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A Direct High Voltage Electrophoresis Study of ^{95}Zr in Na_2CO_3-$UO_2(NO_3)_2 \cdot 6H_2O$ Systems

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TECHNICAL NOTE

A Direct High Voltage Electrophoresis Study of ^{95}Zr in Na_2CO_3–$UO_2(NO_3)_2 \cdot 6H_2O$ Systems

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INTRODUCTION

The recovery and reutilization of nonfissioned residual uranium clearly depends on the separation of the fission products present. Knowledge of the chemical behavior of these species in aqueous solution facilitates the recovery treatment and, with this, the possibility of minimizing the process times and the volumes of liquid wastes. In this recovery process the preparation of the anionic uranyl tricarbonate complex is of significant importance as a soluble species (1–5). The study of the influence of postreaction time on the quality and concentration of the ionic species of fission products allowed us to determine the best purification conditions of residual uranium, optimizing its handling for further treatment.

The purpose of this study has therefore been to characterize the variable postreaction time of the anionic zirconium complex and to minimize its presence in the uranium recovery stage.

EXPERIMENTAL

Hexavalent uranium reacts with sodium carbonate to produce an anionic uranyl tricarbonate species which is soluble and extractable. When this reaction is established in the presence of known activities of ^{95}Zr, the

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reaction product is a mixture of ionic species of uranyl tricarbonate and ^{95}Zr. The ionic ^{95}Zr species in this product tend to vary their conformation due to the effect of the postreaction time. Measurements repeated with the same sample gave different results. This condition is based on the tendency of positive ions to form complexes with anions in aqueous solution, and the dominant species in these complexes appears to be monomeric and/or dimeric forms of some kind. However, the final product is a consequence of the postreaction time. To evaluate this influence, a system was used which allowed for the exclusive production of the anionic uranyl tricarbonate complex as a mononuclear species from hexahydrated uranyl nitrate and anhydrous sodium carbonate (analytical grade obtained from Merck, for 0.05 and 0.15 molar concentrations, respectively). The reaction system was prepared by treating both reactants in equivalent amounts for a total volume of 15 mL with the addition of 1 μCi of ^{95}Zr obtained from the supplier Damri. The final aqueous system had a pH of approximately 9.2.

The separation of the zirconium ionic species was done by using a high voltage electrphoresis technique. The electrolyte solution used was 0.1 M ammonium carbonate, and the potential applied was of 2500 V for 30 minutes. For the experimental measurements, 20 μL of the synthetic solution containing ^{95}Zr was deposited on 1 × 60 cm Whatman No. 2 paper strips. These conditions were optimal for traces amounts which corresponded to the specimens under investigation, with samples of recent preparation (t = 0) and after 24, 48, and 72 hours, under the same experimental conditions. The migration of ^{95}Zr was determinated by γ-spectrometry with a Ge(hi) detector coupled to a Canberra 4096 channel analyzer. ^{95}Zr activity was measured by integrating the area under the 0.724 MeV γ-peak.

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RESULTS AND DISCUSSION

Studies by a wide variety of classical and spectroscopic techniques have demonstrated the complexity of aqueous solutions of zirconium salts (6–8). The direct room temperature electrophoresis technique has been highly valuable for determining the extent of ionic complex formation.

As seen from the results, zirconium reacts to form complexes in association with the sodium cation to produce compounds with different contents of carbonates (9–13). Figure 1(A) shows the electrophoretogram from a recently prepared solution. In this figure the most significant feature is the appearance of eight anionic zirconium complexes of essentially different area displacements. This pattern of behavior prevailed in the electrophoretograms of all solutions with significant changes for different time

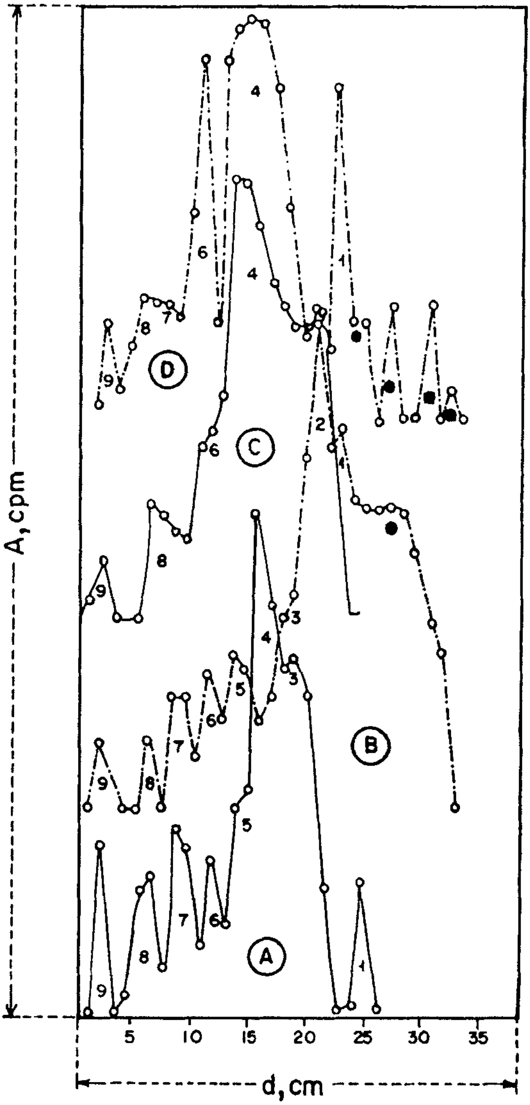

FIG. 1 Electrophoretogram of a sample of the ^{95}Zr carbonated complex analyzed at various times after its preparation.

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TABLE 1
Migration of Anionic Zirconium Complexes in 0.1 M Ammonium Carbonate under a Potential of 2500 V for 30 Minutes

Distance traveled toward anode (cm) at postreaction times of:			
0 hours[a]	24 hours	48 hours	144 hours
2	2	2	2
6	6	6	6
8	8	—	8
11	11	11	11
14	14	—	—
15	—	15	15
19	19	—	—
—	21	21	21
24	23	—	23
—	—	—	25
—	28	—	28
—	—	—	31
—	—	—	33

[a] Freshly prepared solution.

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intervals. Table 1 gives the electrophoretic mobility values and the corresponding differences in behavior. The values show a gradual equilibrium that was reproducible for a period of 4 weeks, indicating the presence of the same dominant species throughout.

After 24 hours (Fig. 1B) the system aqueous showed a new species (Fig. 1B2) arising from unstable molecules. A probable explanation of this behavior is that in freshly prepared solutions the transformation is not yet finished and therefore the concentration of this new species is presumably due to the slow breaking up of the species dominating in freshly prepared solutions (Figs. 1A1, 3, and 4). A similar behavior was found for solutions aged 48 and 72 hours (Figs. 1C and 1D, respectively). Here, the low molecular weight species are strongly hydrolysed and occur mainly as aggregates. In this respect it was found that the higher aggregates are dependent on the age of the solution in which equilibrium is reached slowly with the appearance of other low molecular weight species (Fig. 1D*). This is evident from a comparison of fresh and aged solutions.

It is clear from these results that it would be possible to separate zirconium from uranium by selective sorption of uranium from a nitric acid solution.

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