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Presented at the International Conference on
Chemistry and Migration Behavior of Actinides
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Munich, FRG, September 14–18, 1987,
and to be published in Radiochimica Acta

# Some Effects of Data Base Variations on Numerical Simulations of Uranium Migration

C.L. Carnahan

December 1987

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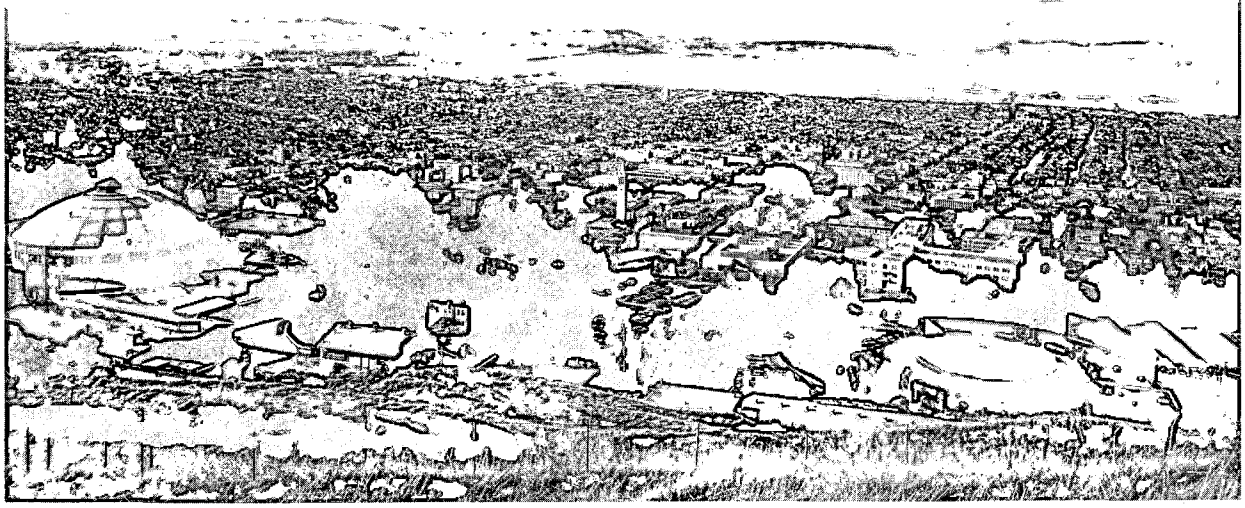

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# SOME EFFECTS OF DATA BASE VARIATIONS ON NUMERICAL SIMULATIONS OF URANIUM MIGRATION

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# Some Effects of Data Base Variations on Numerical Simulations of Uranium Migration

By C. L. CARNAHAN, Earth Sciences Division, Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720, U. S. A.

**Abstract**

Numerical simulations of migration of chemicals in the geosphere depend on knowledge of identities of chemical species and on values of chemical equilibrium constants supplied to the simulators. In this work, some effects of variability in assumed speciation and in equilibrium constants were examined, using migration of uranium as an example. Various simulations were done of uranium migration in systems with varying oxidation potential, pH, and major component content. A simulation including formation of aqueous species $UO_2^{2+}$, $UO_2CO_3^0$, $UO_2(CO_3)_2^{2-}$, $UO_2(CO_3)_3^{4-}$, $(UO_2)_2CO_3(OH)_3^-$, $UO_2^+$, $U(OH)_4^0$, and $U(OH)_5^-$ is compared to a simulation excluding formation of $UO_2^+$ and $U(OH)_5^-$. These simulations relied on older data bases, and they are compared to a further simulation using recently published data on formation of $U(OH)_4^0$, $(UO_2)_2CO_3(OH)_3^-$, $UO_2(CO_3)_3^{5-}$, and $U(CO_3)_5^{6-}$. Significant differences in dissolved uranium concentrations are noted among the simulations. Differences are noted also in precipitation of two solids, $USiO_4$(c) (coffinite) and $CaUO_4$(c) (calcium uranate), although the solubility products of the solids were not varied in the simulations.

## Introduction

In recent years, considerable progress has been made in the development of thermodynamically based computer programs that incorporate homogeneous and heterogeneous chemical reactions, under the assumption of chemical equilibrium, into the solution of multicomponent solute transport equations (e.g., [1–5]). These computational methods rely upon thermodynamic data bases (in particular, values of equilibrium constants and their variations with temperature) for the chemical reactions to be included in numerical simulations of reactive chemical migration.

The work reported here is an investigation of the sensitivity of computed uranium concentrations to variations of data bases used in numerical simulations of uranium transport. The calculations were made with the computer program *THCC* [4,5]. Uranium was chosen as the element of interest for several reasons. First, uranium is interesting chemically because of its possible existence in three principal oxidation states (IV, V, VI) and its ability to form complexes with ligands occurring in natural waters. Second, as explained in detail in the following section, disagreement exists in the literature about the identities and formation constants of certain uranium species in aqueous solution. Third, practical interest in migration of uranium arises in investigations of formation of ore bodies and in performance assessments of geological repositories for nuclear wastes.

## Some differences in uranium data bases

In this work, no attempt was made to create a comprehensive, self-consistent, critically evaluated data base. Instead, reliance was placed on existing compilations [6,7,8] of equilibrium constants for reactions of interest, supplemented by a selection of data from the more recent literature. In order to keep the sensitivity analyses relatively uncomplicated,

attention was focused on equilibria involving the aqueous species $U(OH)_4^0$, $U(OH)_5^-$, $UO_2^+$, $(UO_2)_2CO_3(OH)_3^-$, $UO_2(CO_3)_3^{5-}$, and $U(CO_3)_5^{6-}$.

The older literature, compiled in [6], contains formation constants for a series of hydrolysis products of U(IV) including $U(OH)_4^0$ and $U(OH)_5^-$. The work [9,10] upon which these constants are based, as well as the existence of $U(OH)_5^-$, has been questioned [11]. In addition, new data for the stability constant of $U(OH)_4^0$ have been reported [12,13] that do not support the older data.

The older literature provides data on $UO_2^+$ which indicate a significant field of stability for this species in the Eh-pH domain [7]. However, it has been pointed out [14] that $UO_2^+$ has a strong tendency to disproportionate. Thus $UO_2^+$ may not be an appropriate species for consideration in geological environments. On the other hand, complexation may increase the stability of U(V).

New data have been reported for formation of the U(IV) species $U(CO_3)_5^{6-}$ [15], the U(V) species $UO_2(CO_3)_3^{5-}$ [16], and the binuclear U(VI) species $(UO_2)_2CO_3(OH)_3^-$ [17].

The sensitivity analyses to be described here are intended to demonstrate effects on concentrations of migrating uranium of inclusion or exclusion of certain species and of differences in equilibrium constants. When necessary, published values of equilibrium constants have been corrected to zero ionic strength by use of an extended Debye-Hückel equation [6].

