. Electrokinetics has emerged as a promising *in situ* technique for removing heavy-metal ions and/or radionuclides from soil.

Electrokinetic remediation of soil, which can be described physically as the movement of charged or neutral species in an electrolyte solution within the porous soil matrix under the influence of an electric field,^(1,2) has been studied by numerous researchers for several years.⁽¹⁻²⁴⁾ The technique relies on the fact that many solids undergo surface ionization in the presence of water. The charge-bearing particles and dissolved ions are then attracted to either the positive or negative electrode as an electric field is applied. Positively charged species and water move toward the cathode (negative electrode), and negatively charged species move toward the anode (positive electrode). The primary advantage of the technique is that charged contaminants can be transported or concentrated *in situ*, without soil excavation. This inherently reduces the secondary waste and the cost associated with decommissioning or decontamination of excavation equipment.

This paper examines a new experimental approach that monitors the movement of radioactive ions within soil during the electrokinetic process. The objective of this work is to demonstrate the feasibility of transporting unbound Cs and Co in saturated Hanford soil by using an applied electric field (electrokinetics). The experimental approach allows for *in situ* monitoring of radioactive Cs and Co during the electrokinetic process. In addition to a description of the new experimental apparatus and preliminary results of the study, this paper provides an overview of the mechanisms governing electrokinetic transport in porous media.

Transport Mechanisms

The transport of charged species in porous media under the influence of an electric field can be classified into distinct electrokinetic phenomena: electrophoresis (the movement of charged particles), electroosmosis (the movement of water), and electromigration (the movement of ions).^(1,2) Figure 1 shows a schematic of these electrokinetic phenomena. For the work discussed in this paper, the soil was well-packed, which prevented the movement of large solid particles; therefore, only the latter two mechanisms are relevant. It should be noted that in addition to the electrokinetic phenomena described above, electrolysis reactions take place at the cathode and anode. These reactions include the reduction and oxidation of water and other ionic species near the electrode surface.

The fundamental mathematical expressions which describe transport in porous media via electrokinetic phenomena are provided below. These expressions are discussed widely in the open literature and are used to predict the transport of contaminants in soils. This information is provided to demonstrate the difficulty in predicting contaminant transport without a thorough characterization of the soil chemistry and colloidal properties.

The molar flux, J_j , of an ionic species j ⁽²⁰⁾ is given by

$$\mathbf{J}_j = -D_j \nabla c_j - z_j F c_j u_j \nabla \Phi + c_j \mathbf{v} \quad [1]$$

where D_j is an effective ionic diffusion coefficient, c_j is the ionic concentration, z_j is the valence, F is Faraday's constant, u_j is an effective ionic mobility, Φ is the applied electric potential, and \mathbf{v} is the velocity of the bulk solvent fluid. The first term in Equation (1) is a contribution from diffusion, the second migration, and the third convection. Equation (1) neglects all coupling effects between driving forces and thermal gradients. For this application, the velocity term in Equation (1) is given by:

$$\mathbf{v} = -\kappa_h \nabla h - \kappa_e \nabla \Phi \quad [2]$$

where κ_h is the hydraulic conductivity of the soil, h is the hydraulic head, and κ_e is the electroosmotic permeability. The first term in Equation (2) is Darcy's law, while the second term represents contributions from electroosmosis. The hydraulic conductivity and the electroosmotic permeability are empirical parameters with complex relationships to fundamental variables. The hydraulic conductivity depends primarily on the pore and particle size distribution and on the tortuosity path of the flowing fluid. The electroosmotic permeability is a function of variables such as the soil porosity and the surface charge or zeta potential of the soil matrix particles. The Helmholtz-Smoluchowski equation can be used to relate κ_e to these parameters⁽¹⁾ by

$$\kappa_e = \frac{\epsilon \zeta \theta}{\mu} \quad [3]$$

where ϵ is the solution permittivity, ζ is the zeta potential, θ is the porosity of the soil matrix, and μ is the solution viscosity.

The current, I , measured during electrokinetic remediation is created by the movement of ions and is related to the ionic flux by

$$I = F \sum z_j J_j \quad [4]$$

When Equation (4) is combined with Equation (1), the current can be expressed as

$$I = F \sum -z_j D_j \nabla c_j - \sigma \nabla \Phi \quad [5]$$

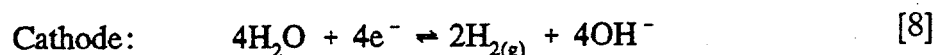
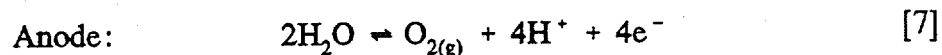
where

$$\sigma = F \sum z_j c_j u_j \quad [6]$$

The quantity σ is defined as an effective electrical conductivity of free fluid in the soil pores and is a function of the ionic mobility of all species in solution. Equation (5) implicitly assumes that electroneutrality holds at all points in the fluid within the soil pores. This assumption is difficult to validate because of the complex heterogeneity of the soil

composition and chemistry, which vary significantly as a function of time during the electrokinetic process. Characterization of transport of using Equations (1) through (6) has obtained adequate predictions for simple, homogeneous soil matrices. However, for field demonstrations, soils are not homogeneous and predicting the transport of contaminants using these expressions is difficult.

