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Modelling uranium solubilities in aqueous solutions: Validation of a thermodynamic data base for the EQ3/6 geochemical codes

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MODELLING URANIUM SOLUBILITIES IN AQUEOUS SOLUTIONS: VALIDATION OF A THERMODYNAMIC DATA BASE FOR THE EQ3/6 GEOCEHMICAL CODES

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ABSTRACT

Experimental solubilities of U^{4+} and UO_2^{2+} that are reported in the literature have been collected. Data on oxides, hydroxides and carbonates have been selected for this work. They include results both at 25°C and at higher temperatures.

The literature data have been compared with calculated uranium solubilities obtained with the EQ3/6 geochemical modelling programs and an uranium thermodynamic data base selected for the Swedish nuclear waste management program.

This verification/validation exercise has shown that more experimental data is needed to determine the chemical composition of anionic uranyl hydroxo complexes as well as their equilibrium constants of formation. There is also a need for more solubility data on well characterised alkaline or alkaline-earth uranates.

For the uranyl carbonate system, the calculated results agree reasonably well with the experimental literature values, which span over a wide range of pH, $[CO_3^{2-}]_T$, CO_2(g)-pressure, and T.

The experimental solubility of UO_2(s) agrees also well with the EQ3/6 calculations for pH>6. However, in more acidic solutions the experimental solubilities are higher than the calculated values. This is due to the formation of polynuclear hydroxo complexes of uranium(IV), which are not well characterised, and are not included in the thermodynamic data base used in this study.

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1 INTRODUCTION

This document reports a comparison beteen experimental uranium solubilities found in the literature, and the results of calculations obtained with a combination of a geochemical computer program (EQ3/6) and the thermodynamic data base given in the Appendices. An extract of this work has been presented elsewhere (Bruno and Puigdomenech, 1988).

The calculation of equilibrium radionuclide solubilities is of importance for nuclear waste disposal programs. This interest is due to,

- equilibrium models are used to gain understanding of the chemical behaviour of radionuclides under many different geological environments.
- in a situation of low groundwater flow within a repository, the source term might be limited by the solubility of the radionuclides at the prevailing physicochemical conditions (T, P, pH, Eh, etc).
- calculated solubility limits under different chemical conditions might be used to design a repository that through its engineered barriers keeps the lowest possible radionuclide concentrations in the waters that eventually might intrude into it.
- chemical equilibrium models can be used to estimate the prevailing geochemical processes affecting the mobility of naturally occurring radionuclides in candidate vault sites. Such an understanding will influence the selection of a repository site.

Because of these reasons, calculations of uranium(IV) oxide* solubility under several conditions have been reported in the literature (e.g., Goodwin, 1982; Kertes and

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* UO_2 is the main component in spent nuclear fuel.

Guillaumont, 1985; Bruton and Shaw, 1987). Another line of research has been to obtain reliable experimental data on radionuclide solubility under several chemical conditions (e.g., Forsyth et al, 1986; Parks and Pohl, 1988).

Agreement between model solubilities and experimental results is necessary in order to have any confidence in the long time predictions of the chemical behaviour of radionuclides in a geological environment. Therefore some comparison between model calculations and laboratory data have been reported (e.g., Kertes and Guillaumont, 1985; Pryke and Rees, 1986; and in the validation of WATEQ4 by Krupka et al, 1983).

The development of workable thermodynamic data bases for safety assessment within the Swedish nuclear waste management program has created a need for similar validation exercises.

1.1 A URANIUM DATA BASE FOR THE SWEDISH NUCLEAR WASTE PROGRAM

The modelling efforts by the different research groups involved in the Swedish program for disposal of spent nuclear fuel, have been made by using different sets of chemical equilibrium constants (compare e.g., Allard, 1983; with Grenthe et al, 1983). This reflected only minor discrepancies in the choice of equilibrium constants within the different reseach institutions. Nevertheless, we expect that future calculations for safety analysis will be more coordinated and will involve a unique set of thermodynamic data. The final goal is to adopt the recommendations arisen from the NEA-TDB data selection groups. In the meantime, there is a need for workable databases, hence, a selection of a thermodynamic data base has to be made for the chemical elements of radiological interest in the Swedish nuclear waste program.

This is a bookkeeping exercise, and a practical solution has been to use an existing data base as a starting material. The EQ3/6 data base has been selected. The choice is based in the following facts:

- the data base contains information on the most important minerals and aqueous species involved in systems with natural waters (about 630 minerals, 660 aqueous species, and 10 gaseous species)
- it also contains data on some of the radionuclides of interest (Th, U, Np, Eu, Pu, Am, Ru, Ra)
- the data base contains both a file with thermodynamic quantities (standard gibbs free energies of formation, standard enthalpies of formation, standard entropies, heat capacities, etc) and a file with equilibrium constants as a function of temperature (log K_{eq}(T) for T=0 to 300°C, and p=1.013 bar (~ 1 atm) up to 100°C and steam/$H_2O(l)$ equilibrium pressure at T>100°C)
- the EQ3/6 code package contains a computer program (MCRT) to calculate equilibrium constants of reaction as a function of temperature using thermodynamic quantities as starting material
- the EQ3/6 code package contains programs to maintain the large files involved in the data base
- the structure of the files that make up the data base includes the possibility to contain literature references, comments, quality description, etc

The main disadvantage with the EQ3/6 data base is that the computer programs that are used to maintain the files that constitute the data base are not documented. It is judged, however, that the advantages compensate for the disadvantage.

The uranium data base, which is validated in this report, is essentially that of Lemire and Tremaine (1980), with the changes introduced by Lemire (1988), and some minor modifications made by us (c.f Section 3). The uranium species included in the original database and not listed either in Section 3 or in the appendices (chloride, fluoride, phosphate and nitrate complexes, etc) were left unmodified.

It is intended at this time, that the data base, henceforth called in this report the SKBU1 data base, shall be used by the groups involved in the Swedish nuclear waste program as a primary data source whenever a need for thermodynamic data arises.

2 LITERATURE SOLUBILITY DATA USED IN THE VALIDATION

The solubility data that is found in the literature for the system UO_2-H_2O-$H_2(g)$-$CO_2(g)$ can be classified into three groups:

- solubility of U(IV) oxide/hydroxide as a function of pH, $[HClO_4]$, $[OH^-]_{tot}$ and T
- solubility of U(VI) hydroxides as a function of pH, $[HClO_4]$, [NaOH], $[CO_3^{2-}]_{tot}$ and T
- solubility of rutherfordine ($UO_2CO_3(c)$) as a function of p_{CO2}, pH, $[CO_3^{2-}]_{tot}$ and T

It seems that the solubility of oxides with other oxidation degrees (U_4O_9 and U_3O_8) has not been reported, except for the study of Gayer et al (1964) that reports the solubility of U_3O_8 in water. That study is however useless for our purposes because Gayer et al (1964) do not specify either the redox potential of the equilibrium aqueous solution, or the redox composition of the initial aqueous solution (partial presure of $O_2(g)$ and/or $H_2(g)$, concentration of residual oxygen, etc). Without a knowledge of the redox conditions, the solubility of U_3O_8 cannot be calculated.

We will give in this section a short introduction to the literature references used by us in this work. A further discussion of the accuracy, etc, is given later on in Sections 2.4, 4 and 5.

2.1 SOLUBILITIES OF U^{IV} OXIDE/HYDROXIDE

The solubility of uranium(IV) oxide and hydroxide depends on the crystallinity and particle size of the solid (Parks and Pohl, 1988; Bruno, 1988).