## Bases of the simulations

All simulations assumed that an influent fluid bearing dissolved uranium at a high Eh and moderate pH flowed into and mixed with an initial fluid containing no uranium at a low Eh and high pH. The composition of the initial fluid matched exactly the reported [18] composition of a Swedish deep groundwater. The composition of the influent fluid matched the major ion content and pH reported [18] for a different Swedish deep groundwater, but the Eh was assumed much higher than the reported value of $-0.22$ v and the fluid was assumed to contain dissolved uranium at a total concentration of $4.31\times10^{-5}M$. This uranium concentration is about one order of magnitude smaller than the calculated solubility of $UO_2(OH)_2 \cdot H_2O(c)$ (schoepite), the uranium-containing solid that is most stable under the conditions assumed in the influent fluid. Detailed compositions of the two simulated fluids are shown in Table 1.

The initial fluid was contained within a one-dimensional spatial domain simulating a porous or fractured geological medium. At time zero, the influent fluid began to penetrate this domain with a fluid velocity of $1\times10^{-6}$ m/s. The domain had a dispersivity of 0.02 m and a diffusion coefficient of $1\times10^{-10}$ $m^2/s$.

The temperature of the simulated system was held constant at 25 °C. The corresponding equilibrium constants (expressed as $\log K$ values) for simulated chemical reactions are shown in Table 2. It is noted that reactions forming aqueous complexes and dissolving solids are expressed in terms of a set of "basis species" consisting of $UO_2^{2+}$, $CO_3^{2-}$, $Si(OH)_4^0$, $H^+$, and $e^-$.

The results of three simulations will be presented here. The same compositions of influent and initial fluids and identical physical properties were used in the simulations. The same equilibrium constants for ionization of $H_2O$ and for formation of the complexes $HCO_3^-$, $H_2CO_3^0$, $UO_2CO_3^0$, $UO_2(CO_3)_2^{2-}$, and $UO_2(CO_3)_3^{4-}$ were used in all cases. The possibility of precipitation of six solid phases was considered, and their solubility products were held constant for all simulations. The solid phases considered were $UO_2(OH)_2 \cdot H_2O(c)$ (schoepite), $UO_2(c)$ (uraninite), $USiO_4(c)$ (coffinite), $Ca(UO_2)_2(SiO_3OH)_2(c)$ (uranophane), $UO_2CO_3(c)$ (rutherfordine), and $CaUO_4(c)$ (calcium uranate). The simulations differed in the identities of certain aqueous species and in values of equilibrium constants for the formation of certain species.

Case 1. Older literature values [6] of $\log K$'s for formation of the species $UO_2^+$, $U(OH)_4^0$, $U(OH)_5^-$, and $(UO_2)_2CO_3(OH)_3^-$ were used.

Case 2. The same $\log K$ values as in Case 1 were used for $(UO_2)_2CO_3(OH)_3^-$ and $U(OH)_4^0$, but $UO_2^+$ and $U(OH)_5^-$ were excluded from the simulation.

Case 3. Newer literature values of $\log K$'s for formation of $(UO_2)_2CO_3(OH)_3^-$ [17] and $U(OH)_4^0$ [13,12] were used, $UO_2^+$ and $U(OH)_5^-$ were excluded, and the species $UO_2(CO_3)_3^{5-}$ [16] and $U(CO_3)_5^{6-}$ [15,16] were added to the chemical system.

## Results and discussion

Computational results for the three cases are displayed in Figs. 1–9.

At a simulated time of $1\times10^6$ s, profiles of Eh and concentrations of $Ca^{2+}$, $Si(OH)_4^0$, $CO_3^{2-}$, and $H^+$ do not differ significantly among the three cases (Figs. 1–3). However, total dissolved uranium concentrations exhibit significant differences attributable to the different data bases used. For ease of comparison, the total dissolved uranium concentrations are displayed together in Fig. 4. Here it is seen that, relative to Case 1, exclusion of $U(OH)_5^-$ from Case 2 decreases total uranium concentrations in the region of precipitation of $USiO_4(c)$ by more than two orders of magnitude and extends this region by a factor of two. On the other hand, total uranium concentrations in equilibrium with $USiO_4(c)$ for Case 3 exceed those for Case 1 by almost three orders, and they exceed those for Case 2 by five orders of magnitude. Beyond the region of precipitation, total uranium concentrations for Case 3 exceed those for Cases 1 and 2 by about two orders of magnitude.

It is noted that, of the six solid phases considered in the simulations, only $USiO_4(c)$ and $CaUO_4(c)$ were found to precipitate at any simulated time less than $2\times10^6$ s. Precipitation of $CaUO_4(c)$ was confined to the region of high Eh near the influent boundary in Cases 1 and 2, and did not occur anywhere in Case 3.

Concentrations of individual uranium species at the simulated time of $1\times10^6$ s are displayed in Figs. 5–7. Carbonato complexes of dioxouranium(VI) are dominant in the region of high Eh, and show only slight differences among the simulations. The concentration of $(UO_2)_2CO_3(OH)_3^-$ is slightly increased in Case 3 (Fig. 7). In Case 1 (Fig. 5), $UO_2^+$ contributes negligibly, and the absence of this species from Cases 2 and 3 does not affect the computations. Similarly, the "new" species $UO_2(CO_3)_3^{5-}$ and $U(CO_3)_5^{6-}$ contribute

negligibly in Case 3 (Fig. 7). The major differences in the species' profiles occur in the region of low Eh and are attributable to $U(OH)_4^0$ and $U(OH)_5^-$.

"Concentrations" of uranium solid phases (moles of solid per $dm^3$ of fluid phase) at $1\times10^6$ s are displayed in Fig. 8, and show the same trends as the fluid-phase concentrations in Fig. 4. Taken together, the two figures indicate that "new" data (Case 3) for formation of $U(OH)_4^0$ predict total migration of uranium significantly larger than that predicted by the older data. The difference between the data sets is increased if $U(OH)_5^-$ is excluded from consideration (Case 2).

Fig. 9 shows total uranium concentrations in the fluid phase at a point located 0.18 m from the influent boundary as functions of time. A change of solid composition from $USiO_4(c)$ to $CaUO_4(c)$ is observed as a result of increasing Eh with time at this point. However, migration of the Eh front is retarded, relative to the fluid velocity, by a factor of about five. Significant differences among the three cases simulated can be seen at early times when Eh values are close to the initial value assumed for the spatial domain.


## Acknowledgement

This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Division of Engineering and Geosciences, U. S. Department of Energy, under Contract No. DE-AC03-76SF00098.