Also, as mentioned earlier, Faradaic electrochemical reactions occur at both the anode and cathode. Of primary importance are the oxidation and reduction of water. These reactions are listed below:



Note that hydrogen ions are produced at the anode and hydroxyl ions are generated at the cathode. The ionic mobilities of these ions are typically 5 to 10 times higher than other cations or anions, as is shown in Table 1. The high mobilities of H^+ and OH^- ions create significant pH gradients within the soil matrix. An acid front will develop from the anode and a base front from the cathode. At a point between the electrodes, the fronts will meet and neutralize the soil pH. This point is typically close to the cathode because electroosmosis transports water from the anode to the cathode and, therefore, aids in the transport of H^+ and inhibits the transport of OH^- . The majority of the potential drop across the soil occurs at the neutralization point because of the low ionic strength of the solution within the soil pores.

The pH fronts within the soil matrix create many heterogeneous conditions, especially, variations in the soil solubility chemistry and zeta potential. The change in solubility of soil salts and oxides can cause unwanted precipitation. Also, it can reduce the efficiency of transporting an ionic contaminant because of increased salt dissolution. The zeta potential of soil particles is a strong function of pH.^(1,2) At low pH the zeta potential is positive, while at high pH the surface charge is negative. It is clear from Equations (1) and (3) that zeta potential variations will strongly affect the ionic and electroosmotic transport.

The above discussion was provided to demonstrate the complicated chemistry and heterogeneity within a soil matrix during electrokinetic remediation. Because of this unpredictable behavior, it is advantageous to develop experimental techniques to probe the soil matrix *in situ* for both chemical and physical properties. Here, a nondestructive experimental approach is introduced to monitor the transport of radioactive species in real-time during electrokinetic treatment.

Experimental Apparatus

The experimental apparatus was designed to allow real-time measurements of electrokinetic transport of radioactive species in porous media. The system consists of an electrokinetic cell, a NaI scintillation detector, a multichannel analyzer, and a high-voltage power supply. A schematic of the equipment is shown in Figure 2.

The electrokinetic cell, which was constructed from Plexiglas, is rectangular in shape with the dimensions 2 in. x 6 in. x 10 in. For the experiments, the soil was sandwiched between two flat-plate, stainless steel electrodes. A threaded rod was welded to each electrode so that the volume of soil between the electrodes could be varied. The rods were sealed at the cell wall with Kynar O-rings. Two ports on the anode and cathode sides were available to vent gaseous oxygen and hydrogen generated by the electrolysis of water.

The cell was placed above a NaI scintillation counter with a 2-in.-thick lead collimator having a 3/8-in.-diameter circular aperture. The cell was positioned as close to the shield as possible to minimize radioactive decay due to detector-source separation. A 3/8-in.-thick aluminum plate was placed between the collimator and the cell as a platform to provide support for the cell as it was manually moved on top of the detector. The cell was moved in a grid pattern while the NaI detector remained stationary to obtain a two-dimensional map of the radioactive species in the soil.

A Tracer Northern, Model TN-2700, multichannel analyzer was used to measure the gamma radiation spectrum from the NaI detector. The analyzer used an auxiliary high voltage supply to power the NaI detector. The detector efficiency was optimized for Cs and Co gamma rays, and good counting statistics were obtained with an integration time of 180 sec. The optimal detector operating voltage was at +1080 V. Although a full energy spectrum was measured, only the primary emission lines of Cs and Co were recorded. The radioactivity was measured as peak areas with centerlines located at 662 keV, 1173 keV, and 1333 keV. The 662 keV emission line was produced by ^{137}Cs , while the other lines were emitted by ^{60}Co .

The potential necessary to enable electrokinetic ionic movement was provided by a Kepco, Model HB8A, power supply. An HP, Model 3438A, multimeter was placed in series with the electrical circuit to measure the cell current. For all experiments, the power supply was operated in potentiostat mode with an applied voltage of 200 V. The corresponding cell current varied from 16 to 48 mA.

Nonradioactive soil, obtained from the Hanford Site 200 East Grout Vault, was used for the experiments. The soil was initially sieved using a U.S. mesh size of 10 to eliminate large rocks which would cause channeling within the cell. The soil was saturated with deionized water, and the moisture content was measured to be 16.40% by weight. Before the saturated soil was added to the cell, it was mixed in a plastic bag to ensure a uniform moisture content. Then, the soil was evenly distributed within the cell and packed down with a 5-lb brick to minimize void spaces.

After the soil was added to the cell, a small portion was removed and replaced with saturated soil containing a "spike" mixture of radioactive and nonradioactive components. The ion exchange capacity of the soil required that the additional nonradioactive material be added to the soil to ensure there was unbound or mobile Cs and Co ions. The volume of soil removed and replaced was approximately 5 mL. Two experiments were run: first, a spike of ^{137}Cs and CsNO_3 was added to the center of the rectangular cell and the movement was monitored for approximately 10 days; and second, three spikes containing ^{137}Cs , CsNO_3 , ^{60}Co , and $\text{Co}(\text{NO}_3)_2$ were added to the soil close to the anode, and the ionic movement of the binary system was monitored for 13 days.