Therefore, in the SKBU1 data base we have included three oxides of U(IV): UO_2(am), UO_2(fuel) and uraninite, in order of decreasing solubility.

The data for UO_2(am) should reflect the properties of a hydrous and X-ray amorphous solid that is obtained by precipitation in alkaline aqueous solutions. In contrast, uraninite corresponds to well crystallized UO_2(c), while UO_2(fuel) is intended to be used for an intermediate solid (particle size 1-5 µm), which corresponds to the average particle size of UO_2 in spent fuel.

The measurements of the solubility of U(IV) oxides are complicated by oxidation of U(IV) to U(VI) hydroxo and carbonato complexes which is caused by the residual oxygen and carbonate that might be present in the initial solutions and by the residual oxygen present in the surface of the UO_2 solid phase.

The experimental difficulties and the varying solubility with particle size and crystallinity, is reflected in Figure 2.1, where some literature data from several authors and for ~25°C are shown. The difference between the highest and lowest reported solubility in Figure 2.1, is a factor of then thousand.

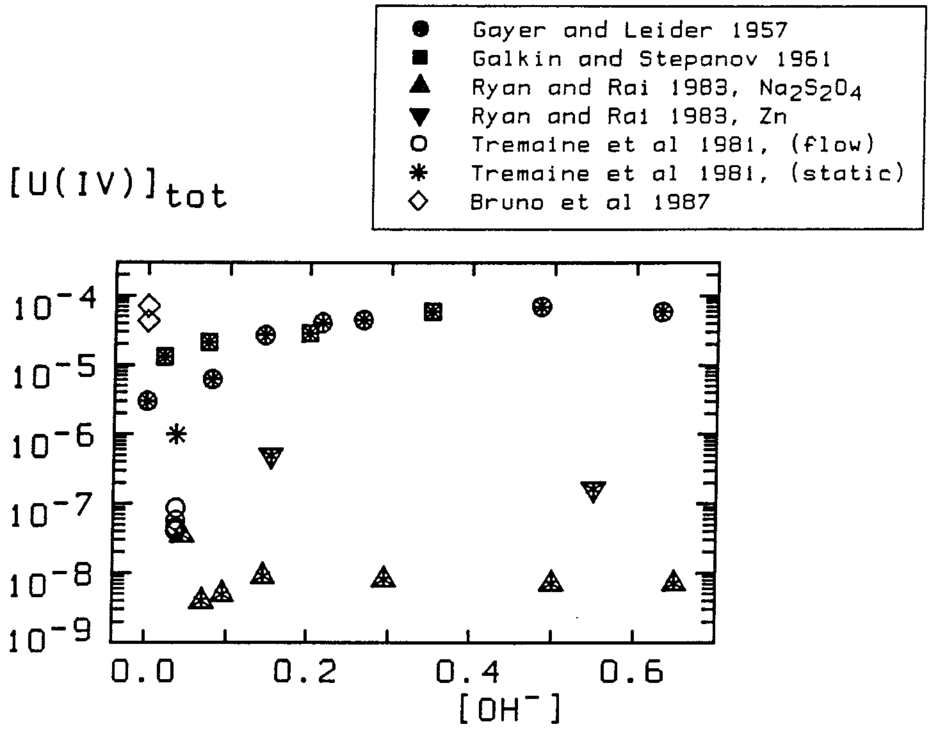

Figure 2.1 Experimental Solubility of Uranium(IV) oxides/hydroxides at about 25°C in alkaline aqueous solutions.

For the verification and validation of the UO_2(am) thermodynamic data, we have selected the data at 25°C by Nikolaeva and Pirozhkov (1978), Bruno et al (1987), Gayer and Leider (1957) and Galkin and Stepanov (1961).

The selected solubility data for uraninite (UO_2(c)), is that of Parks and Pohl (1988), Redkin and Omelyanenko (1987), Nguyen-Trung (1985), Nikolaeva and Pirozhkov (1978) and Tremaine et al (1981). The data covers the temperature range between 20 and 300°C and the pH range between 2 and 10.

2.2 SOLUBILITIES OF URANYL HYDROXIDES

The solid phase stable at 25°C is schoepite ($UO_2(OH)_2 \cdot H_2O$) while $UO_2(OH)_2$(c) is the stable form above ~60°C (Robins, 1966).

The measurements of the solubility of U(VI) hydroxides are complicated by the possible contamination with $CO_2(g)$ from the air, and by the formation of alkali uranates (at pH>10 at 25°C).

For the validation at 25°C, we have used the data of Gayer and Leider (1955), Miller(1958), Nikitin et al (1972), Silva and Yee (1981), Babko and Kodenskaya (1960) and Krupka et al (1985).

The data covers the pH range 3 to 12. Krupka et al (1985) is the only source of data above pH=10, because they used tetramethylammonium hydroxide as titrant to prevent the formation of insoluble uranates.

The different sets of solubility data for uranium(VI) hydroxide do not give a unique dependence of $[U(VI)]_{tot}$ versus pH (c.f. Figures 2-2 and 5.1). The data by Krupka et al (1985) shows a comparatively large spread, probably due to some shortcomings in the experimental technique used by Krupka et al (1985). All the solubility data at 25°C for U(VI) hydroxide used in this report, excluding, for the sake of clearness, the data of Krupka et al (1985) are shown in Figure 2.2.

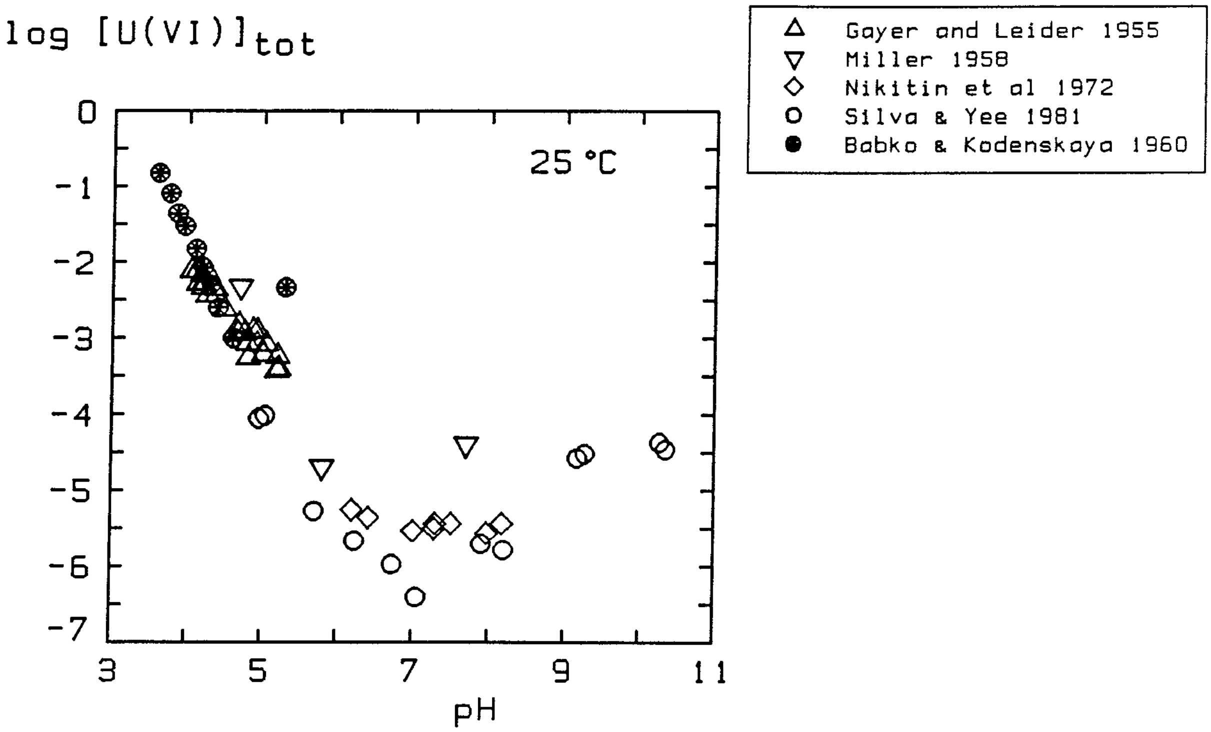

Figure 2.2 Some of the experimental literature data for the solubility of U(VI) hydroxide as a function of pH at 25°C.