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## Tables

Table 1. Composition of initial and influent solutions

Table 2. Chemical reactions and $\log K$ values used in the simulations

Table 1. Compositions of initial and influent solutions

| Component | Initial | Influent |
|---|---|---|
| $Na^+$ | $1.30\times10^{-2}M$ | $1.60\times10^{-3}M$ |
| $Ca^{2+}$ | $1.00\times10^{-3}M$ | $6.70\times10^{-4}M$ |
| $Cl^-$ | $1.47\times10^{-2}M$ | $5.34\times10^{-4}M$ |
| $Si(OH)_4^0$ | $1.10\times10^{-4}M$ | $1.10\times10^{-4}M$ |
| total C (a) | $2.60\times10^{-4}M$ | $3.00\times10^{-3}M$ |
| total U (b) | 0. | $4.31\times10^{-5}M$ |
| Eh | −0.35 v | +0.30 v |
| pH | 9.05 | 6.93 |

a. $CO_3^{2-} + HCO_3^- + H_2CO_3^0$
b. sum of all U species

Table 2. Chemical reactions and $\log K$ values used in the simulations

| Reaction | $\log K$ | Source |
|---|---|---|
| used in all cases: | | |
| $H_2O = H^+ + OH^-$ | −13.99 | [6] |
| $H^+ + CO_3^{2-} = HCO_3^-$ | 10.34 | [6] |
| $2H^+ + CO_3^{2-} = H_2CO_3^0$ | 16.70 | [6] |
| $UO_2^{2+} + CO_3^{2-} = UO_2CO_3^0$ | 10.09 | [6] |
| $UO_2^{2+} + 2CO_3^{2-} = UO_2(CO_3)_2^{2-}$ | 17.13 | [6] |
| $UO_2^{2+} + 3CO_3^{2-} = UO_2(CO_3)_3^{4-}$ | 20.64 | [6] |
| $UO_2(OH)_2 \cdot H_2O(c) + 2H^+ = UO_2^{2+} + 3H_2O$ | 5.40 | [7] |
| $Ca(UO_2)_2(SiO_3OH)_2(c) + 6H^+ = Ca^{2+} + 2UO_2^{2+} + 2Si(OH)_4^0$ | 17.21 | [7] |
| $UO_2CO_3(c) = UO_2^{2+} + CO_3^{2-}$ | −14.29 | [6] |
| $CaUO_4(c) + 4H^+ = Ca^{2+} + UO_2^{2+} + 2H_2O$ | 15.00 | [8] |
| $USiO_4(c) + 2H_2O = UO_2^{2+} + Si(OH)_4^0 + 2e^-$ | −18.54 | [7] |
| $UO_2(c) = UO_2^{2+} + 2e^-$ | −13.19 | [6] |
| used in case 1: | | |
| $2UO_2^{2+} + CO_3^{2-} + 3H_2O = (UO_2)_2CO_3(OH)_3^- + 3H^+$ | −1.92 | [6] |
| $UO_2^{2+} + e^- = UO_2^+$ | 2.11 | [6] |
| $UO_2^{2+} + 2e^- + 2H_2O = U(OH)_4^0$ | −0.02 | [6] |
| $UO_2^{2+} + 2e^- + 3H_2O = U(OH)_5^- + H^+$ | −4.65 | [6] |
| used in case 2: | | |
| $2UO_2^{2+} + CO_3^{2-} + 3H_2O = (UO_2)_2CO_3(OH)_3^- + 3H^+$ | −1.92 | [6] |
| $UO_2^{2+} + 2e^- + 2H_2O = U(OH)_4^0$ | −0.02 | [6] |
| used in case 3: | | |
| $2UO_2^{2+} + CO_3^{2-} + 3H_2O = (UO_2)_2CO_3(OH)_3^- + 3H^+$ | −0.24 | [17] |
| $UO_2^{2+} + 3CO_3^{2-} + e^- = UO_2(CO_3)_3^{5-}$ | 14.93 | [16] |
| $UO_2^{2+} + 2e^- + 2H_2O = U(OH)_4^0$ | 4.40 | [13,12] |
| $UO_2^{2+} + 5CO_3^{2-} + 4H^+ + 2e^- = U(CO_3)_5^{6-} + 2H_2O$ | 52.00 | [15,16] |

## Figure captions

**Fig. 1.** Case 1: Eh and concentrations of fluid components at time $1\times10^6$ s. Filled symbols denote presence of solid phases.

**Fig. 2.** Case 2: Eh and concentrations of fluid components at time $1\times10^6$ s. Filled symbols denote presence of solid phases.

**Fig. 3.** Case 3: Eh and concentrations of fluid components at time $1\times10^6$ s. Filled symbols denote presence of solid phases.

**Fig. 4.** Total concentrations of uranium in fluid phases for Cases 1, 2, and 3 at time $1\times10^6$ s. Filled symbols denote presence of solid phases.

