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Authors
Phillips, S.L.
Silvester, L.F.

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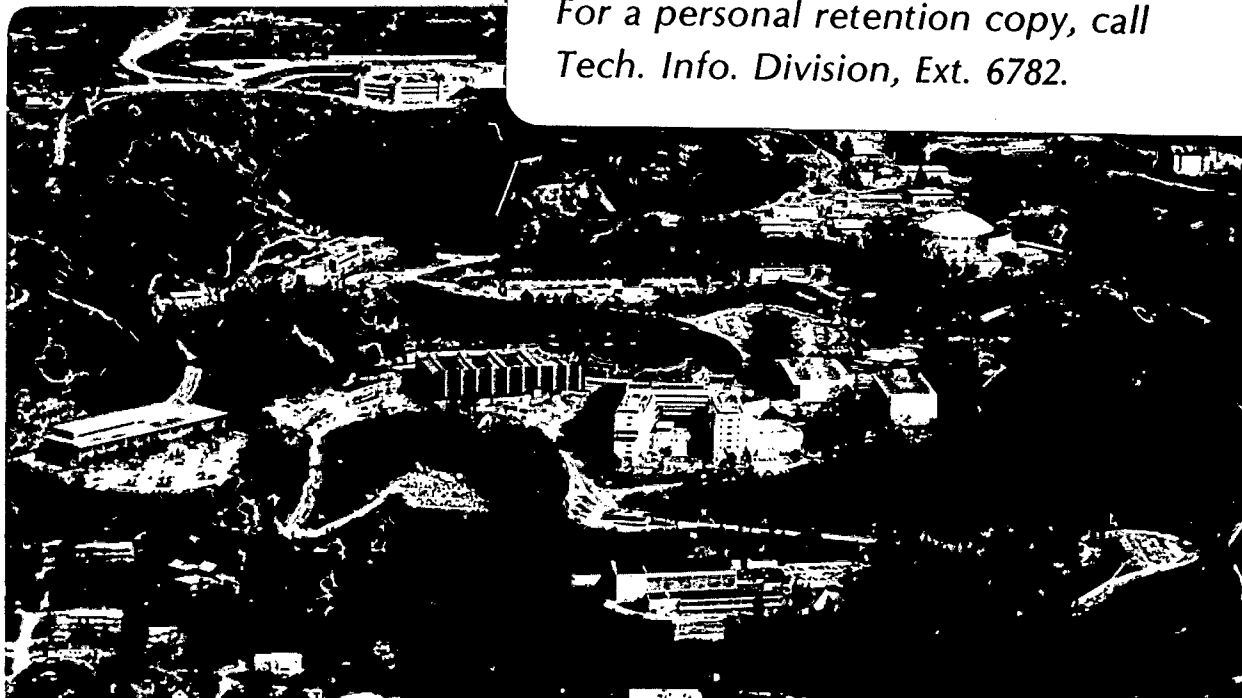
A DATABASE FOR NUCLEAR WASTE DISPOSAL FOR
TEMPERATURES UP TO 300°C

Sidney L. Phillips and Lenard F. Silvester

September 1982

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Sidney L. Phillips
and
Lenard F. Silvester

Engineering & Technical Services Division
Lawrence Berkeley Laboratory
University of California
Berkeley, CA 94720

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ABSTRACT

A computerized database is compiled of evaluated thermodynamic
data for aqueous species associated with nuclear waste storage. The
data are organized as hydrolysis and formation constants; solubilities
of oxides and hydroxides; and, as Nernstian potentials. More emphasis
is on stability constants. Coefficients are given to calculate sta-
bility constants at various ionic strengths and to high temperatures.
Results are presented as tables for selected species including uranium,
amorphous silica and actinides.

A computerized database is established to provide critically evaluated thermodynamic data for application to nuclear waste disposal. The data are organized into that for leaching studies, and that for migration. More emphasis is given to stability constants; solubility of oxides and hydroxides; and, to Nernstian potentials. However, this report is limited to hydrolysis and formation constants for selected waste species such as uranium, and to the solubility of amorphous silica.

Introduction

The past three decades have seen great progress in developing new sources of energy to heat our homes, light our cities, and for manufacturing. Nuclear power is an example. Now there is increased research in disposal of wastes from nuclear power production using modern scientific and engineering methodology. Underground isolation is preferred.

Central to underground isolation is the estimation of leaching of species from a glass or ceramic solid containing the wastes by underground waters, followed by migration via natural waters. Predictions of nuclide leaching are based partly on accelerated laboratory tests, for example the study by Jantzen et al. (1982). Polyphase ceramic materials were used as the solid waste; leaching solutions were deionized water (pH 7.9 - 8.5), simulated brine (pH 5.5 - 6.0) and simulated silicate water (pH 8.0 - 9.0). Test conditions followed procedures of the Materials Characterization Center; 40°C, 90°C and 150°C temperatures were used. Results indicated that dissolution is controlled mainly by the solubilities of the individual phases of the various constituents of the ceramic materials; and, by heterogeneous chemical reactions at the solid-solution interface such as adsorption of metal hydroxy complexes. Similar evaluation is done on borosilicate glass. See, for example, Plodinec et al. (1982).

Computer codes such as EQ6/EQ3, PROTOCOL and SOLMNEQ are used to estimate the concentration of dissolved uranium or silica in groundwater. Goodwin (1982) calculated uranium solubilities on the

order of 10^{-10} M for a range of conditions representing a Canadian disposal site. Both Wolery (1980), and Paquette and Lemire (1981) have developed and utilized potential -pH diagrams for speciation and other studies of uranium in natural groundwaters. One result is the inclusion of UO_2^+ as a soluble species, another is that $UO_2(s)$ is probably the stable solid phase.