Data presented by Brush (1980) (also reported in Holland and Brush (1978)) have been used for the validation of the U(VI) data base at 90°C. The data selected from Brush (1980), include experimental points without added NaCl and where the final solid was α-$UO_2(OH)_2$.

Data at temperatures higher than 25°C (other than Brush, 1980), have been obtained by Miller (1958) and Nikitin et al (1972). The data obtained by the various authors show some spread (up to a factor of 100 in the uranium concentration, c.f. Figure 5.6), the solubilities of Miller (1958) being the highest.

The solubility of schoepite in Na_2CO_3 solutions at 25°C that has been reported by Babko and Kodenskaya (1960) has also been used (c.f. Figure 5.4).

2.3 SOLUBILITY OF URANYL CARBONATE (RUTHERFORDINE)

The solubility of rutherfordine ($UO_2CO_3(c)$) has been investigated both as function of Na_2CO_3-concentration (Blake et al 1956), and as a function of pH at constant p_{CO2} (Sergeyeva et al 1972; Grenthe et al 1984). All data from these references, except that of Grenthe et al (1984) at 3.0 M $NaClO_4$, have been used in this study. The data at 3.0 M $NaClO_4$ of Grenthe et al (1984) were not selected because the method for activity factor corrections used in the EQ3/6 calculations fails at such high ionic strengths.

The data of Sergeyeva et al (1972) include the only available solubilities of rutherfordine at temperatures higher than 25°C.

2.4 THE pH VALUES AT HIGH TEMPERATURE

As it will be mentioned in section 4, in this report we try to compare calculated results in the molal scale (mols/1000 g H_2O) with experimental data that sometimes must be converted from molar (mols per litre) to molal units before the comparison.

For measured pH-values in dilute solutions at low temperature (~25°C), we have assumed that the correction from molar to molal units is negligible. In concentrated aqueous solutions at ~25°C we have converted the pH values from molar to molal units as discussed in section 4.

At higher temperatures a correction should also be made. This is because the density of aqueous solutions changes with temperature (for example, the density of water is 0.997 (g/cm^3) at 25°C and 0.96 at 100°C), and because protolitic equilibria in aqueous solutions are temperature dependent (e.g., a 10^{-6} m HCl solution has pH=6.0 at 25°C and pH=5.55 at 300°C). Furthermore, a pH measuring equipment calibrated at ~25°C will give erroneous results when used at other temperatures because of the temperature dependence of the Nerst equation.

The following references report uranium solubilities versus pH at temperatures higher than 25°C: Miller (1958), Nikitin et al (1972), Sergeyeva et al (1972), Nikolaeva and Pirozhkov (1978), Brush (1980), Nguyen-Trung (1985), and Parks and Pohl (1988).

Some of the experimental pH-values were measured at the temperature of the experiment (Nikolaeva and Pirozhkov 1978; Brush 1980; and the values at 50°C of Sergeyeva et al 1972 and Nikitin et al 1972). On the other hand, Nguyen-Trung (1985) reports pH values measured at 20°C. In some cases, however, it is not clear whether the pH values refer to high or low temperature measurements, and we assume that room temperature pH for quenched solutions is reported (Miller 1958; Sergeyeva et al 1972 at T>50°C).

Parks and Pohl (1988) give high temperature pH values of their experiments as calculated with EQ3NR from the low temperature pH and composition of their solutions.

Similarly, we have calculated (with the EQ3NR program) the high temperature pH for the lithium hydroxide solutions in the experiments of Tremaine et al (1981) and the values (that were used in Figures 5.14 to 5.16) varied from pH=12.45 at 25°C to pH=9.66 at 300°C.

Summarizing, we note that:

- the pH values at 90°C of Brush (1980) in Figure 5.5 were measured at 90°C by Brush (1980)
- the pH values at 50°C of Sergeyeva et al (1972) in Figure 5.10 were measured at 50°C by Sergeyeva et al (1972)
- the pH values att 100°C of Nikolaeva and Pirozhkov (1978) in Figure 5.14 were measured at 100°C

- the pH values at 100 and 200°C in Figures 5.14 and 5.15 for the solubilities of Parks and Pohl (1988) and Tremaine et al (1981) were calculated with the EQ3NR program by Parks and Pohl (1988) and us, respectively
- the pH values at 25°C in Figure 5.12 for the solubilities of UO_2(am) in NaOH solutions reported by Gayer and Leider (1957) and Galkin and Stepanov (1961), were also calculated by us with the EQ3NR program

3 THE THERMODYNAMIC DATA BASE: SKBU1

The SKBU1 thermodynamic data base consists of:

- the uranium data included in the EQ3/6 thermodynamic data base where the following aqueous species have been withdrawn:

  $U(OH)_5^-$
  $(UO_2)_2(CO_3)(OH)_3^-$
  $U_6(OH)_{15}^{9+}$

- the data listed in the Appendices. These data were replaced or added to the MDAS and DATA0 files of the EQ3/6 package

The data listed in the Appendices originate mainly from Lemire and Tremaine (1980). The modifications introduced by Lemire (1988) have also been included (except for the data on $U(OH)_5^-$) as well as the following changes:

a - $\Delta G_f°$ for U^{4+}, UO_2^+ and UO_2^{2+} was taken from the NEA-OCDE compilation (Grenthe et al 1988). We give here a comparison between the free energies of formation used by us and the data selected by Lemire and Tremaine (1980):

	NEA-TDB Grenthe et al 1988a	Lemire & Tremaine 1980
U^{4+}	-126.65 k cal/mol	-126.89 k cal/mol
UO_2^+	-229.59 "	-231.50 "
UO_2^{2+}	-227.67 "	-227.70 "

The differences between the two sets of data may be translated, for example, into the U^{4+}/UO_2^{2+} redox potentials for the reaction:

$$UO_2^{2+} + 4H+ + 2e^- = U^{4+} + 2H_2O$$

Using the $\Delta G_f°$ for water in the EQ3/6 data base (-56.686 k cal/mol), we obtain the following values for E° (= $59.16 \cdot (\frac{1}{2} \log K_{eq})$):

	NEA-TDB Grenthe et al 1988a	Lemire & Tremaine 1980
$U^{4+}/UO_2 2+$	267.8 mV	272.4 mV

The differences between the two data sets are not large (4.6 mV), and may be compared with values of E° reported by Bruno et al (1985): 260±3 and 248±3 mV (depending on the equations used for activity coefficient corrections).