**Fig. 5.** Case 1: Concentrations of uranium solution species at time $1\times10^6$ s.

**Fig. 6.** Case 2: Concentrations of uranium solution species at time $1\times10^6$ s.

**Fig. 7.** Case 3: Concentrations of uranium solution species at time $1\times10^6$ s.

**Fig. 8.** Concentrations of uranium solid phases for Cases 1, 2, and 3 at time $1\times10^6$ s.

**Fig. 9.** Total concentrations of uranium in fluid phases for Cases 1, 2, and 3 at $x = 0.18$ m.

FIGURE 1

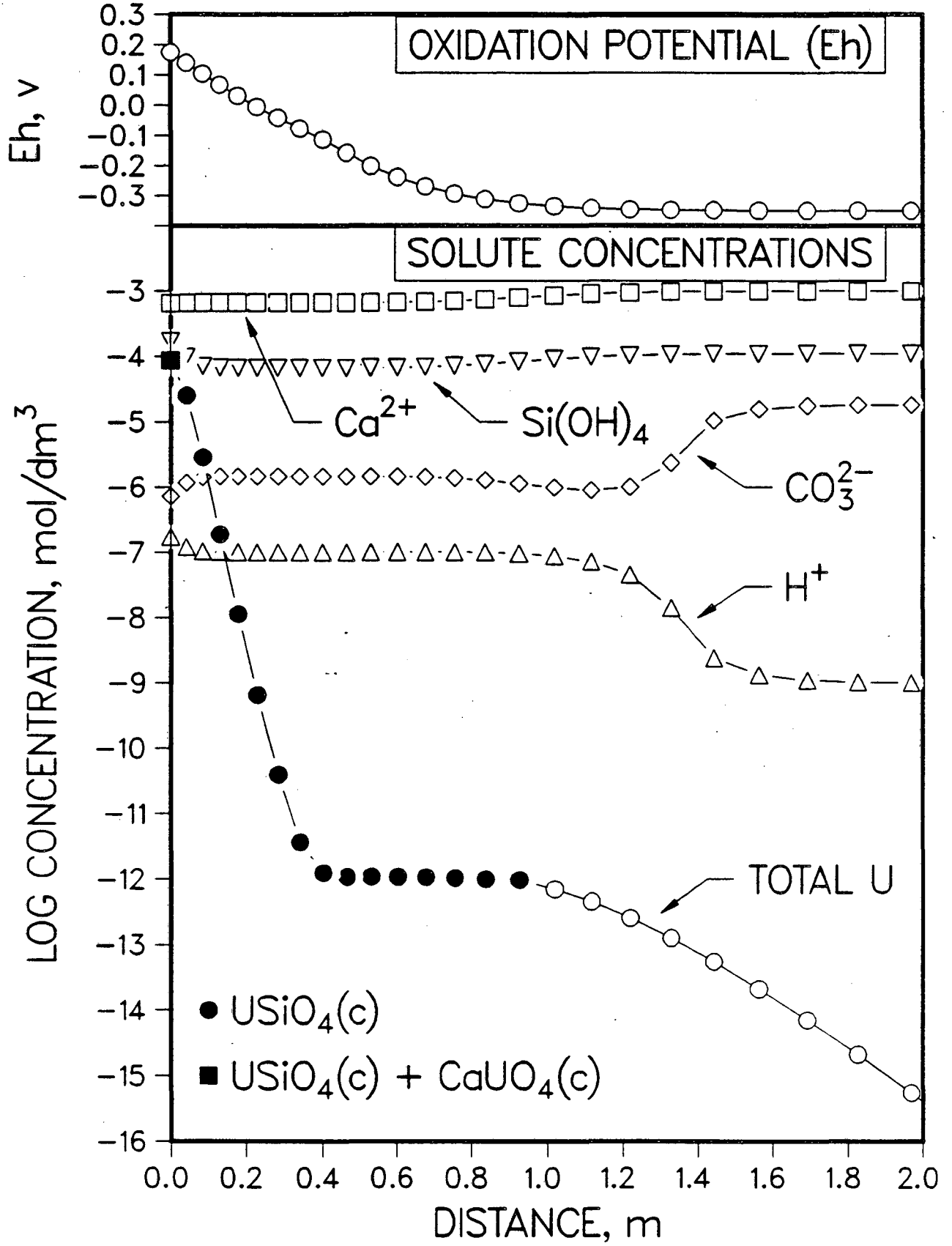

OXIDATION POTENTIAL (Eh)
Eh, v
0.3
0.2
0.1
0.0
−0.1
−0.2
−0.3
SOLUTE CONCENTRATIONS
LOG CONCENTRATION, mol/dm3
−3
−4
−5
−6
−7
−8
−9
−10
−11
−12
−13
−14
−15
−16
Ca2+
Si(OH)4
CO32−
H+
TOTAL U
USiO4(c)
USiO4(c) + CaUO4(c)
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0
DISTANCE, m

FIGURE 2

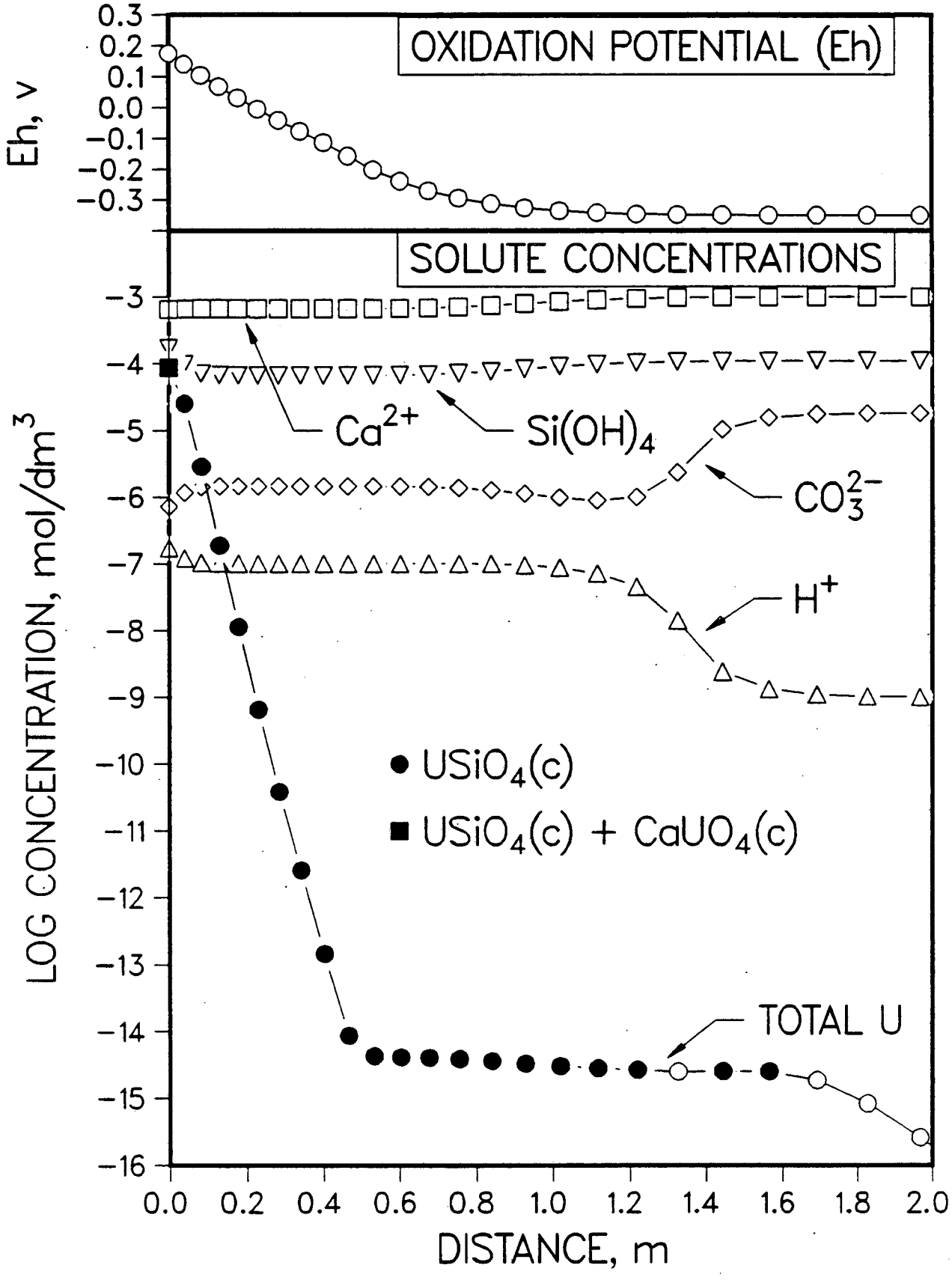

OXIDATION POTENTIAL (Eh)
Eh, v
0.3
0.2
0.1
0.0
−0.1
−0.2
−0.3
SOLUTE CONCENTRATIONS
LOG CONCENTRATION, mol/dm3
−3
−4
−5
−6
−7
−8
−9
−10
−11
−12
−13
−14
−15
−16
Ca2+
Si(OH)4
CO32−
H+
USiO4(c)
USiO4(c) + CaUO4(c)
TOTAL U
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0
DISTANCE, m