Current worldwide research for nuclear waste disposal includes leaching studies at Savannah River Laboratory, Rockwell International, Battelle Pacific Northwest, and Hahn-Meitner Institut. Migration studies include those at Sandia National Laboratory, Whiteshell Nuclear Research Establishment, and Los Alamos National Laboratory. Stability constants, solubility and other basic data are published by Lawrence Berkeley Laboratory, Oak Ridge National Laboratory, Argonne National Laboratory, Whiteshell Nuclear Research, U. S. Geological Survey, Pennsylvania State University, Catholic University, Florida State University, Chalmers University and Colorado School of Mines. Evaluated thermochemical data are published by Lawrence Berkeley Laboratory, National Bureau of Standards, U. S. Geological Survey and Whiteshell Nuclear Research. Finally, development of computer codes are at Lawrence Livermore National Laboratory, Battelle Pacific Northwest and Whiteshell Nuclear Research Establishment. Jenne (1981) has critically surveyed over 10 computer models with possible application to nuclear waste isolation. Included in his survey are the widely used WATEQ codes; these are a virtual benchmark for computer-assisted computer calculations. A recent model called PROTOCOL is being developed by Jackson (1982) to predict dissolution reactions of solidified nuclear wastes.

In this report, more detail is given to estimating stability constants, acid ionization and solubility to high concentrations and temperatures. Examples are given for silicic acid ionization to 300°C; stability constants of selected uranium and actinide species; and, to the solubility of amorphous silica. Refer for detailed information on nuclear waste species to Phillips (1982) for about 20 metal species at 25°C and ionic strengths from 0 to 3; Lemire and Tremaine (1980) for uranium and plutonium; Wolery (1982) for uranium and other species

important to migration; and, Jackson (1982) for species important to leaching. More generally, Cobble et al. (1982) have published a report covering thermodynamic properties of aqueous species important to nuclear reactor chemistry; some data useful to nuclear waste disposal are included.

Stability Constants at 25°C

Reliable ionization, formation and hydrolysis constants at 25°C and zero ionic strength are used to estimate stability constants at high concentrations and temperatures. These data are useful in predicting likely species in leachants and in natural waters; together with other data, e.g., solubility and Nernstian potentials. Hydrolysis of UO_2^{2+}, complexation of UO_2^{2+} with CO_3^{2-}, and ionization of silicic acid typify stability data.

Table 1 lists selected hydrolysis and formation constants for UO_2^{2+} together with coefficients for the following modified Debye-Huckel equation:

$$\log K = \log K° + \frac{A \, \Delta Z^2 \, I^{1/2}}{I + BI^{1/2}} + CI \qquad (1)$$

where

K = apparent stability constant

$K°$ = stability constant at zero ionic strength

A = 0.5115 at 25°C

ΔZ^2 = sum of squares of ionic charge of products of hydrolysis or formation reaction minus sum of the squares of charges of reactants

I = ionic strength

B = constant

C = constant

From eq(1) and the values given in Table 1, apparent stability constants for UO_2^{2+} are calculated over the range of ionic strength, $0 < I < 3$; see Tables 2 and 3. Baes and Mesmer (1976) cite -5.8 for UO_2OH^+, $\log K°_{11}$; Dongarra and Langmuir (1980) calculate -5.1.

3

We calculate -5.3 using the Davies equation from log K_{11} = -5.5 at I = 0.1 (Phillips 1982). Langmuir found -5.1 gave better values in calculating log K for UO_2^{2+} phosphate complex than -5.7; also, our estimated constant is closer to -5.1. Accordingly, the best value is -5.3. Figure 1 plots log K versus ionic strength for the following three values of log K°: -5.1, -5.3, -5.8. The B and C coefficients are taken from Turner et al.(1981). The effect of the change in log K° from -5.3 is to lower or raise the log K values, while the shape of the curve is not significantly changed.

Similarly, we calculate for log K_{22}^o -5.68 which compares with -5.54 found by Vainiotalo and Makitie (1981) and compares with -5.62 in Baes and Mesmer (1976) for $(UO_2)_2(OH)_2^{2+}$; log K_{35}^o = -15.43 for $(UO_2)_3(OH)_5^+$ using the Davies equation compared with -15.63 in Baes and Mesmer; and -15.64 calculated from their measurements by Vainiotalo and Makitie. We select for $UO_2CO_3(aq)$ log $K°_{11}$ = 9.9 calculated from Langmuir's data and in better agreement with 10.1 from Lemire and Tremaine (1980), and 9.16 from Ciavatta et al. (1979), than 7.50 from Turner et al. (1981). The log K_{12}^o and log K_{13}^o values selected here (16.7, 21.4) agree well with published data (Phillips 1982). The B and C coefficients are mostly from Turner et al.

The first ionization constant for silicic acid in Table 1 is from the careful measurements by Busey and Mesmer (1977). We calculate -9.41 at I = 0.6, agreeing with -9.472 calculated by Sjoberg et al. (1981) to within 0.6%. Log K_{12}^o of -22.92 was adopted from Baes and Mesmer; this second ionization constant changes only by about 4% over the range 0 < I < 3. The B and C coefficients are from Baes and Mesmer.

Additional data on hydrolysis and formation constants for nuclear waste applications are found in a number of publications, including Phillips (1982), Lemire and Tremaine (1980), Rai et al. (1980), and Langmuir (1978).

Stability Constants to 300°C

Calculational methods are needed for estimating thermochemical data at high temperatures where data are scarce, using the values at other temperatures such as 25°C where data are more plentiful. The Criss-Cobble (1964) and Helgeson (1969) approaches are widely used. Other procedures include empirical fitting to polynomials, use of the integrated van't Hoff equation, and estimation of heat capacity (see Cobble et al., 1982). We have used a general equilibrium equation derived by Clarke and Glew (1966) for estimating stability constants up to 300°C using enthalpy and entropy data published at 25°C.