Preliminary Results

The following results were obtained from the initial studies using the experimental apparatus. Continuing experiments are being conducted to refine the experimental approach.

Single Component: ^{137}Cs

A spike of saturated soil containing 20 μCi of ^{137}Cs and 0.5M CsNO_3 was added to the center of the cell with an electrode separation of 9 in. The total weight of the saturated soil used was 2.3 kg. Initially, the cell radioactivity was mapped out and no electric field was applied for 24 hr, which allowed the effects of Cs concentration gradients to be elucidated. Figure 3 shows the radioactive peak area of Cs as a function of centerline distance without an applied electric field. The data points clearly overlap and indicate that the Cs movement due to a concentration gradient is negligible for a 24-hr period. This conclusion cannot be generalized to all applications, but for this case, it is reasonable to assume that ionic transport resulting from concentration gradients is not significant.

After the initial 24-hr period, a potential of 200 V was applied across the cell and the movement of Cs was monitored approximately every 24 hr. The center spike moved in a uniform, axial fashion toward the negative electrode with negligible radial dispersion. Therefore, the Cs movement can be depicted by monitoring the radioactivity along the centerline distance between the anode and cathode. Figure 4 shows a plot of the detected peak area as a function of centerline distance for various times. The data points are the measured radioactivity, and the curves shown were arbitrarily drawn to provide qualitative interpretation. Ideally, the curves shown for various times should have the same area.

From Figure 4, the radioactivity moves dramatically toward the cathode as a function of time. The peak heights of the curves decrease and broaden as the ^{137}Cs moves. There are many mechanisms to consider for a complete explanation of this observation. These mechanisms include electroosmosis, electromigration, ion exchange capacity of the soil, uniformity of the applied electric field, and others. Clearly, this is a complicated problem to model and extract quantitative information. However, qualitatively, Figure 4 demonstrates that unbound, mobile Cs can be transported through saturated Hanford soil using electrokinetics. In addition, this transport is significantly faster than pure diffusion, as shown by comparing Figures 3 and 4. Finally, it should be noted that the radioactive measurement at the center position, Position 4.5 shown in Figure 4, does not decrease to the baseline value. This appears to represent the residual ^{137}Cs that cannot be moved significantly without the addition of a sequestering agent.

The DC current response over the experiment described above is shown in Figure 5. The current initially decreases sharply and then maintains a relatively constant value. Finally, as the experiment continues beyond 200 hr, the current begins to decrease again. The local fluctuations of current were approximately ± 5 mA as determined visually over a 2-hr period. The current variations can be attributed to either electrochemical deposition of soluble metals or unstable electrode overpotentials.

After the experiment was terminated, soil samples were removed at various positions and the moisture content was measured. Because the electrokinetic cell is a closed system, the moisture content was expected to remain constant throughout the experiment, except for the water that electrolyzed at the electrodes. Table 2 shows the moisture content at

various positions within the cell. As expected, the moisture content is relatively constant. The radioactive region from the center to the cathode was not sampled because an oven for drying radioactive samples was not available.

The volume of water consumed by oxidation at the anode and reduction at the cathode is given by

$$V_{H_2O} = \frac{I t M}{F \rho z} \quad [9]$$

where t is the time period the voltage was applied, M is the molecular weight of water, and ρ is the density of water. The volume of water destroyed is estimated by using an average current of 35 mA (see Figure 4) for a 10-day period and assuming 100% current efficiency. From these conditions, a volume of 3.8 mL of water is converted to hydrogen and hydroxyl ions. This value represents approximately 1% of the total water volume added to the soil and less than 0.2% of the total weight of saturated soil used. Therefore, the water consumed via electrolysis does not introduce significant error to the moisture content measurements.

Binary System: ^{137}Cs and ^{60}Co

The second experiment involved adding three spikes approximately 1 in. in front of and parallel to the anode. The spikes were separated by 1 in. and were evenly distributed. Figure 6 shows the spike locations. The purpose of the experiment was twofold: 1) demonstrate that the experimental approach can monitor simultaneous movement of multicomponent species, and 2) determine the influence of electric-field edge effects and nonuniform electroosmotic flow developed in a closed cell on ionic movement.

Figure 7 shows three-dimensional plots of ^{60}Co radioactivity as a function of cell position and time. Figure 8 shows similar plots for the radioactivity of ^{137}Cs obtained simultaneously. The spike composition was 20 μCi ^{137}Cs , 0.25M CsNO_3 , 50 μCi ^{60}Co , and 0.25M $\text{Co}(\text{NO}_3)_2$. The soil water content was 15% by weight. The peak areas plotted in Figure 7 are a sum of the 1173 and 1333 keV emission lines. The measured ^{60}Co and ^{137}Cs radioactivity map is significantly broader than the single-component ^{137}Cs shown in Figure 4. This is caused by the high-energy gamma rays emitted by ^{60}Co , which were not effectively screened by the NaI detector shield.

From Figures 7 and 8, the Cs and Co located in the centerline of the cell moved at approximately the same velocity as the material located near the edges of the cell. This similar velocity indicates that the electric field lines are nearly uniform within the cell and edge effects can be neglected.