b - $\Delta G_f°$ for UO_2(am) and UO_2(fuel) have been calculated from the equilibrium constants K_{s0} for the reaction

$$UO_2(s) + 4H^+ = U^{4+} + 2H_2O$$

K_{s0} was obtained by combining values of K_{s4} and K_4. The value of K_{s4} for the reactions

$$UO_2(s) + 2H_2O = U(OH)_4(aq)$$

have been determined by Bruno et al (1987) and (1988) for UO_2(am) and UO_2(fuel) respectively (we have assumed that the value of K_{s4}, which involves only electrically neutral species, is independent on the concentration of the ionic media). The value of K_4 for the reaction

$$U^{4+} + 4H_2O = U(OH)_4(aq) + 4H^+$$

is given by Lemire (1988). As previously mentioned, UO_2(fuel) corresponds to a uranium (IV) oxide with a particle size in the range of 1 to 5 µm.

c - $\Delta G_f°$ for the U(IV) hydrolysis complexes ($U(OH)_n^{4-n}$) has been calculated from the equilibrium constants for the reactions

$$U^{4+} + n\ H_2O = U(OH)_n^{4-n} + n\ H^+$$

The equilibrium constants were obtained as follows:

- $U(OH)^{3+}$: the experimental value ($\log K_1 = -0.51 \pm 0.03$) reported by Grenthe et al (1988b)

- $U(OH)_4$: the value reported by Lemire (1988) is $\log K_4 \doteq -5.28$

- $U(OH)_2^{2+}$ and $U(OH)_3^+$: estimated by interpolation between the equilibrium constants for $U(OH)^{3+}$ and $U(OH)_4$

A plot of the equilibrium constants versus T is shown in Figure 3.1.

d - S° for $U(OH)_4(aq)$ has been estimated with the "dquant" approximation (Helgeson, 1969, eqn. 16) for the reaction

$$U^{4+} + 4H_2O = U(OH)_4(aq) + 4\ H^+$$

in a procedure similar to that used by Lemire (1988). The "dquant" equation has been used because the MCRT program (of the EQ3/6 package) uses this equation when the aqueous complex formed is electrically neutral. The calculated equilibrium constant versus T is shown in Figure 3.1. This procedure differs from the one used by Lemire (1988) who assumed a value of zero for the increment in heat capacity for the reaction

$$UO_2(c) + 2H_2O = U(OH)_4(aq)$$

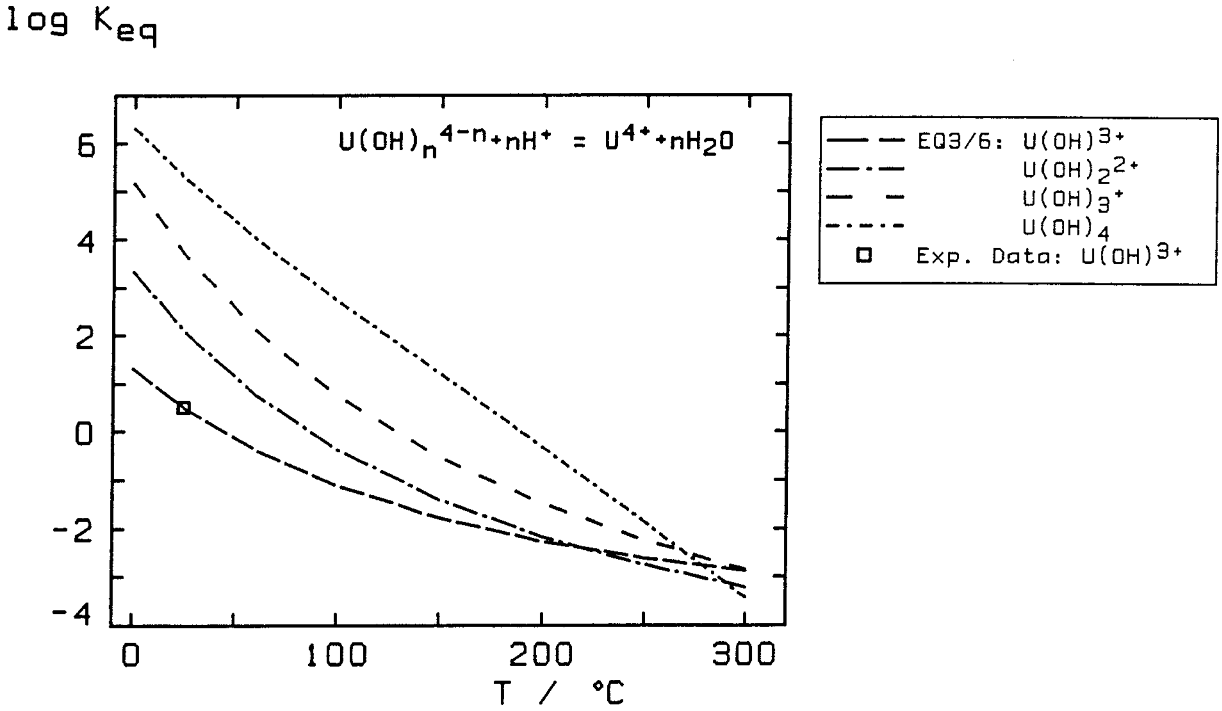

Figure 3.1 The equilibrium constants for U(IV) hydroxide complexes (calculated by the MCRT program of the EQ3/6 package) versus temperature. The experimental value reported by Grenthe et al (1988b) is also shown for comparison.

e - Similarly, the value of S^o for the neutral species $UO_2CO_3(aq)$ which is reported as 0±15 cal/mol K by Lemire and Tremaine (1980), was changed to +20 cal/mol K. The resulting values for the mononuclear carbonate complexing constants as a function of temperature are shown in Figure 3.2, together with the experimental values of Pirozhkov and Nikolaeva (1976).