By definition

$$R \ln K = \Delta S°_T - \frac{\Delta H_T°}{T} \tag{2}$$

$$\Delta H_T° = \Delta H°_{298.15} + \int_\Theta^T \Delta C_p \, dT \tag{3}$$

$$\Delta S_T° = \Delta S°_{298.15} + \int_\Theta^T \frac{\Delta C_p}{T} \, dT \tag{4}$$

From Clark and Glew (1966) we assume both $\Delta S°_T$ and ΔCp can be expanded in a Taylor's series with 298.15 K as the reference temperature to obtain eq(5) in which $\Theta = 298.15$ K.

$$R \ln K = A + \frac{B}{T} + C \ln T + D T + ET^2 + FT^3 \tag{5}$$

where

$$A = \Delta S°_\Theta - (C_0 - \frac{\Theta^2}{4} C_2 + \frac{10\Theta^3}{72} C_3) - \ln \Theta$$
$$(C_0 - \Theta C_1 + \frac{\Theta^2}{2} C_2 - \frac{\Theta^3}{6} C_3) \tag{6}$$

$$B = -\Delta H°_\Theta + \Theta C_0 - \frac{\Theta^2}{2} C_1 + \frac{\Theta^3}{6} C_2 - \frac{\Theta^4}{24} C_3 \tag{7}$$

$$C = C_0 - \Theta C_1 + \frac{\Theta^2}{2} C_2 - \frac{\Theta^3}{6} C_3 \tag{8}$$

$$D = \frac{C_1}{2} - \frac{\theta}{2} C_2 + \frac{\theta^2}{4} C_3 \qquad (9)$$

$$E = \frac{C_2}{12} - \frac{\theta}{12} C_3 \qquad (10)$$

$$F = \frac{C_3}{72} \qquad (11)$$

and $C_0 = \Delta C_{p\theta}$, $C_1 = (d \Delta C_p/dT)_\theta$, $C_2 = (d^2 \Delta C_p/dT^2)_\theta$, $C_3 = (d^3 \Delta C_p/dT^3)_\theta$. For a constant heat capacity over the temperature range considered, eq(5) becomes:

$$R \ln K = \Delta S_\theta^\circ - \frac{\Delta H_\theta^\circ}{T} + \Delta C_p \left(\frac{\theta}{T} - 1 + \ln \frac{T}{\theta} \right) \qquad (5a)$$

Table 4 consists of coefficients for eq(5) for estimating thermochemical constants up to 300°C for the reactions shown. Enthalpy and entropy values of the reactions at 25°C are calculated from the fit to eq(5a).Coefficients D, E and F are zero, except where noted.

We have fit data for the chemical equations shown in Table 4 to eq(5), except for $Si(OH)_4$ and $SiO(OH)_3^-$ ionizations: these are from Busey and Mesmer (1977) and Truesdell and Jones(1974). The first ionization of silicic acid has an almost parabolic shape from 0-350°. Extrapolation to predict log K values at higher temperatures only yields limited accuracy. The accuracy is improved by writing the chemical reaction as a neutralization, then subtracting the log K_w values for water. See Table 5.

Fleming and Crerar (1982) found that the neutralization of silicic acid: $H_4SiO_4 + OH^- = H_3SiO_4^- + H_2O$ is represented up to 350°C by the relation:

$$\log K = -0.6496 + \frac{1479}{T(K)} \qquad (12)$$

We calculate from eq(12) the following values of log K at the temperatures given: 25°C, 4.31; 50°C, 3.93; 100°C, 3.31; 200°C, 2.48; 300°C, 1.93. Adding the dissociation constants of water at each temperature (Marshall and Franck, 1981) gives the following values for the ionization

of silicic acid: 25°C, -9.69; 50°C, -9.35; 100°C, -8.96; 200°C, -8.81; 300°C, -9.48. The calculated values agree within an average of 1% with those calculated from the correlation developed by Busey and Mesmer (1977). Figure 2 compares the fit for the neutralization of silicic acid using eq(5) and the three coefficients from Table 4 (Busey and Mesmer, 1977) with our approximate fit: $\log K° = -0.130 + 1240.60/T(K)$. Figure 3 shows a similar plot for $\log K°$ vs. T^{-1}. Differences exceed about 1% for temperatures beyond about 300°C.

From A and B we find enthalpy (ΔH_r) and entropy (ΔS_r) using data from Choppin and Unrein as follows for I = 1.0M (ClO_4), 25°C:

Reaction	ΔH_r, cal/mol	ΔS_r, cal/mol/deg.
$Am^{3+} + F^- = AmF^{2+}$	2933	21.3
$Cm^{3+} + F^- = CmF^{2+}$	2825	21.4
$Th^{4+} + HF = ThF^{3+} + H^+$	-2615	11.9
$Np^{4+} + HF = NpF^{3+} + H^+$	-2551	12.5
$U^{4+} + HF = UF^{3+} + H^+$	-2606	12.1

Enthalpy and entropy values for AmF^{2+} and CmF^{2+} are lower than those of Choppin and Unrein. However, our values of A and B in Table 4 from which we computed ΔH_r and ΔS_r give values of log K that agree within the uncertainty of their experiment for AmF^{2+}:10°C, 2.38(2.39);25°C, 2.49(2.49);40°C, 2.59 (2.57);55°C, 2.69(2.71). On the other hand their values of ΔH_r and ΔS_r yield A=7.32 and B=-1422.3 with log K at I=1 of:10°C, 2.23;25°C, 2.48;40°C, 2.71; 55°C, 2.92. We find a similar difference for CmF^{2+}. Table 6 gives log K at I=1 for these reactions from a fit to eq(5); C, D, E and F are equal to zero.

Formation constants for the reaction $U^{4+} + F^- = UF^{3+}$ were calculated at I = 0 from the data in Choppin and Unrein (1976) using eq(1) with coefficients taken from Phillips (1982); and log K° values for dissociation of HF to 200°C from Ellis (1963). The effect of ionic strength on log K for HF was calculated using the correlation in Phillips (1982). Corrections from I = 1 to I = 0 for the formation of UF^{3+} were made using the Debye-Huckel coefficients, A, for the following temperatures: 25°C (0.511), 40°C (0.524), 90°C (0.582). We assumed that the C coefficient did not change over this temperature range, and otherwise used the fit published by Turner et al. (1981).