Conclusions

The experimental approach introduced here is being used to further understand ionic transport during electrokinetic soil remediation. The technique proved useful in demonstrating *in situ* monitoring of radioactive species transport in Hanford soil. Both single-component and binary mixtures were used. The transport of unbound ^{137}Cs and ^{60}Co within a soil matrix was measured by a gamma counter at various times during the electrokinetic process. For an applied potential of 200 V over a period of approximately 200 hr, ^{137}Cs and ^{60}Co were transported a distance of 4 to 5 in. Both ions transported at

approximately the same rate, both migrating toward the cathode. This experimental technique demonstrates that individual components can be monitored simultaneously. Therefore, this approach represents a powerful tool to optimize operating conditions for complex multicomponent systems seen in field applications.

Acknowledgments

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Table 1. Ionic mobilities of cations and anions at infinite dilution in aqueous solution and 25°C (25).

Ion	Ionic Mobility, u_j ($\text{cm}^2 \text{sec}^{-1} \text{V}^{-1} \times 10^4$)	Ion	Ionic Mobility, u_j ($\text{cm}^2 \text{sec}^{-1} \text{V}^{-1} \times 10^4$)
H ⁺	36	OH ⁻	20
K ⁺	7.6	Cl ⁻	7.9
Na ⁺	5.2	Br ⁻	8.1
Li ⁺	4.0	ClO ₄ ⁻	7.1
1/2 Ca ⁺²	6.2	NO ₃ ⁻	7.4
		1/2 SO ₄ ⁻²	8.3

Table 2. Soil moisture content as a function of cell position after a 200 V potential was applied for approximately 200 hours. All measurements are shown in weight percent water. The table depicts a top view and the shaded region represents the radioactive soil.

ANODE (+)				
15.02		16.04		16.16
16.30		16.22		16.50
15.80				16.18
15.59				16.64
15.42				16.40
CATHODE (-)				

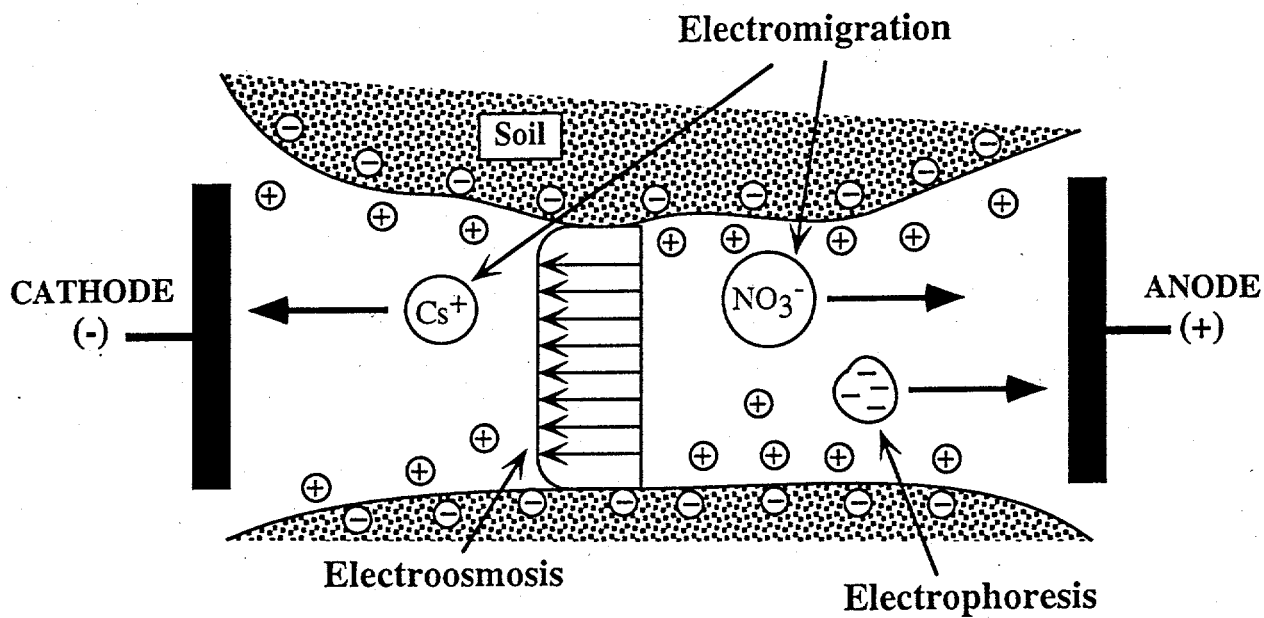


Figure 1. Schematic depicting electrokinetic phenomena of electromigration, electroosmosis, and electrophoresis.