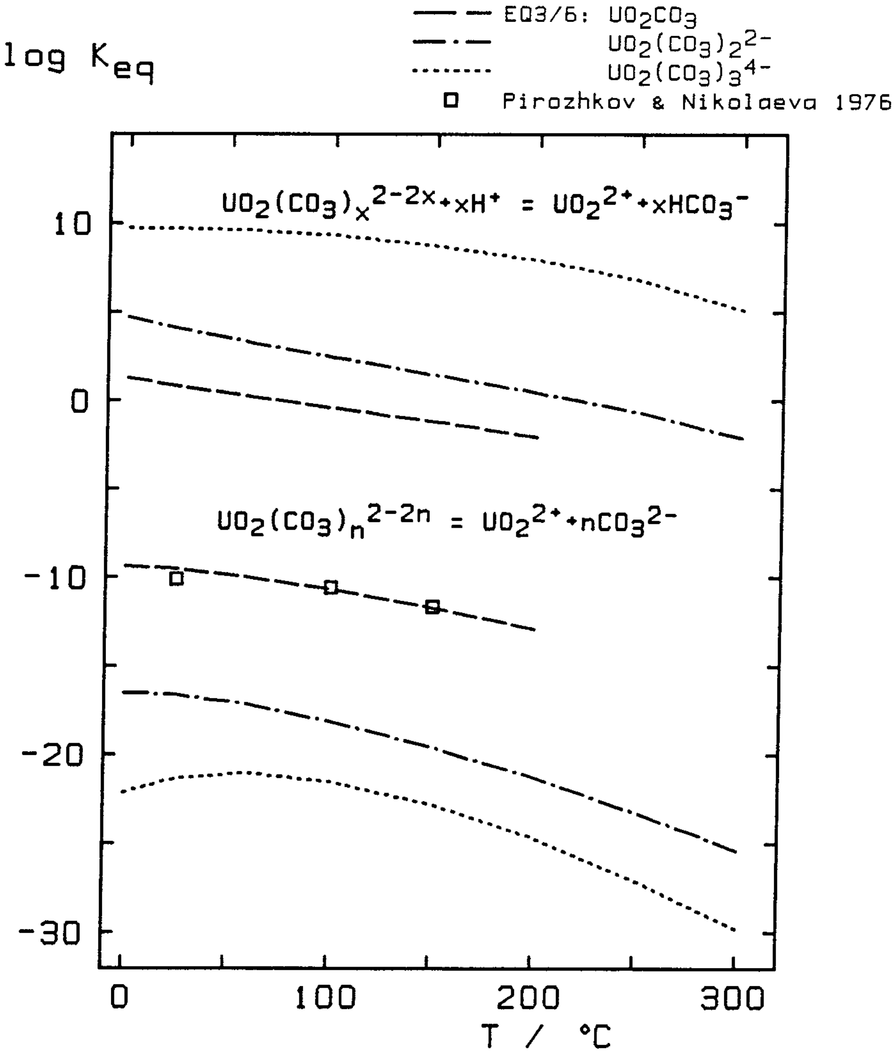

Figure 3.2 The equilibrium constants for mononuclear uranyl carbonato complexes (calculated by the MCRT program of the EQ3/6 package) versus temperature. The experimental values of Pirozhkov and Nikolaeva (1976) are also shown for comparison.

f - $\Delta G_f°$ for $(UO_2)_3(OH)_7^-$ was changed from -1034.9 k cal/mol (Lemire, 1988) to -1035.6 k cal/mol, which gives an equilibrium constant of -32.40 (at 25°C) for the following reaction

$$3\ UO_2^{2+} + 7\ H_2O = (UO_2)_3(OH)_7^-$$

This value results in somewhat lower calculated solubilities for U(VI) hydroxides at pH>8, which are in better agreement with recent available experimental evidence (Bruno and Sandino 1988).

g - $\Delta G_f°$ for $(UO_2)_3(CO_3)_6^{6-}$ corresponds to an equilibrium constant for the reaction

$$3UO_2^{2+} + 6CO_3^{2-} = (UO_2)_3(CO_3)_6^{6-}$$

equal to +53.36 at 25°. This is in agreement with the value of 53.4 $\pm$ 0.8 reported by Grenthe et al (1984).

4 METHODS OF CALCULATION

The EQ3/6 code package (versions: EQ2NR: 3245R90, EQ6: 3245R79) has been used both to calculate uranium solubilities in aqueous solutions as well as to calculate the equilibrium constants (given in Appendix B) from the thermodynamic data for uranium (given in Appendix A). The EQ3/6 code package is described in Wolery (1979) and (1983), Wolery et al (1984), Delany and Wolery (1984), Bourcier (1985) and Delany et al (1986).

For the calculation of chemical equilibrium in aqueous systems, the EQ3NR code is based both on the charge balance equation, and on the mass balance equations for all elements except hydrogen and oxygen, while the EQ6 code is based on mass balance equations for all elements (including oxygen and hydrogen). For the specific case of solid solubility calculations with the EQ3NR program, the alternative constraint of phase equilibrium with a mineral substitutes the mass balance equation for uranium. For calculations with a fixed partial pressure of $CO_2(g)$, this constrain will substitute the mass balance equation for carbon in the EQ3NR calculations. The redox constraint used was either oxygen or hydrogen fixed gas fugacity.

The principal unit of concentration on which thermodynamic activities, mass balances, equilibrium constants and activity coefficients are based, is molality (mols/1000 g H_2O). The activity coefficents (based on this molality scale) are calculated with the $B^{\bullet}$ equation (Helgeson, 1969).

All EQ3NR and EQ6 calculations were performed for aqueous solutions electrically neutral, achieved by adjusting the concentration of either Na^+ of ClO_4^- in the modeled solutions. This is needed to make realistic calculations of the activity coefficients for the aqueous species. In the case of solubility calculations in aqueous solutions containing a supporting electrolyte, the individual concentrations of the ions in the ionic media were not adjusted, and therefore, in these cases the electrical charge balance sometimes was in error by a few percent, although this does not have much influence on the value of the calculated activity coefficients in such concentrated solutions.

We have compared calculated solubilities (in mols per 1000 g H_2O) as a function of pH (= -log a_{H+}, activity in molal scale) with experimental values, which are not always given as molalities. The difference between molar (M = mols/l) and molal (m = mols/1000 g H_2O) is not important for dilute solutions at 25°C, where uranium solubilities are low and pH is within the range 2-12. In other cases, however, a correction is necessary, as for the experimental data from the following sources:

- the solubility of uranyl carbonate in sodium carbonate solutions of Blake et al (1956) was converted from molar to molal units using the densities of the equilibrium solutions reported by Blake et al (1956).

- the solubility data of uranyl carbonate in 0.5 M $NaClO_4$ of Grenthe et al (1984) was converted from molar to molal scale (both log h (=log$[H^+]$) and log B (=log$[U(VI)]_{tot}$)) using a density of 1.0362 g/cm^3. Thus, in Figure 5.8, calculated values of -log$[H^+]$ (instead of -log a_H+=pH) are compared with the experimental values of Grenthe et al (1984) corrected to molal units.

- the results of Babko and Kodenskaya (1960) on the solubility of uranyl hydroxide in 0.2 M NH_4NO_3 were converted to molal scale using a density of 1.0034 g/cm^3.

- the data of Bruno et al (1987) at 0.5 M $NaClO_4$ for the solubility of UO_2(am), were corrected to the molal scale using a density of 1.0362 g/cm^3.

  Bruno et al (1987) report solubility as a function of -log $[H^+]$. In order to obtain approximate pH values, we have used an activity coefficient correction for H^+:

  $$-\log a_{H+}=-\log [H^+]-\log f_{H+}=-\log [H^+]+0.10$$

  The correction should be approximately valid in 0.5 M $NaClO_4$ at pH>2 and uranium solubilities less than 0.01 mols/l. The pH values thus obtained are plotted in Figure 5.12.

The results of Silva and Yee (1981), although they were measured in 0.001 M $NaClO_4$ solutions, were not corrected for the missmatch of concentration units.

The data of Parks and Pohl (1988) and of Nikolaeva and Pirozhkov (1978) on the solubility of uraninite versus pH at temperatures > 25°C is given in mols/l, as well as the solubility of uranyl hydroxide by Brush (1980) and Miller (1958). However, we have assumed that the data refers to uranium analysis made at about 25°C, and therefore, as molar uranium concentrations are practically equal to molal concentrations in diluted samples at about 25°C, and molal uranium concentration values are temperature independent, we have not made any correction on the data tabulated by either Parks and Pohl (1988), Nikolaeva and Pirozhkov (1978), Miller (1958) or Brush (1980).

The data from the following sources are also given in molar units, but were used in this report without corrections: Galkin and Stepanov (1961), Ryan and Rai (1983), and Krupka et al (1985).

The solubilities reported by the following authors are given in molality scale, and therefore did not need any correction: Tremaine et al (1981), Gayer and Leider (1955) and (1957), Sergeyeva et al (1972), Redkin and Omelyanenko (1987), Nikitin et al (1972), and Nguyen-Trung (1985).

5 RESULTS AND DISCUSSION

The results presented in this section include calculated uranium solubilities for many aqeous solutions in equilibrium with several uranium solid phases. Agreement between the results of the calculations and experimental solubilities might be taken as a validation of the SKBU1-EQ3/6 combination.

However, a distinction must be made between validation and verification calculations. A validation of a model is only possible when the experimental data used are not related to the model, i.e., when the model has been obtained independently of the experimental data used in the validation.