We find the following for log K° at the temperatures shown: 8.95 (3°C); 8.97 (25°C); 8.99 (47°C). These were fit to eq(5), where A = 9.31, B = -101.46. Assuming the term for the heat capacity change is negligible, we estimate log K values as shown in the following table:

Log K° Values For The Reaction $U^{4+} + F^- = UF^{3+}$
At The Temperatures Indicated:

		log K°			
T,°C	Eq(5)	Lemire and Tremaine (1980)	Borin and Koneva (1981)	Langmuir (1978)	Turner et al. (1981)
25	8.97±.08	9±1	8.31	8.6	10.23
50	8.99±.09	-	-	-	-
60	9.01±.09	9±1	-	-	-
100	9.04±.12	9±1	-	-	-
150	9.07±.13	10±2	-	-	-
200	9.09±.15	11±2	-	-	-

The formation constant for UF^{3+} increases by only about 1% over this temperature range, and by about 20% according to Lemire and Tremaine (1980). See Figure 4. The above table also indicates that the best value for log K° at 25°C is 8.9. Using log K° = 8.97 and the A,B,C coefficients from Turner et al. (1981) in eq(1), we calculate at 25°C: log K = 7.73 for I=1; log K= 7.57 for I=2. These compare with log K = 7.34 at I=1 measured by Borin and Koneva (1981), and log K = 7.15 at I=2 as measured by Vdovenko at 20°C (1963). Log K values from eq(5) were calculated using uncertainties given by Choppin and Unrein (1976). We expect extrapolation from 60-200°C to have an increasing uncertainty, perhaps ±1 log K unit at 200°C.
Solubility

This section is limited to the solubility of amorphous silica in water and sodium nitrate solutions. From Fournier and Rowe (1977), the solubility of amorphous silica at the vapor pressure of the solution, from 0° to 250°C is given for water by

$$\log C = 4.52 - \frac{731}{T(K)} \tag{13}$$

where C = silica concentration, mg/Kg.

At a constant pressure of 1034 bars, the solubility from 0° to 380°C is calculated from

$$\log C = 4.82 - \frac{810}{T(K)} \tag{14}$$

Marshall (1980) has measured the solubility of amorphous silica in 0 - 6 molal $NaNO_3$ solutions over the temperature range 25-300°C. Fleming and Crerar (1982) recently correlated data on silica solubility as a function of both temperature and pH. We have fit both sets of data to the following equation which relates solubility, S, as a function of both temperature and $NaNO_3$ concentration

$$\log S = -0.320 + 0.00837m - \frac{(697.12 + 22.12m)}{T(K)} \tag{15}$$

where S is in units of molality.

Table 7 and Figure 5 show the change in solubility of amorphous silica up to 300°C and 5.98m concentrations. Solubilities calculated with eq(15) in water are compared with other data in the following table:

T°C	eq(15)	Fournier and Rowe,1977	Rimstidt and Barnes,1980*	Marshall,1980
25	0.00220	0.00195	0.00193	0.00218
50	.00333	.00301	.00304	.00346
100	.00648	.00606	.00619	.00676
150	.01078	.01031	.01044	.01088
200	.01609	.01571	.01546	.01568
250	.02226	.02209	.02087	.02108
300	.02909	--	.02631	.02698

*See Geochim.Cosmochim.Acta,1980, 44, 1683-1699.

Leaching experiments to predict the likelihood of removal of undesirable nuclear wastes such as actinides from solidified waste forms are done in deionized water, simulated brines and simulated silicate water. The data on solubility as a function of temperature and pH and $NaNO_3$ should be useful in predicting the solubility of amorphous silica in these solutions. However, research is needed to predict the effects of temperature, pH and indifferent electrolytes on silica solubility. Results to date indicate that the solubility increases with both temperature and pH, but decreases with increasing $NaNO_3$ concentrations.

The chemistry of silica including solubility in aqueous solutions is important to geochemistry. Research is active as attested by the large number of publications during the past 25 years. However, few publications deal with the chemistry of silica as applied to disposal of nuclear wastes. A systems approach is needed to permit computerized calculation of leaching from silica as a function of actinide concentrations, temperature, leachant composition, radiation effects, pH, cracks, solubility, and other parameters such as rates of solution and reprecipitation of colloidal particles by an adsorption mechanism. This is an area of active research (Jackson 1982, Nowak 1982, Plodinec et al. 1982). A complication in relating solubilities calculated from thermodynamic data to leaching studies is that equilibrium solubility may not be obtained under leaching test conditions.

Acknowledgment

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Table 1. Coefficients for eq(1) to estimate apparent stability constants for reactions shown, 25C

Reactions	logK(0)	AΔz2	B	C
$UO2^{2+} + H2O = UO2OH^+ + H^+$	-5.30	-1.022	2.32	0.3
$2UO2^{2+} + 2H2O = (UO2)2(OH)2^{2+} + 2H^+$	-5.54	-1.022	0.91	0.007
$3UO2^{2+} + 5H2O = (UO2)3(OH)5^+ + 5H^+$	-15.64	-3.054	1.90	0.014
$Si(OH)4 = SiO(OH)3^- + H^+$	-9.82	1.022	1.0	-0.06
$Si(OH)4 = SiO2(OH)2^{2-} + 2H^+$	-22.92	3.066	1.0	-0.39
$UO2^{2+} + CO3^{2-} = UO2CO3(aq)$	9.9	-4.09	1.63	0.05
$UO2^{2+} + 2CO3^{2-} = UO2(CO3)2^{2-}$	16.7	-4.09	1.61	-0.22
$UO2^{2+} + 3CO3^{2-} = UO2(CO3)3^{4-}$	21.4	0	--	-1.17

Table 2. Hydrolysis of $UO2^{2+}$ at 25 C

log K

I	$UO2OH^+$	$(UO2)2(OH)2^{2+}$	$(UO2)3(OH)5^+$
0.0	-5.30	-5.54	-15.61
.5	-5.42	-5.98	-16.52
1.0	-5.31	-6.07	-16.65
2.0	-5.04	-6.16	-16.75
3.0	-4.75	-6.21	-16.80

Table 3. Formation of $UO2^{2+}$ Carbonates at 25 C

log K

I	$UO2CO3(aq)$	$UO2(CO3)2^{2-}$	$UO2(CO3)3^{4-}$
0.0	9.90	16.70	21.40
.5	8.58	15.24	20.82
1.0	8.39	14.91	20.23
2.0	8.25	14.49	19.06
3.0	8.20	14.17	17.89

Table 4. Coefficients for eq(5) to estimate apparent stability constants for reactions shown