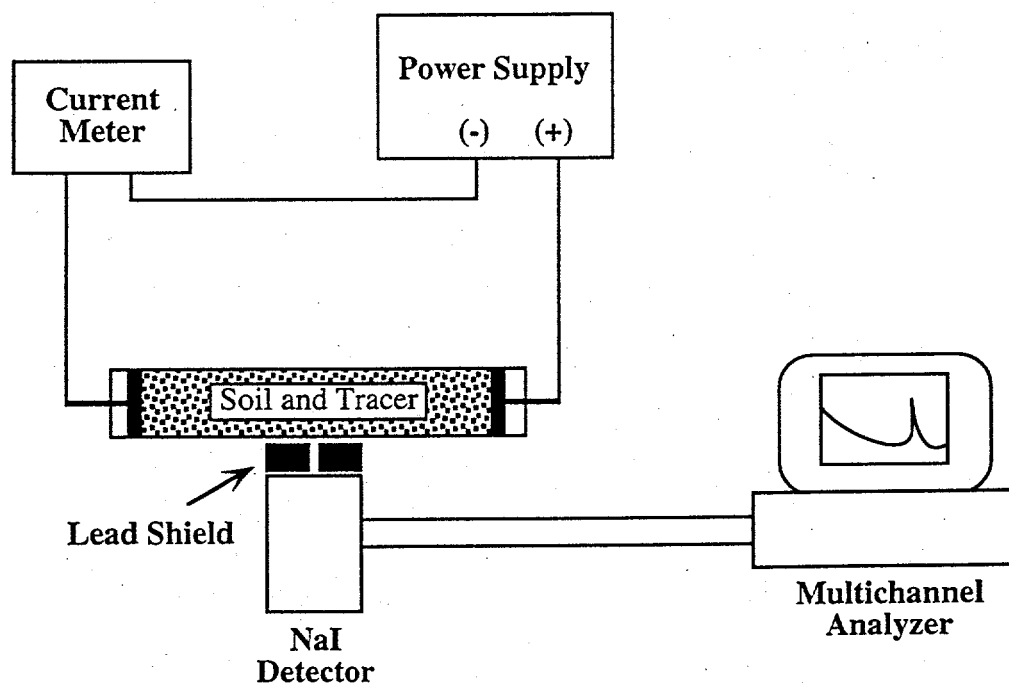


Figure 2. A schematic diagram of the experimental equipment used to monitor radioactive ^{137}Cs and ^{60}Co ions in Hanford soil.

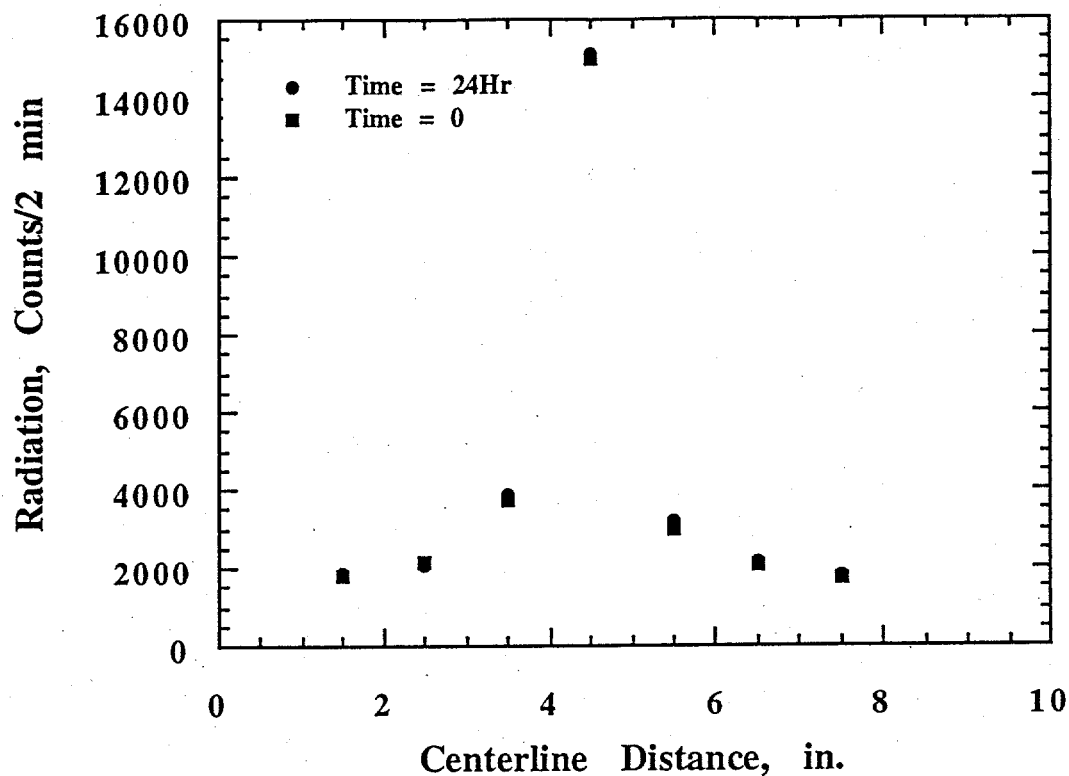


Figure 3. Radioactivity of ^{137}Cs spike as a function of centerline distance for 24-hour period.