FIGURE 3

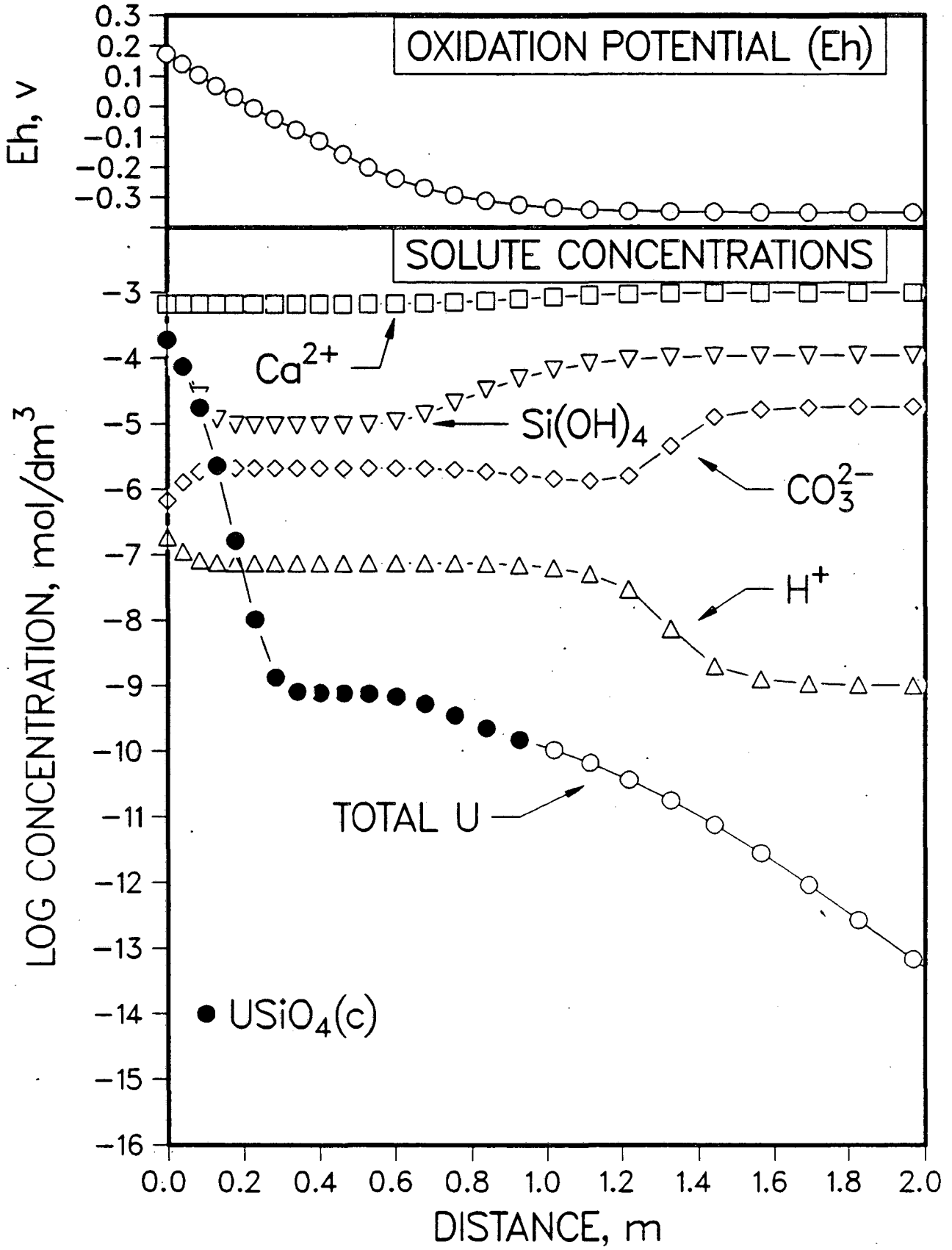

OXIDATION POTENTIAL (Eh)
SOLUTE CONCENTRATIONS
Eh, v
0.3
0.2
0.1
0.0
−0.1
−0.2
−0.3
LOG CONCENTRATION, mol/dm3
−3
−4
−5
−6
−7
−8
−9
−10
−11
−12
−13
−14
−15
−16
Ca2+
Si(OH)4
CO32−
H+
TOTAL U
USiO4(c)
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0
DISTANCE, m

FIGURE 4

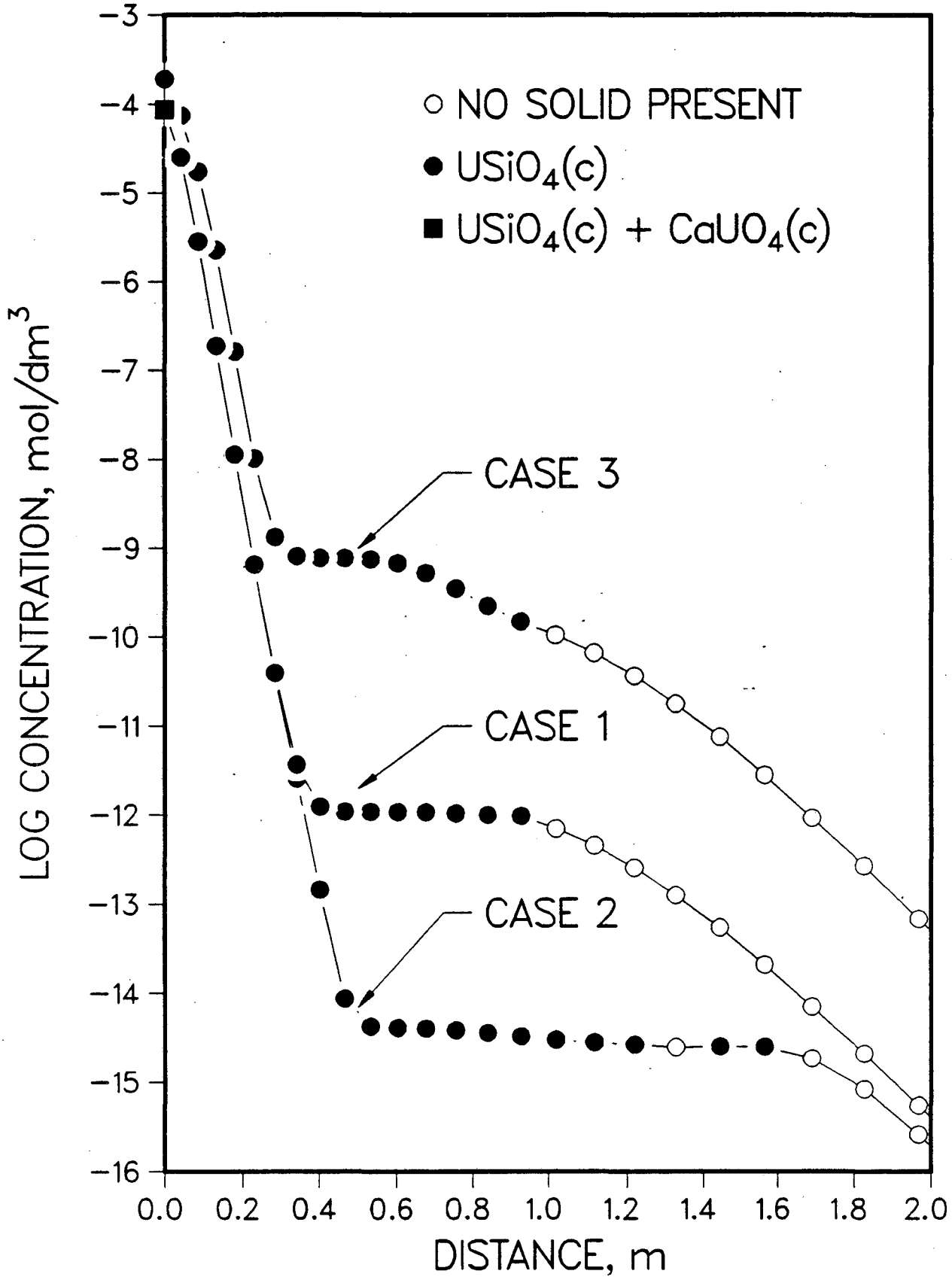

NO SOLID PRESENT
USiO4(c)
USiO4(c) + CaUO4(c)
CASE 3
CASE 1
CASE 2
LOG CONCENTRATION, mol/dm3
DISTANCE, m
−3
−4
−5
−6
−7
−8
−9
−10
−11
−12
−13
−14
−15
−16
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0