	Reaction	A	B	C
*1	$Si(OH)_4(aq)+OH^-=SiO(OH)_3^- +H_2O$	-18.4014	+2346.69	+2.57979
*2	$Si(OH)_4(aq)=SiO_2(OH)_2^{2-} +2H^+$	+39.4780	-12355.10	+0.00000
*3	$Am^{3+} +F^-=AmF^{2+}$	+4.6430	-640.90	+0.00000
*4	$Cm^{3+} +F^-=CmF^{2+}$	+4.6760	-617.40	+0.00000
*5	$Th^{4+} +HF=ThF^{3+} +H^+$	+2.6040	+571.40	+0.00000
25	$U^{4+} +HF=UF^{3+} +H^+$	+2.6460	+569.50	-6.41000
*6	$Np^{4+} +HF=NpF^{3+} +H^+$	+2.7430	+557.50	+0.00000
*7	$U^{4+} +H_2O=UOH^{3+} +H^+$	+7.5966	-2449.66	+0.00000
*8	$U^{4+} +2H_2O=U(OH)_2^{2+} +2H^+$	+9.8504	-3705.12	+0.00000
*9	$U^{4+} +3H_2O=U(OH)_3^+ +3H^+$	+12.9360	-5566.64	+0.00000
10	$U^{4+} +4H_2O=U(OH)_4(aq)+4H^+$	+10.3362	-6112.23	+0.00000
11	$U^{4+} +5H_2O=U(OH)_5^- +5H^+$	+5.0460	-5863.45	+0.00000
12	$Fe^{3+} +H_2O=FeOH^{2+} +H^+$	+12.2578	-2141.43	-1.27470
13	$Fe^{2+} +H_2O=FeOH^+ +H^+$	+9.4238	-3286.69	-3.12060
14	$Ni^{2+} +H_2O=NiOH^+ +H^+$	-1.1860	-2566.30	+0.00000
15	$Ni^{2+} +2H_2O=Ni(OH)_2(aq)+2H^+$	+.0430	-5665.87	+0.00000
16	$Co^{2+} +H_2O=CoOH^+ +H^+$	+.9376	-3178.07	+0.00000
17	$UO_2^{2+} +H_2O=UO_2OH^+ +H^+$	+2.1049	-2347.85	+0.00000
21	$UO_2^{2+} +2H_2O=UO_2(OH)_2(aq) +2H^+$	-1.3669	-3194.51	+0.00000
19	$2UO_2^{2+} +2H_2O=(UO_2)_2(OH)_2^{2+}+2H^+$	+58.9659	-4644.37	-19.79000
20	$3UO_2^{2+} +5H_2O=(UO_2)_3(OH)_5^+ +5H^+$	+138.6846	-11511.34	-46.75000
22	$UO_2^{2+} +CO_3^{2-}=UO_2CO_3(aq)$	+16.2083	-1889.14	+0.00000
23	$UO_2^{2+} +2CO_3^{2-}=UO_2(CO_3)_2^{2-}$	+20.8569	-1125.79	+0.00000
24	$UO_2^{2+} +3CO_3^{2-}=UO_2(CO_3)^{4-}$	+28.2075	-2321.27	+0.00000

NOTE: For reaction *2, D=-0.065927

Table 5. Ionization of Silicic Acid,25-300 C

log K°

T/C	log K11	log K12
25	-9.82	-21.62
50	-9.5	-20.06
100	-9.1	-18.23
200	-8.85	-17.83
300	-9.22	-19.86

$K11=[SiO(OH)3^-][H^+]/[Si(OH)4(aq)]$

$K12=[SiO2(OH)2^{2-}][H^+]^2/[Si(OH)4(aq)]$

Table 6. Formation of Selected Actinide Fluoro Complexes,I=1;25-300 C

log K

T/C	AmF^2+	CmF^2+	ThF^3+	NpF^3+	UF^3+
25	2.49	2.61	4.61	4.52	4.56
50	2.66	2.77	4.47	4.37	4.41
100	2.93	3.02	4.24	4.14	4.17
200	3.29	3.37	3.92	3.81	3.85
300	3.52	3.60	3.72	3.60	3.64

Reactions

$Am^{3+} +F^- = AmF^{2+}$
$Cm^{3+} +F^- = CmF^{2+}$
$Np^{4+} +HF = NpF^{3+} + H^+$
$Th^{4+} +HF = ThF^{3+} + H^+$
$U^{4+} +HF = UF^{3+} + H^+$
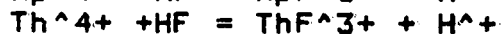
Table 7. Solubility of amorphous silica in NaNO3 solutions calculated from eq(15).