Unfortunately, in this case we know that some of the experimental data have been used to develop the data base. For example, the standard free energies of formation for UO_2(am) and $U(OH)_4$(aq) have been derived from the solubility data of Bruno et al (1987). Therefore, agreement between EQ3/6-SKBU1 calculated results and the experimental data of Bruno et al (1987) in Figure 5.12 will only give a proof of self consistency and show that no serious errors have been introduced in the procedure of developing the SKBU1 data base. Note, however, that in the same Figure 5.12, other solubility data is shown that may be used as a validation.

Having this in mind, lets proceed to the examination of the results obtained in our calculations.

5.1 THE SOLUBILITY OF $UO_2(OH)_2(c)$ AND SCHOEPITE $(UO_2(OH)_2 \cdot H_2O)$

A comparison between experimental and calculated solubility for schoepite and $UO_2(OH)_2(c)$ is shown in Figure 5.1. The theoretical solubility curve for $Na_2U_2O_7(c)$ has also been plotted to indicate the pH range where alkali uranates might be expected to be the stable solid phase.

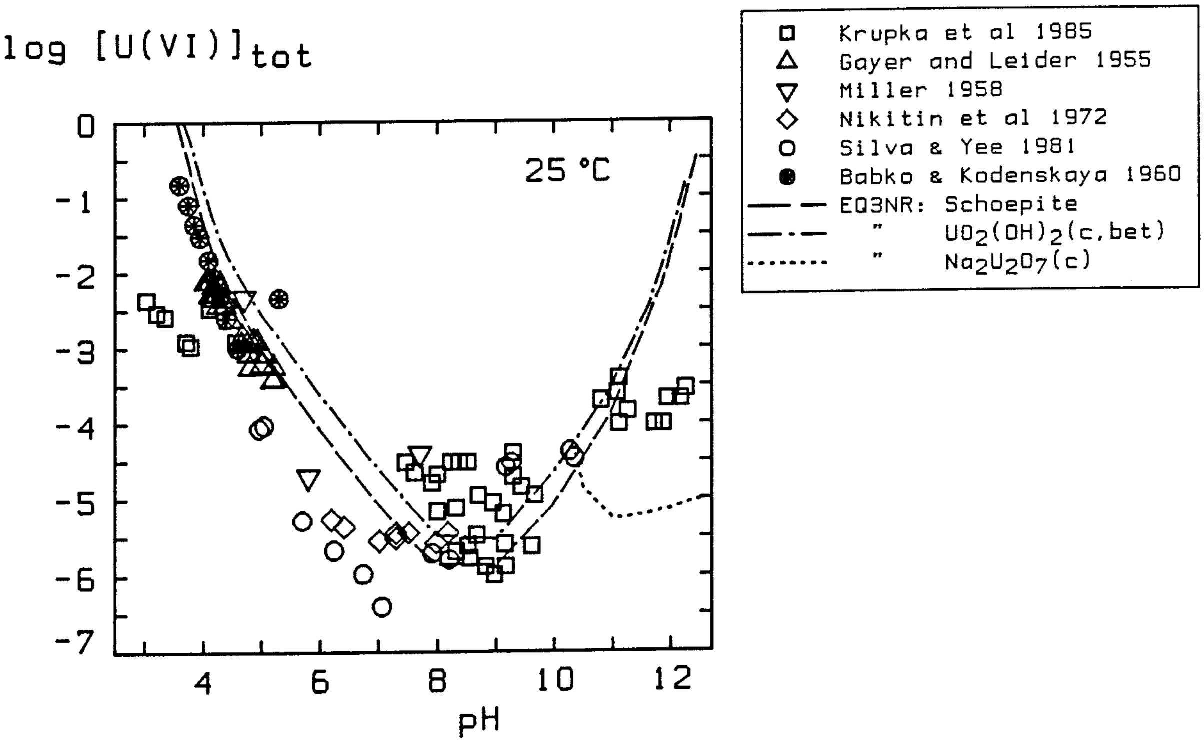

Figure 5.1 The solubility at 25°C of $UO_2(OH)_2(c)$, schoepite and $Na_2U_2O_7(c)$ as a function of pH.

The agreement between calculated and experimental solubilities is rather good. Only some of the data by Krupka et al (1985) deviate from the general trend. The spread of the data from Krupka et al (1985) difficults a more sensitive analysis of the solubilities in the pH range 6 to 12. Previously recomended values of the equilibrium constant β_{37} for the equilibrium

$$3UO_2^{2+} + 7H_2O = (UO_2)_3(OH)_7^- + 7H^+$$

gave calculated values for $[UO_2^{2+}]_T$ that were too large in the alkaline pH range, because $(UO_2)_3(OH)_7^-$ appears to be the dominating complex in this pH range. Recent data by Bruno and Sandino (1988) give a preliminary value of log β_{37}= -32.1±0.1. As mentioned in Section 3e, we have used a value of log β_{37}=-32.4.

The data given by Gayer and Leider (1955) are shown in Figures 5.2 and 5.3. In perchloric acid solutions the agreement between the model and the experiments is satisfactory. At high concentrations the model deviates from reality probably because the $B^{\bullet}$-equation for activity coefficients fails, as expected.

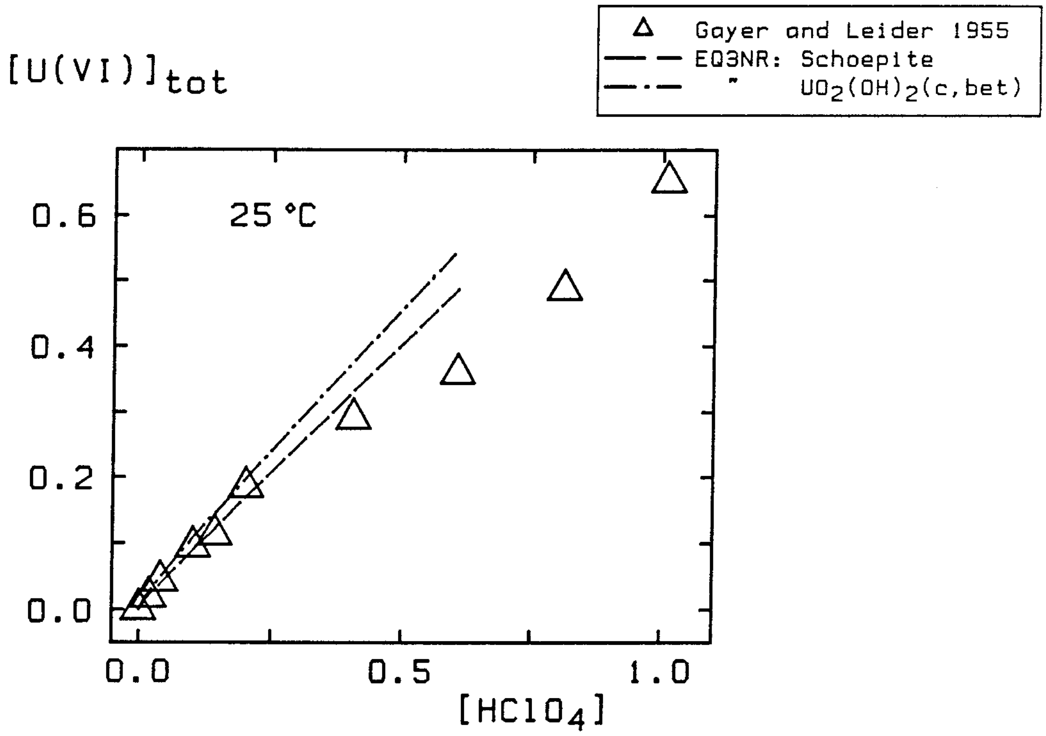

Figure 5.2 The solubility at 25°C of $UO_2(OH)_2$(c) and schoepite as a function of perchloric acid concentration.