FIGURE 5

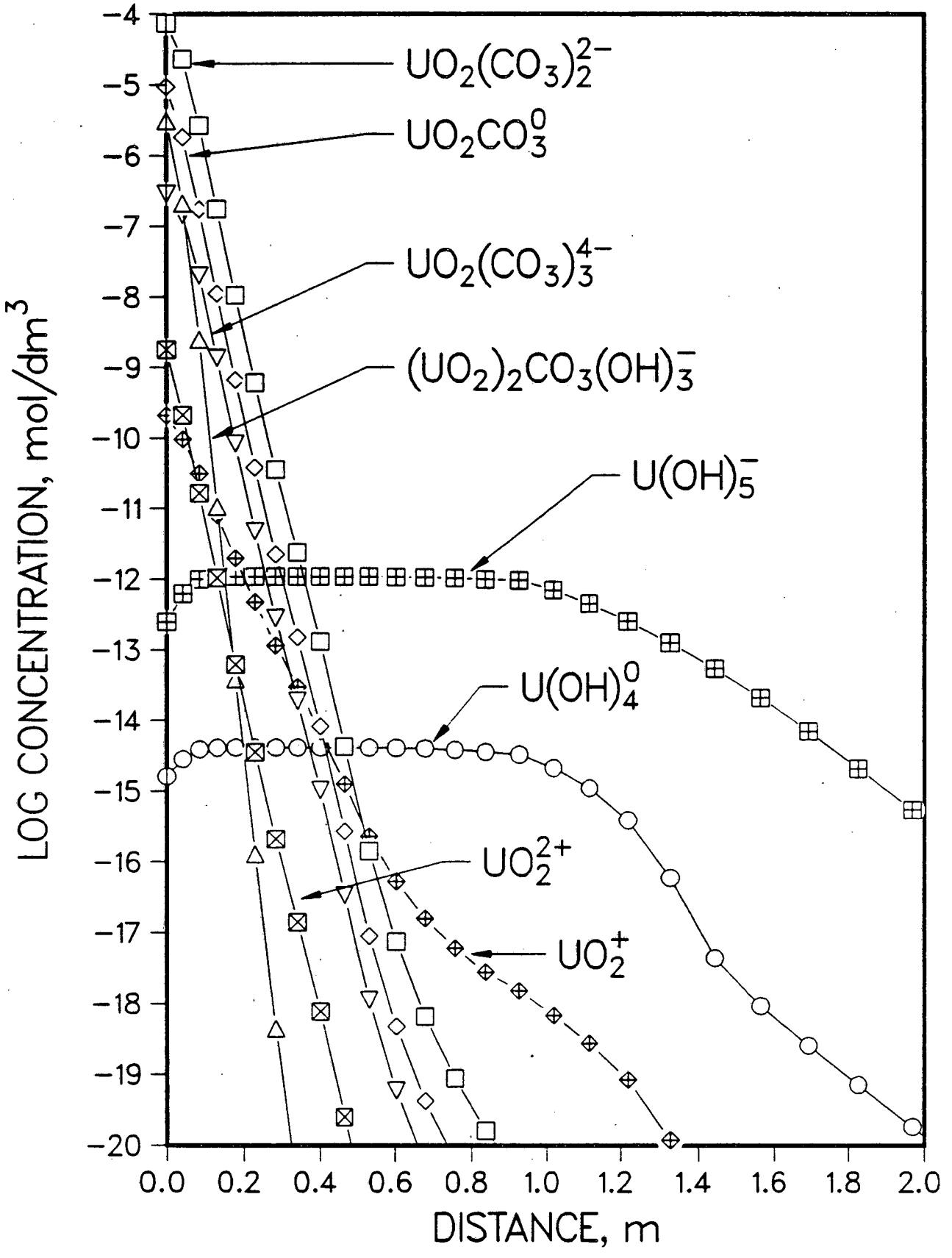

$UO_2(CO_3)_2^{2-}$
$UO_2CO_3^0$
$UO_2(CO_3)_3^{4-}$
$(UO_2)_2CO_3(OH)_3^-$
$U(OH)_5^-$
$U(OH)_4^0$
$UO_2^{2+}$
$UO_2^+$
LOG CONCENTRATION, mol/dm$^3$
DISTANCE, m
-4
-5
-6
-7
-8
-9
-10
-11
-12
-13
-14
-15
-16
-17
-18
-19
-20
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0