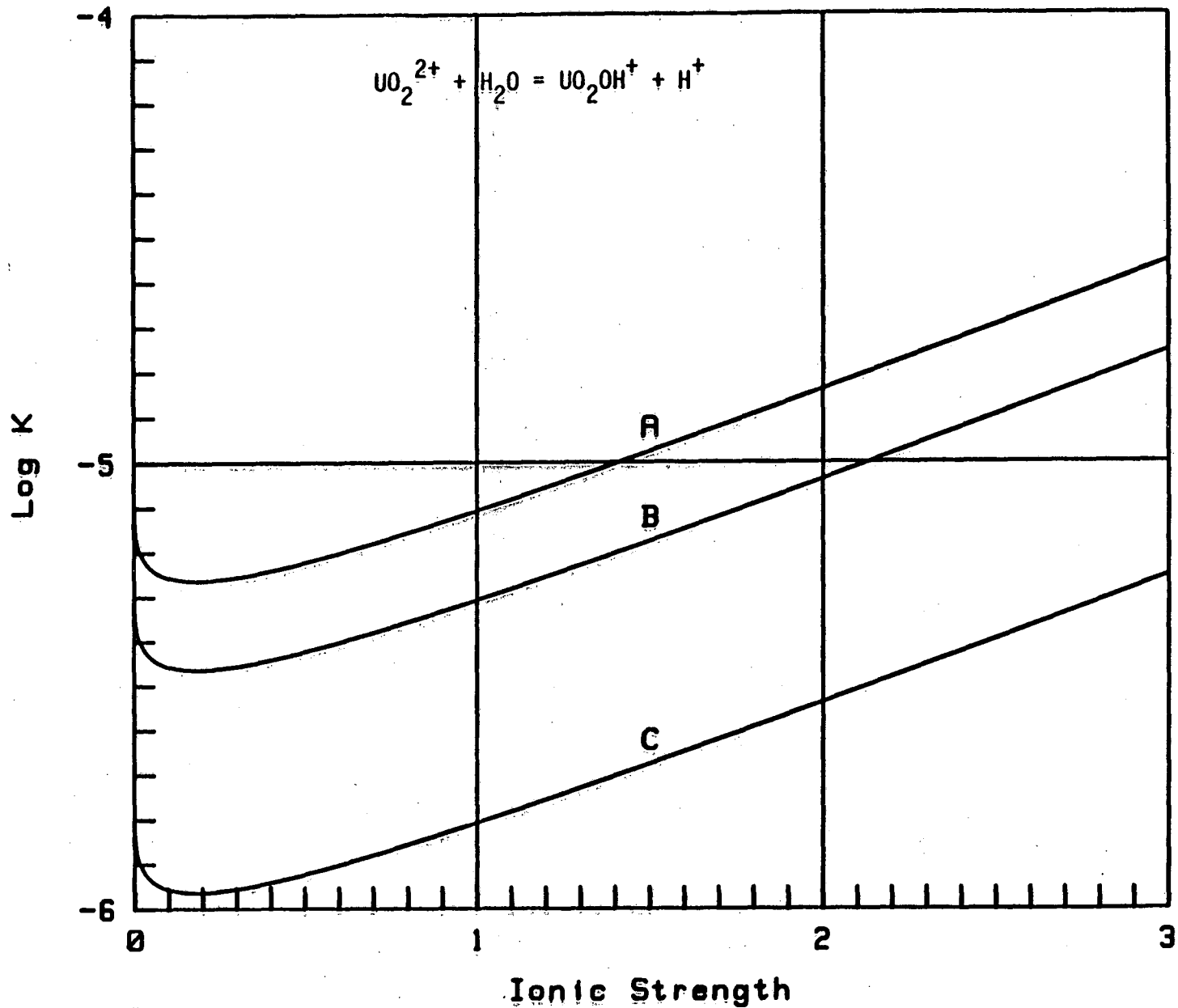| T/C	Molal solubility of SiO2 at mNaNO3 indicated 0m	.51m	2.12m	5.98m
25	.00220	.00203	.00159	.00089
50	.00333	.00311	.00249	.00146
100	.00648	.00611	.00506	.00322
150	.01078	.01024	.00870	.00589
200	.01609	.01538	.01334	.00949
250	.02226	.02139	.01886	.01395
300	.02909	.02807	.02510	.01919

Figure 1. Variation in log K with ionic strength for log K° values of:
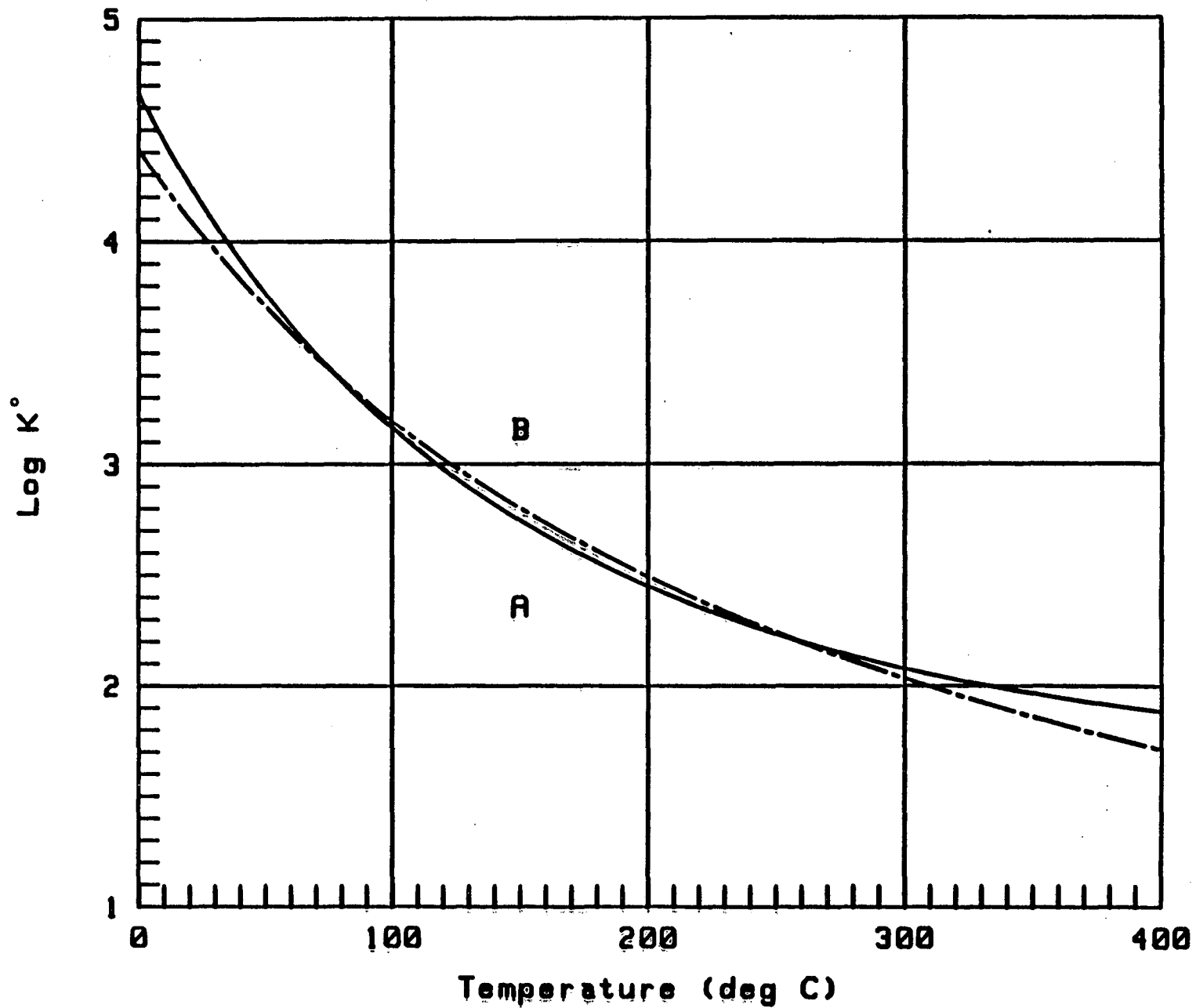A. -5.10, B. -5.30, C. -5.80.