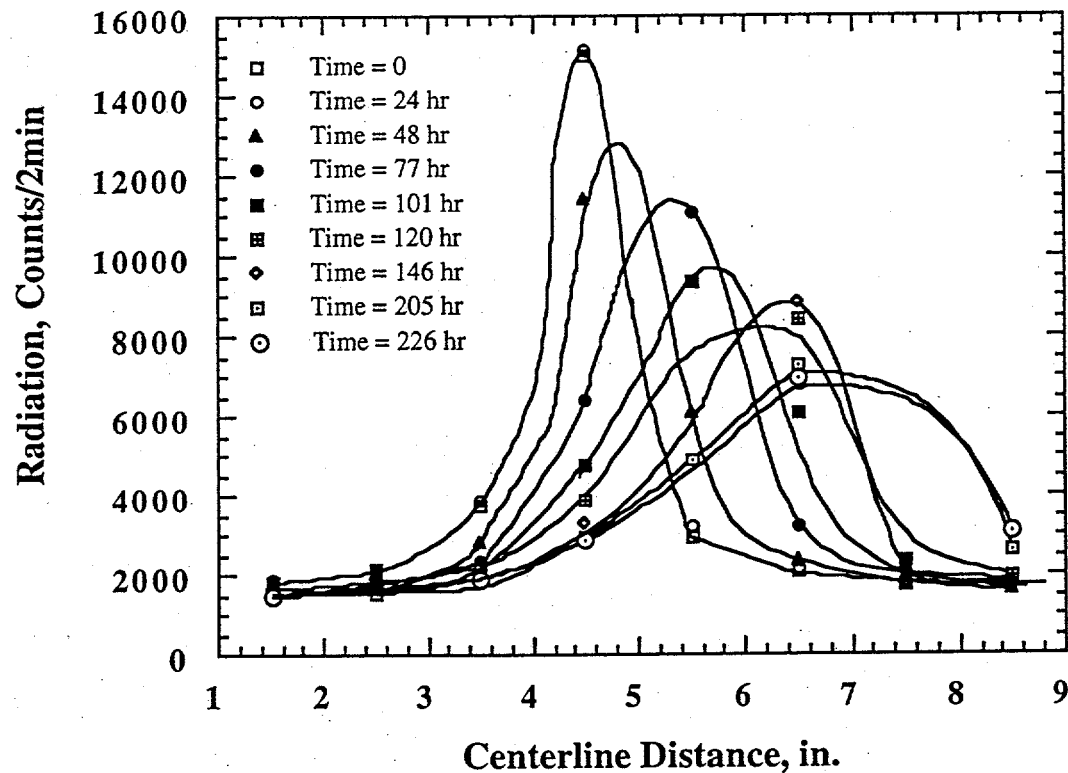


Figure 4. Peak area of 662 keV emission line as a function of centerline distance for a ^{137}Cs spike with an applied potential of 200 V.

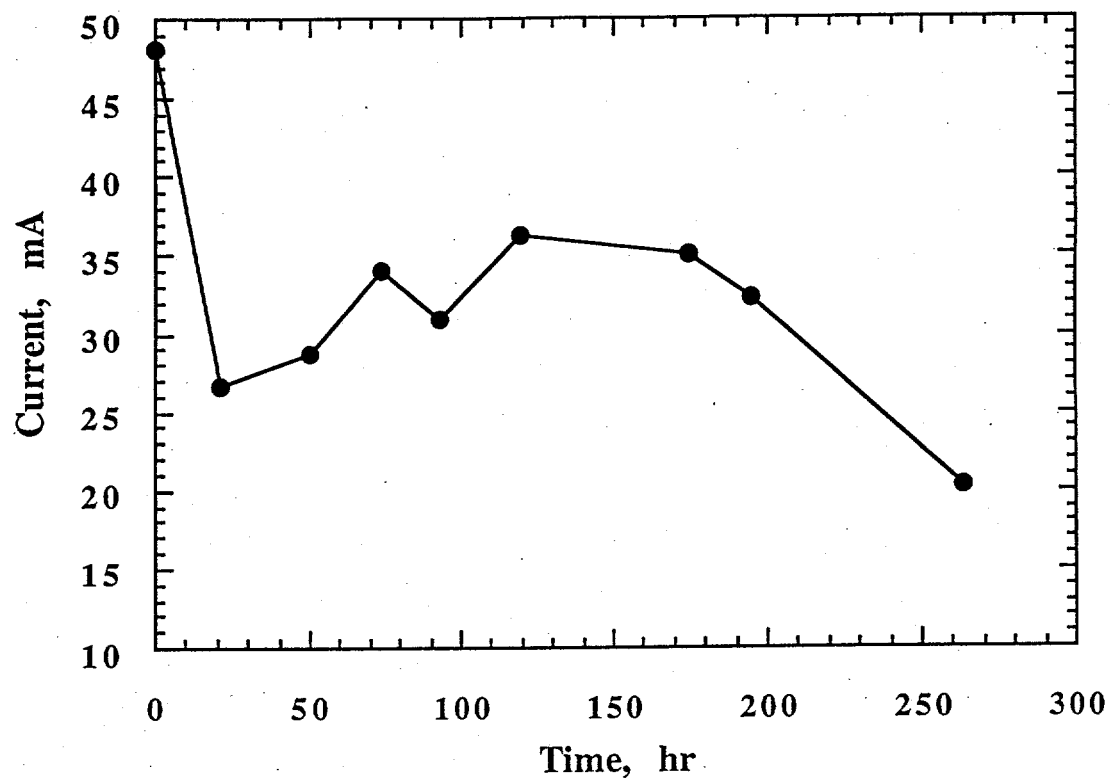


Figure 5. Current as a function of time for the single-component ^{137}Cs spike.