FIGURE 6

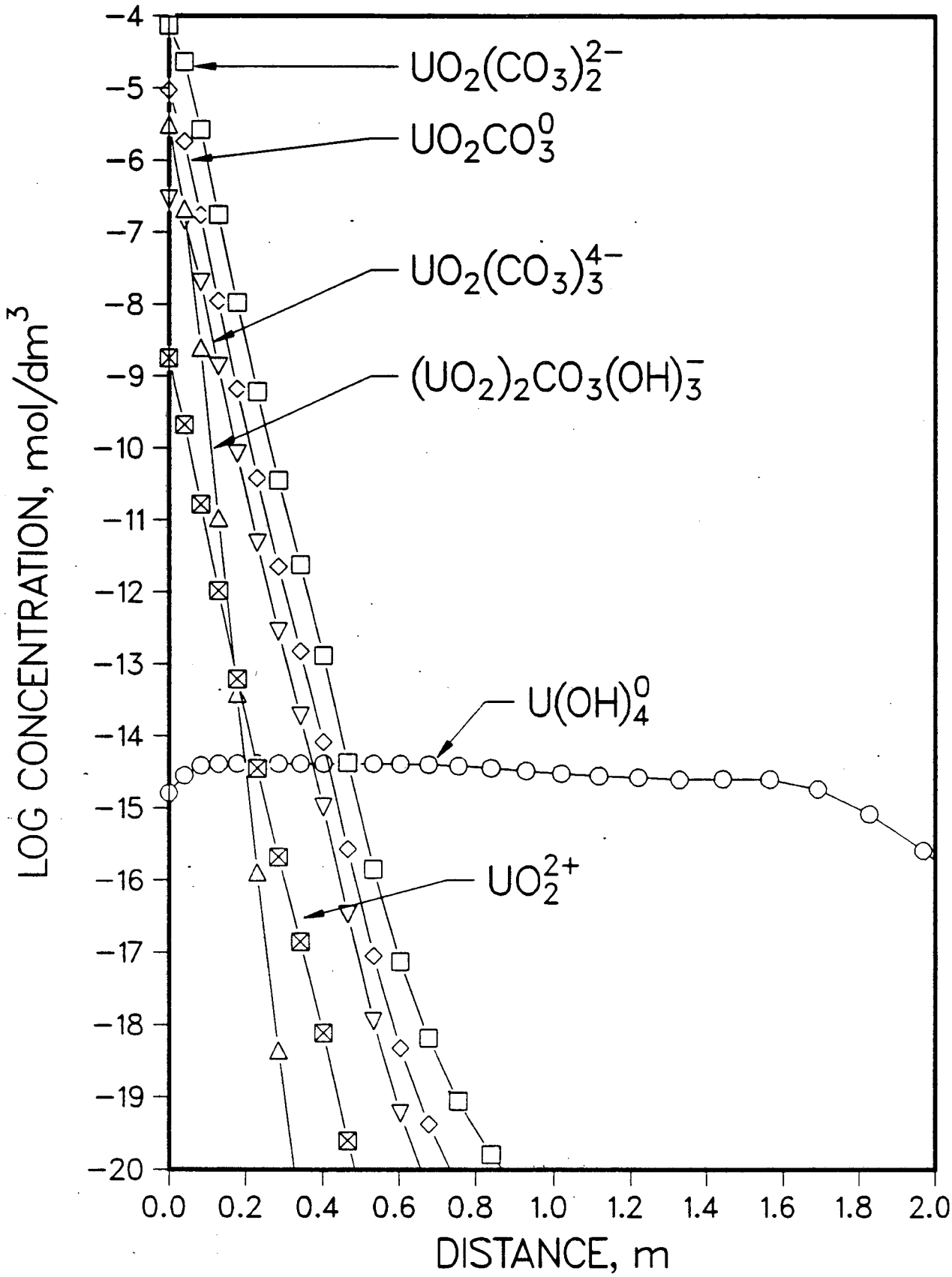

$UO_2(CO_3)_2^{2-}$
$UO_2CO_3^0$
$UO_2(CO_3)_3^{4-}$
$(UO_2)_2CO_3(OH)_3^-$
$U(OH)_4^0$
$UO_2^{2+}$
LOG CONCENTRATION, mol/dm$^3$
DISTANCE, m
−4
−5
−6
−7
−8
−9
−10
−11
−12
−13
−14
−15
−16
−17
−18
−19
−20
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0

FIGURE 7

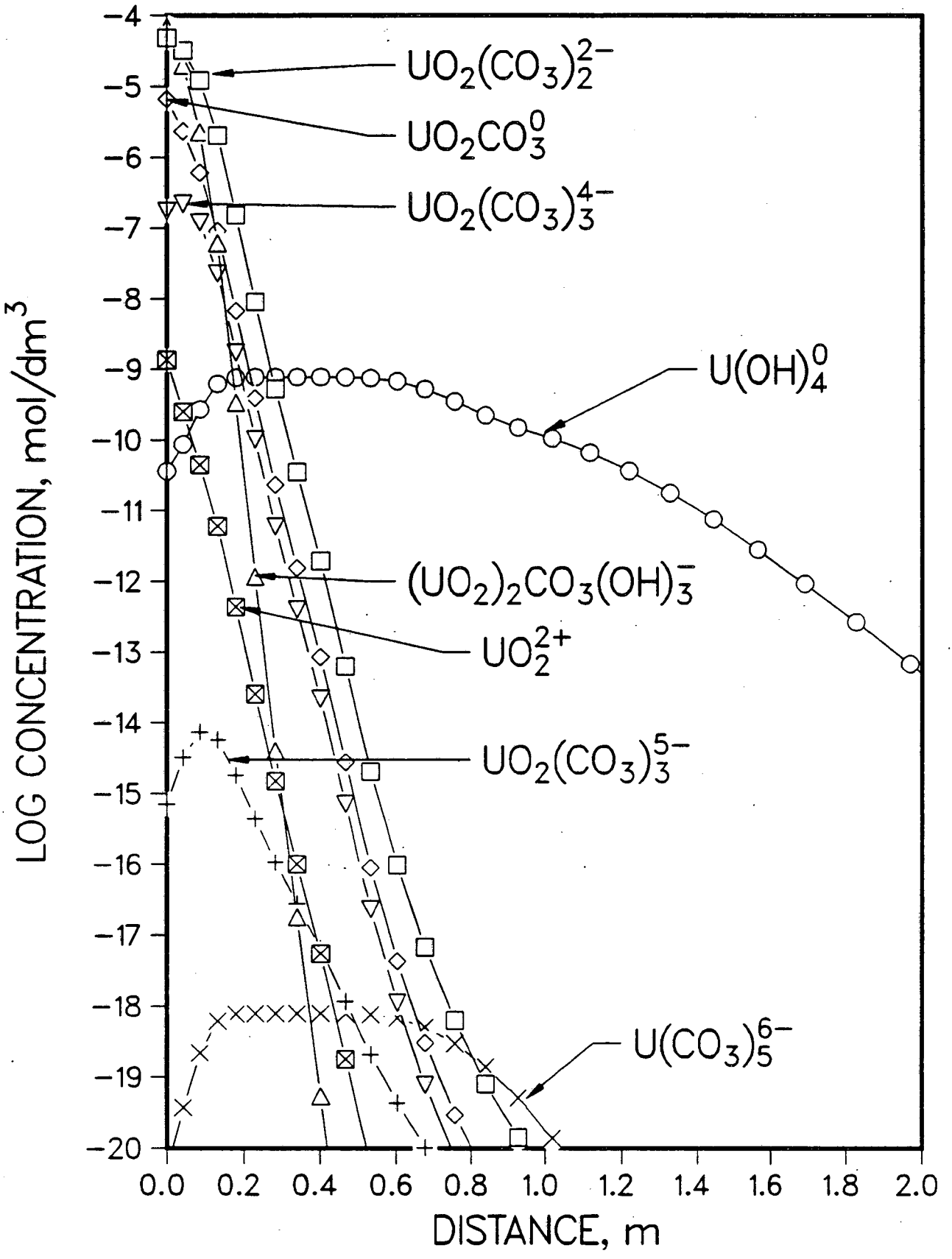

$UO_2(CO_3)_2^{2-}$
$UO_2CO_3^0$
$UO_2(CO_3)_3^{4-}$
$U(OH)_4^0$
$(UO_2)_2CO_3(OH)_3^-$
$UO_2^{2+}$
$UO_2(CO_3)_3^{5-}$
$U(CO_3)_5^{6-}$
LOG CONCENTRATION, mol/dm$^3$
DISTANCE, m
−4
−5
−6
−7
−8
−9
−10
−11
−12
−13
−14
−15
−16
−17
−18
−19
−20
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0