Figure 2. Neutralization constant for silicic acid. A. Fit by Busey and Mesmer (1977), B. Fit to log $K° = -0.130 + 1240.60/T(K)$, This work.
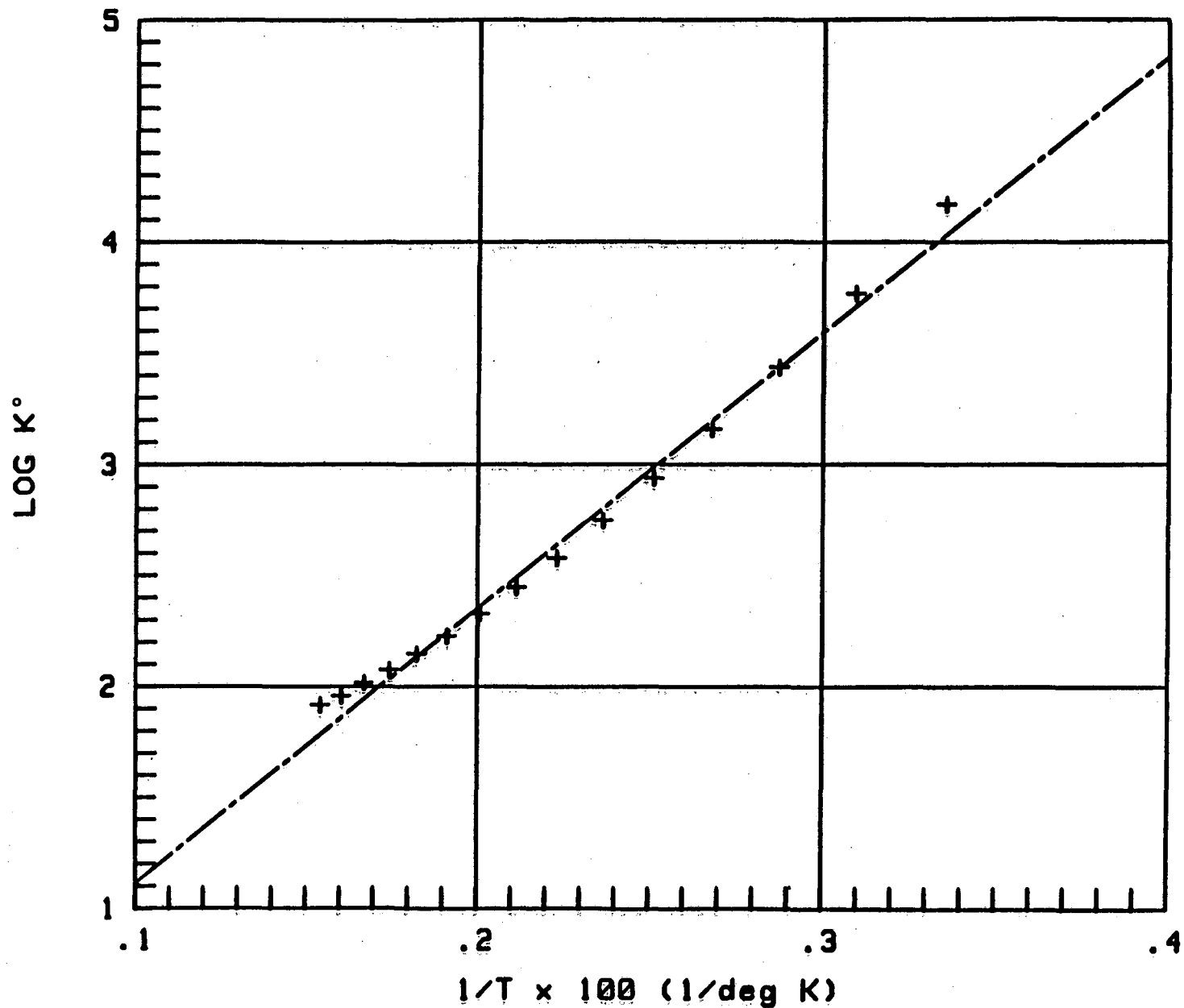
Figure 3. First neutralization constant of silicic acid. ++++ is from Busey and Mesmer (1977). ———·———·——— is fit to equation -0.130+1240.60/T(K).
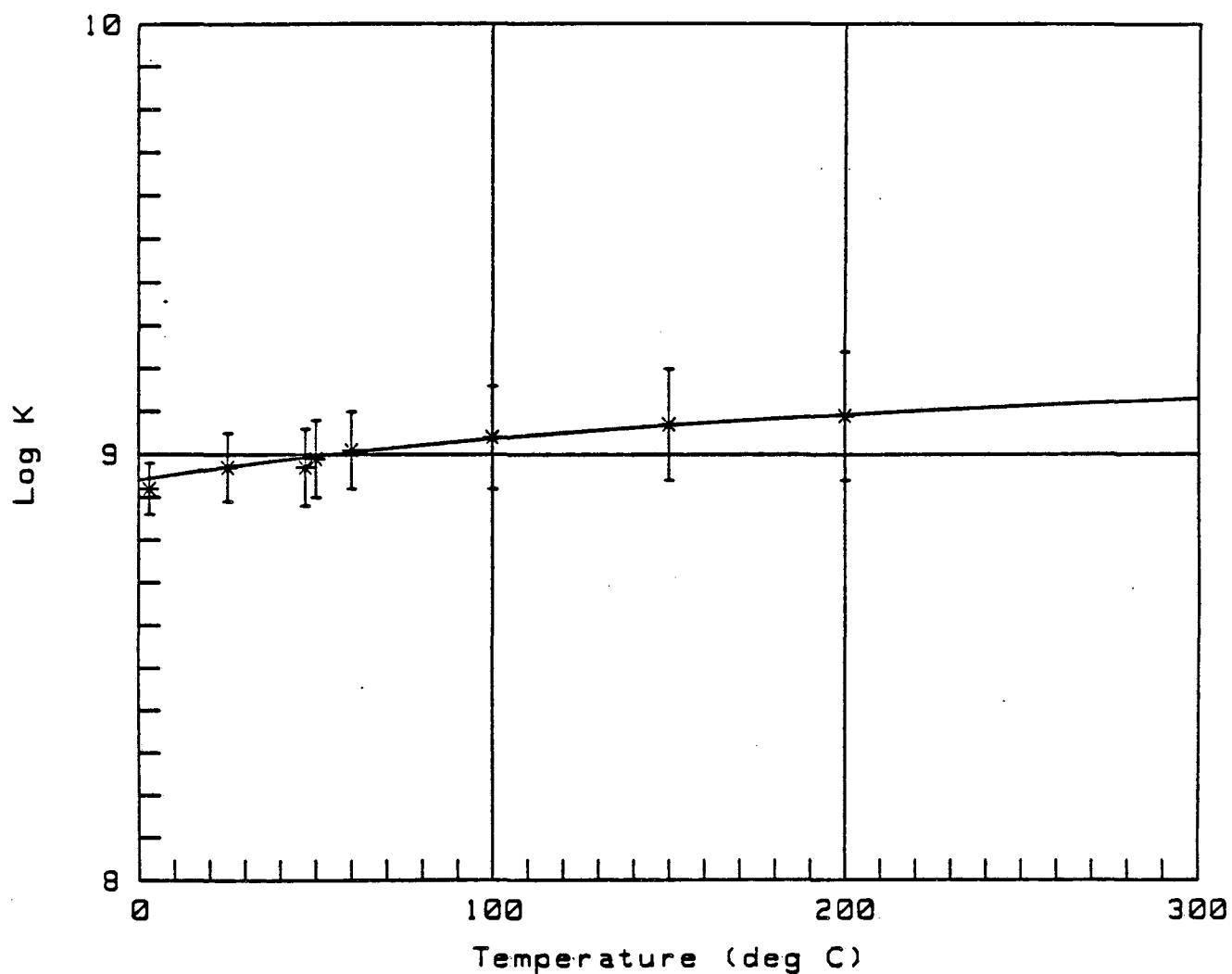