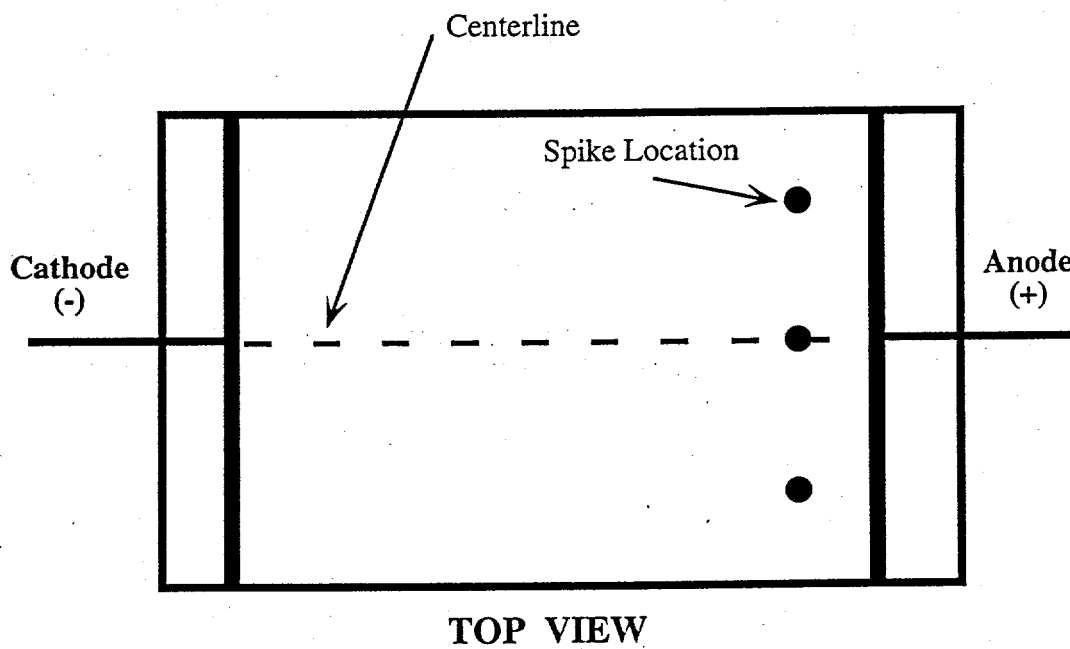


Figure 6. Location of Cs and Co radioactive spikes (top view of cell).

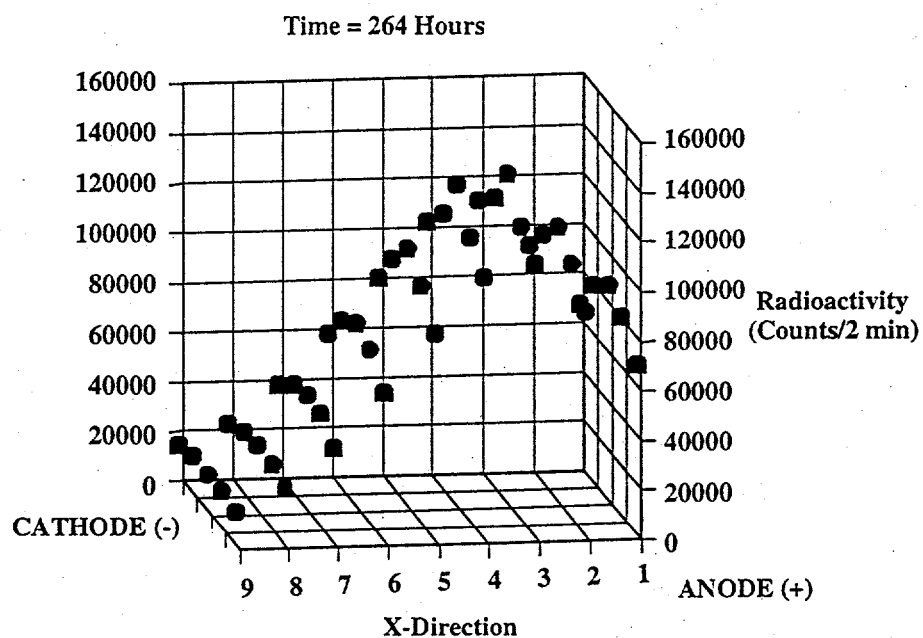
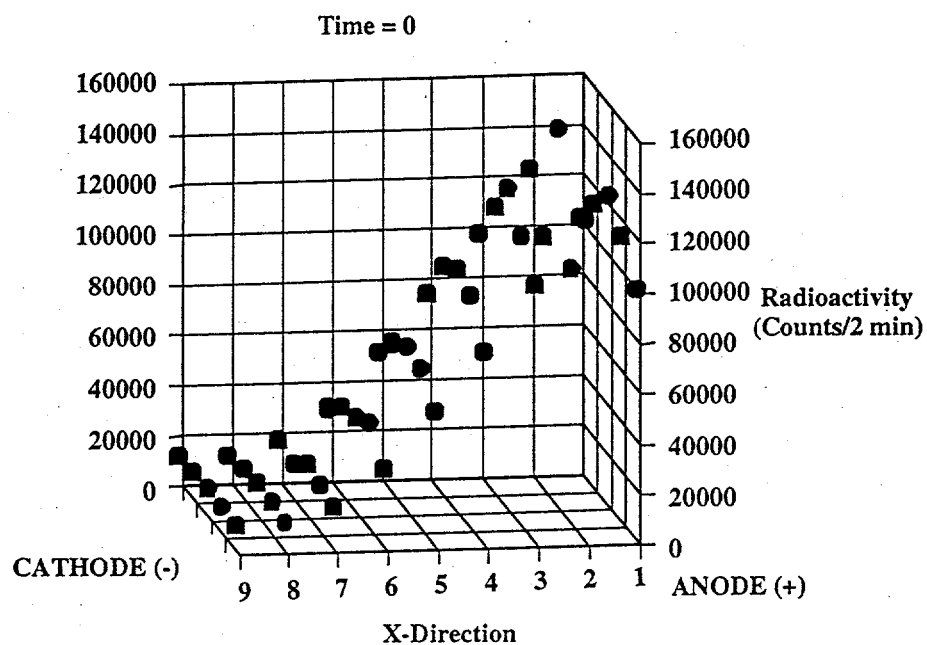