FIGURE 8

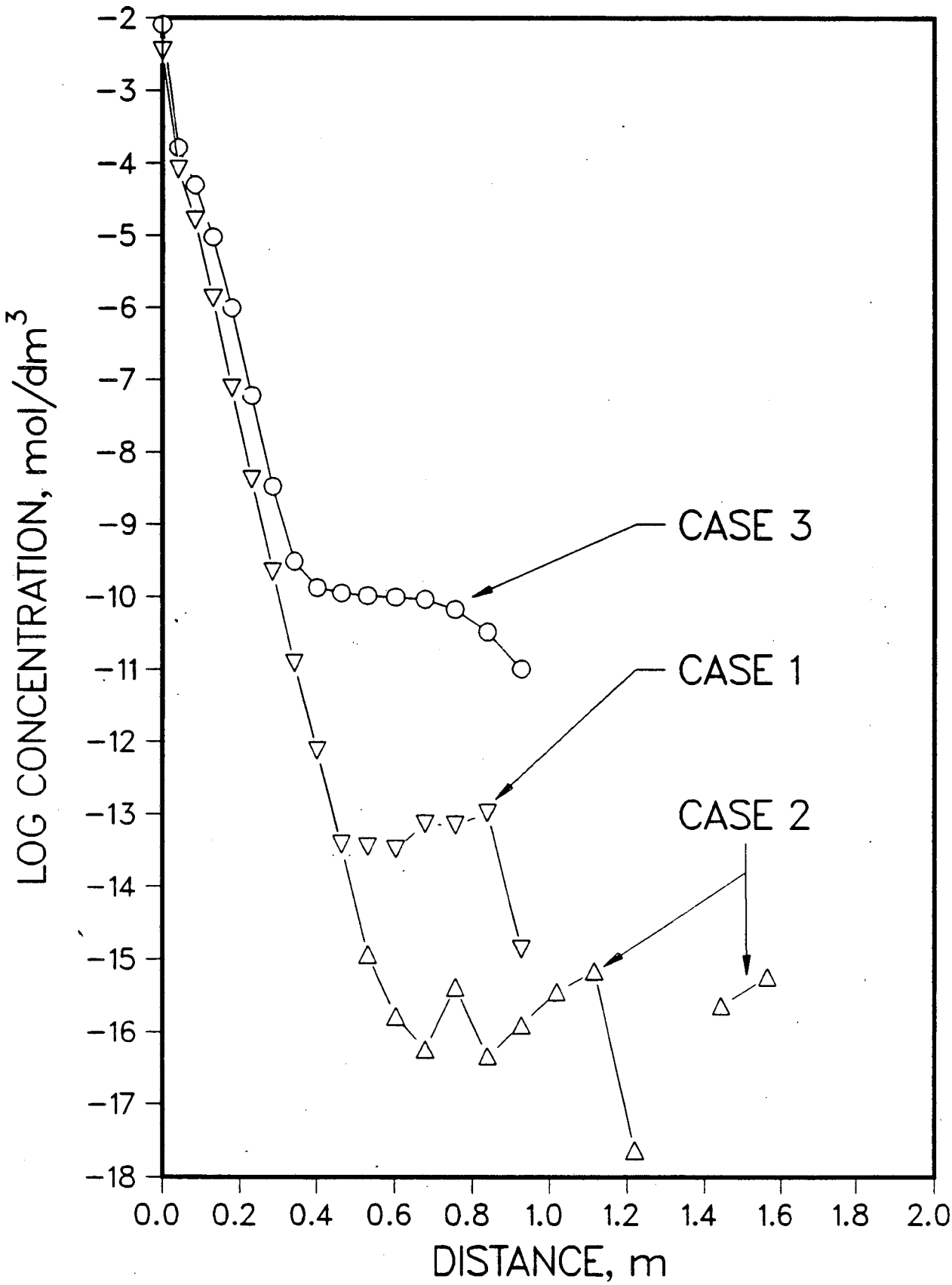
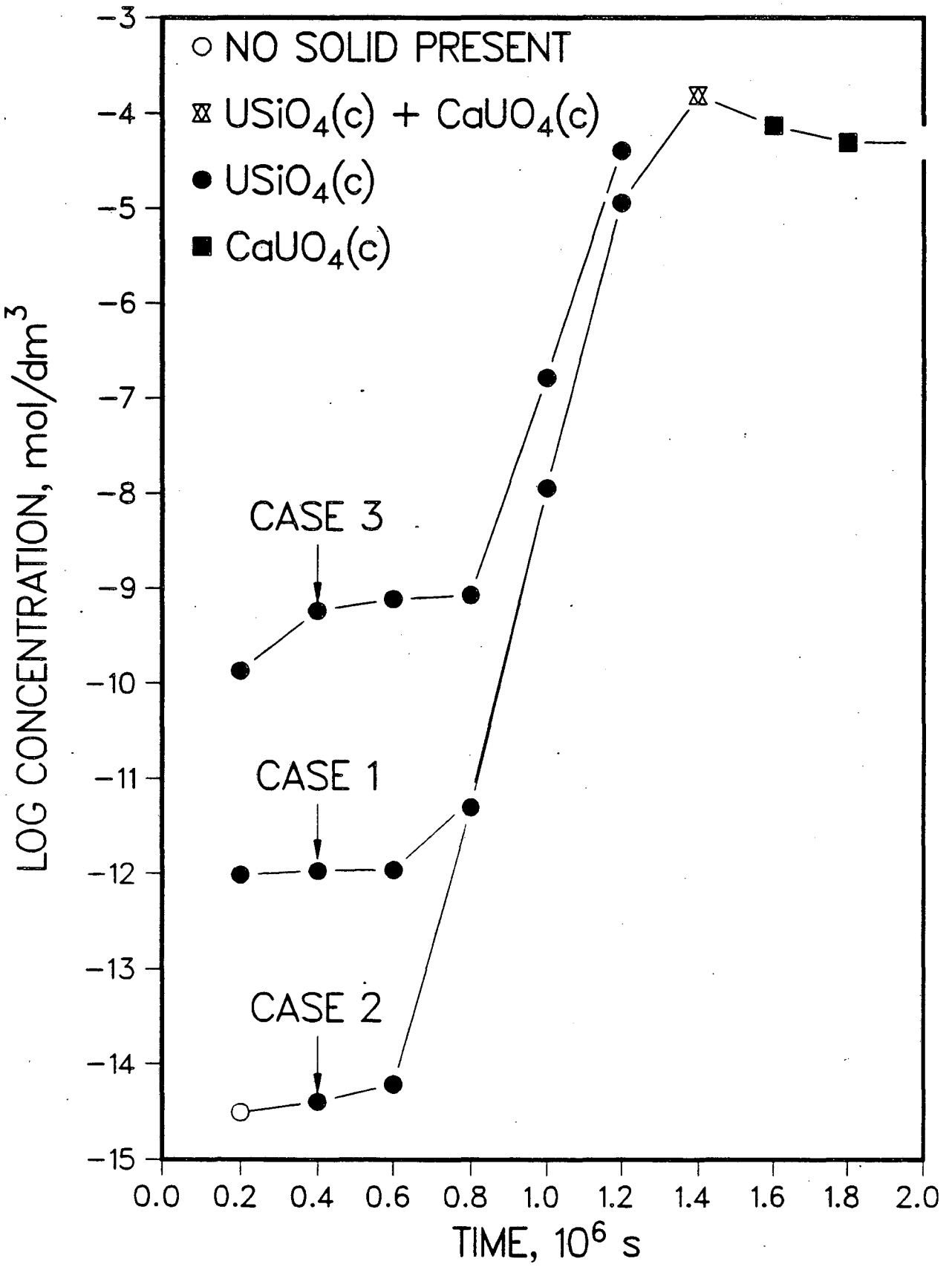

LOG CONCENTRATION, mol/dm3
DISTANCE, m
CASE 3
CASE 1
CASE 2
−2
−3
−4
−5
−6
−7
−8
−9
−10
−11
−12
−13
−14
−15
−16
−17
−18
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0

FIGURE 9


NO SOLID PRESENT
USiO4(c) + CaUO4(c)
USiO4(c)
CaUO4(c)
CASE 3
CASE 1
CASE 2
LOG CONCENTRATION, mol/dm3
TIME, 10^6 s
-3
-4
-5
-6
-7
-8
-9
-10
-11
-12
-13
-14
-15
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0

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