In sodium hydroxide solutions, the presence of alkali uranates is expected. Gayer and Leider (1955) state "the solid phase from basic samples contained sodium". The conclusion extracted from Figure 5.3 is that further studies on the solubility of well characterised alkali uranates are needed.

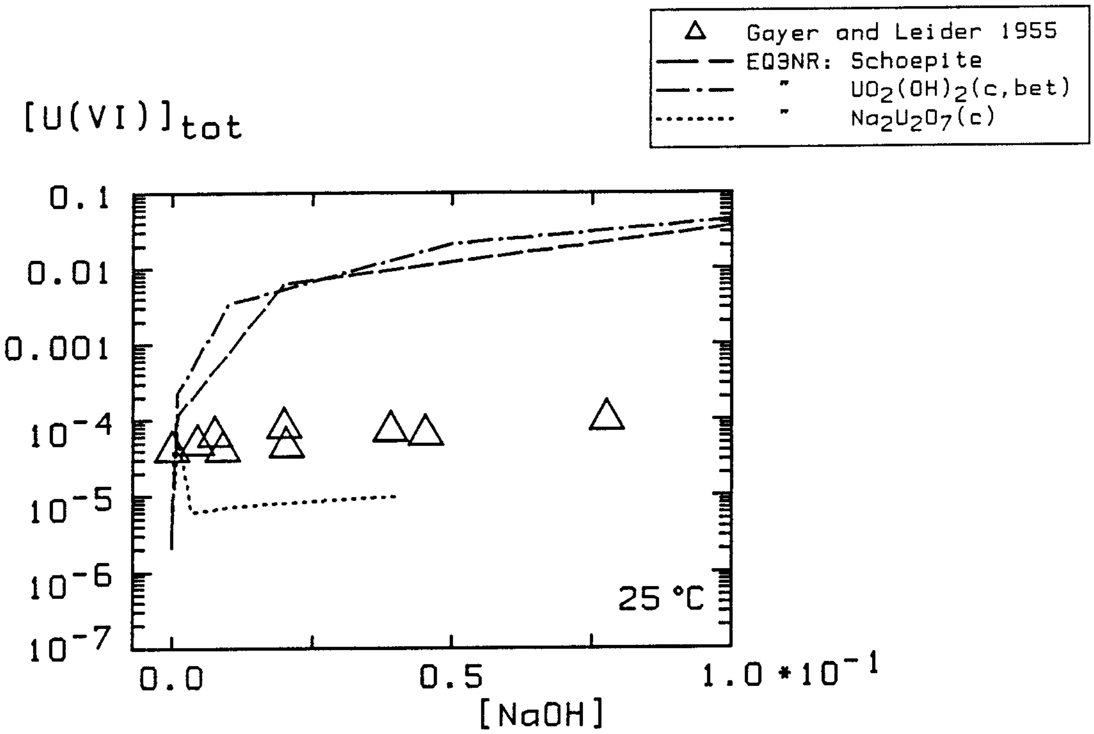

Figure 5.3 The solubility at 25°C of $UO_2(OH)_2(c)$, schoepite and $Na_2U_2O_7(c)$ as a function of sodium hydroxide con centration.

The solubility of schoepite in carbonate solutions at 25°C is reported in Babko and Kodenskaya (1960). The results are shown in Fig 5.4 together with the calculated curves. Except for two experimental points at $[CO_3^{2-}]_T<0.01$ and $pH\cong7.3$, the agreement is quite satisfactory (the disagreement is less than a factor of about two in the uranyl concentrations).

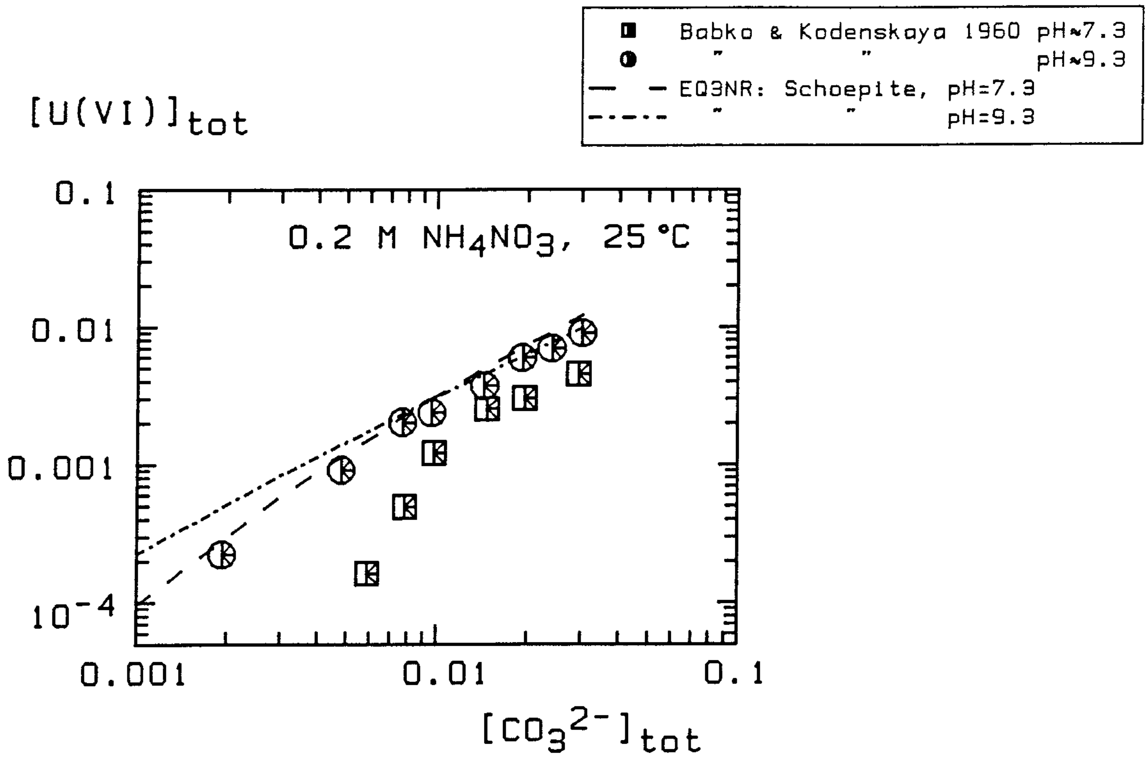

Figure 5.4 The solubility at 25°C of schoepite and as a function of carbonate concentration in 0.2 M NH_4NO_3.

The data of Brush (1980) have been used in order to test the thermodynamic data at high temperature. The $[U(VI)]_T$ versus pH at 90°C is compared with the EQ3NR results in Figure 5.5. The agreement between the experimental and calculated solubility of $UO_2(OH)_2(c)$ (the stable phase at T>60°C) is good up to pH=6. The model concentrations appear to be too large in the alkaline range. The thermodynamic data for $UO_2(OH)_2(c)$ is rather well stablished, and the disagreement is probably a consequence of erroneous values for the standard entropies of anionic UO_2^{2+} hydroxide complexes ($UO_2(OH)_4^{2-}$ and $(UO_2)_3(OH)_7^-$).