Figure 7. Three-dimensional view of ^{137}Cs radioactivity as a function of time.

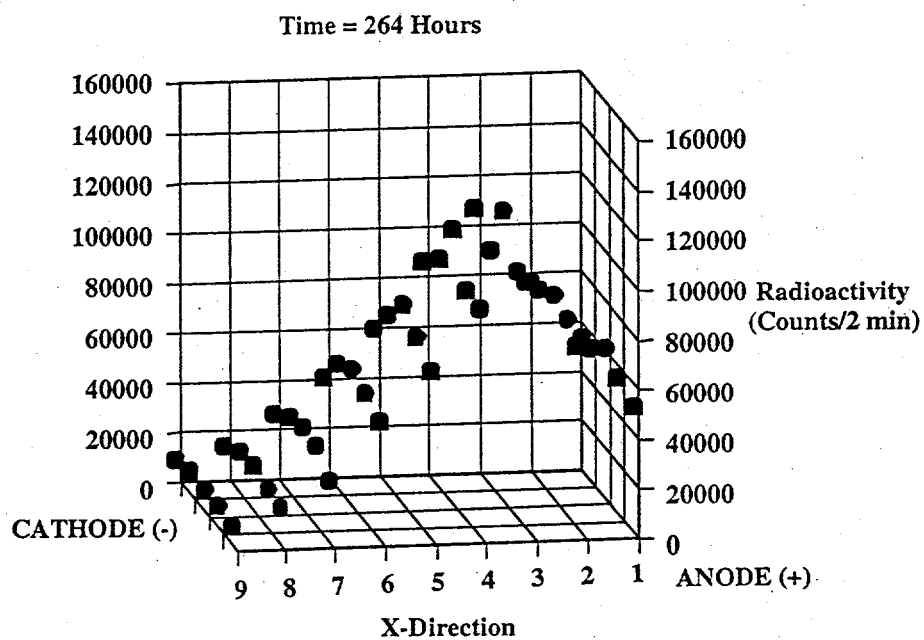
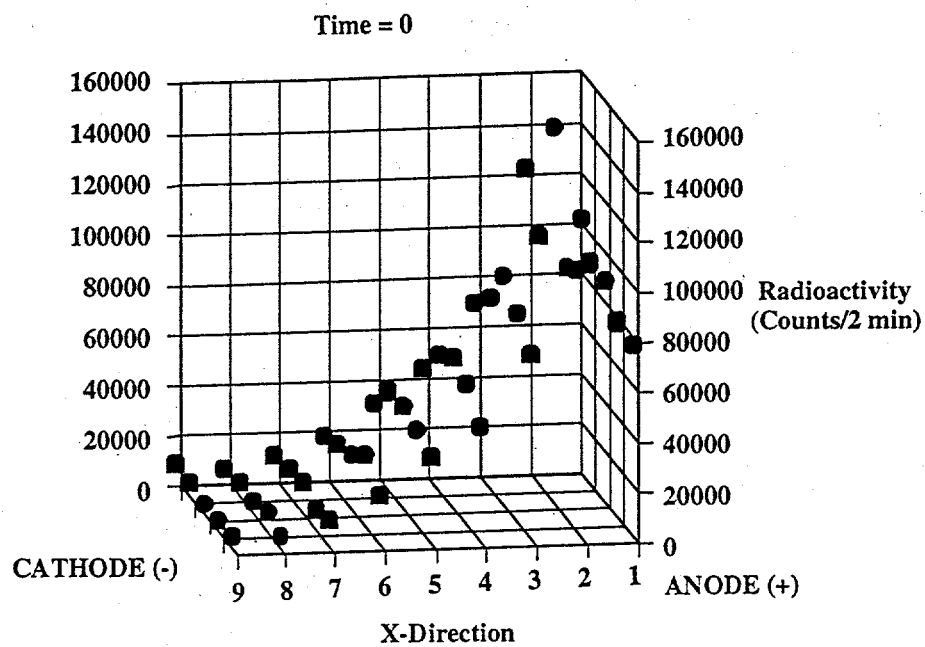


Figure 8. Three-dimensional view of ^{60}Co radioactivity as a function of time.

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