Figure 4. Variation in logK° with temperature for the reaction:
$U^{4+} + F^- = UF^{3+}$. Experimental points shown at 3°C and 47°C were
calculated from data published by Choppin and Unrein (1976).
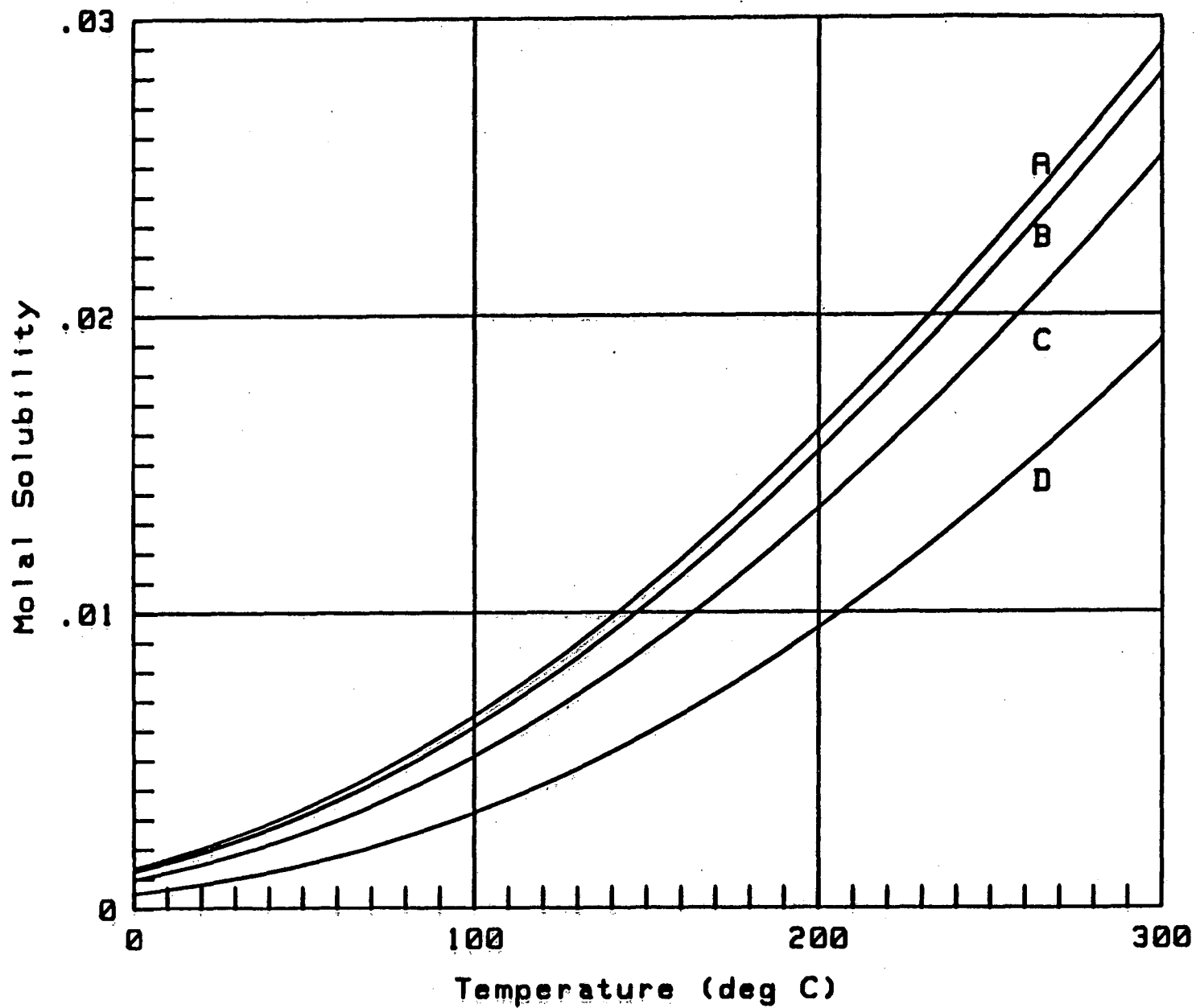
Figure 5. Solubility of amorphous silica calculated by eq (15). A. Water, B. 0.5m NaNO$_3$, C. 2m NaNO$_3$, D. 6m NaNO$_3$