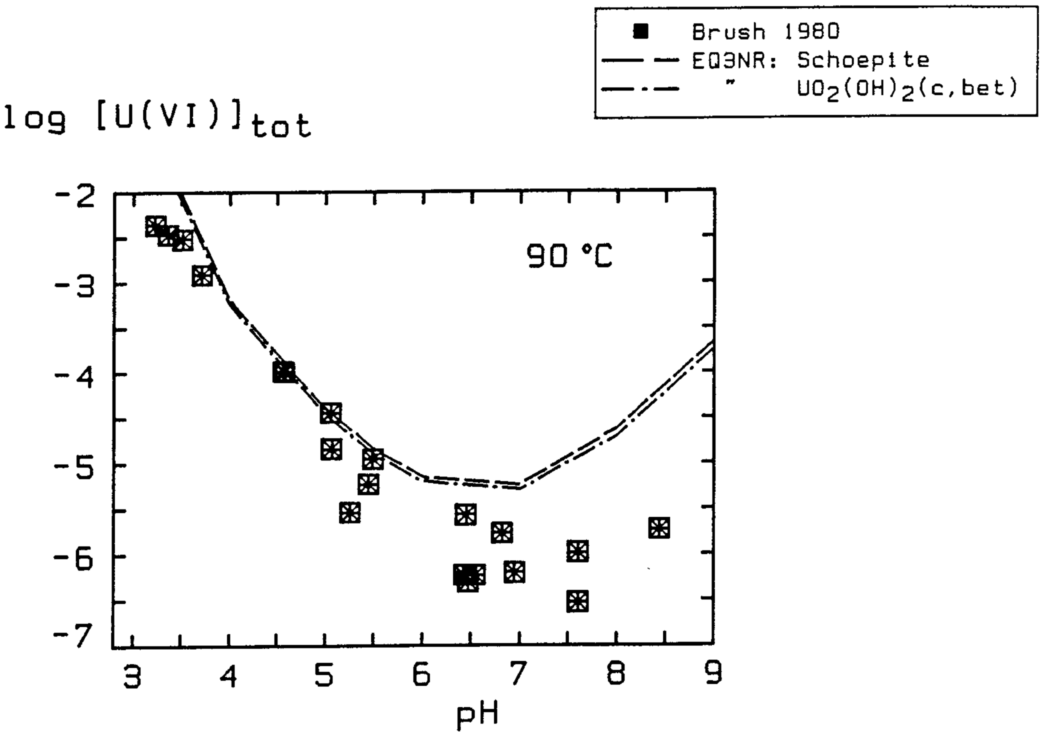

Figure 5.5 The solubility at 90°C of $UO_2(OH)_2(c)$ as a function of pH.

In Figure 5.6 we show an overall comparison of calculated solubilities of schoepite and $UO_2(OH)_2(c)$ in water with experimental values as a function of temperature in the pH range 7 to 8 (except for the values at 155 and 215°C of Miller (1958), that have pH=6 and 5 respectively, however, the pH values reported by Miller (1958) were probably measured at low temperature). The phase transition from schoepite to $UO_2(OH)_2(c)$ is reflected in the results of the calculations. The solubilities of Nikitin et al (1972) and Brush (1980) appear to follow the same curve, while the values of Miller (1958) are about a factor of ten higher, which might be due to lower pH-values for the solutions investigated by Miller (1958). The EQ3NR calculations are in satisfactory agreement with the solubilities of Nikitin et al (1972) and Brush (1980). The EQ3NR calculations show that the dominant species responsible for the uranium solubilities in neutral solutions are $UO_2(OH)_2(aq)$, $(UO_2)_3(OH)_5^+$ and $(UO_2)_3(OH)_7^-$.

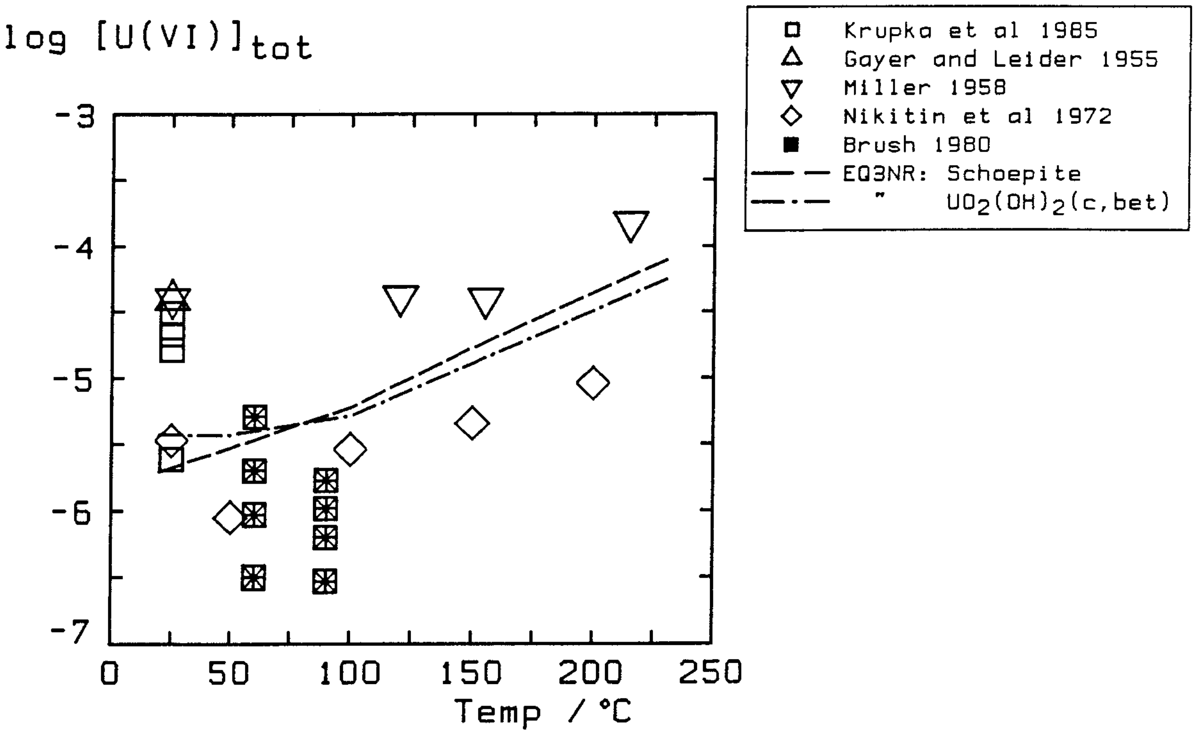

Figure 5.6 The calculated solubility of schoepite and $UO_2(OH)_2(c)$ in water as a function of T, compared with experimental values in the pH range 7 to 8.

5.2 THE SOLUBILITY OF RUTHERFORDINE ($UO_2CO_3(c)$)

The uranyl solubilities of rutherfordine at 25°C have been measured at constant partial presure of $CO_2(g)$ by Sergeyeva et al (1972) and Grenthe et al (1984). The agreement is fair (c.f. Figure 5.7) for the experiments in diluted solutions of Sergeyeva et al (1972) at p_{co2} = 1 atm. The agreement between EQ3NR calculations and the data of Grenthe et al (1984) at 0.5 $NaClO_4$ and P_{co2}=9.97 and 0.098 atm is also rather good, as shown in Figure 5.8. The equilibrium constants for the formation of uranyl cabonato complexes and the solubility product of rutherfordine at infinite dilution that are included in the SKBU1 data base were obtained by Grenthe et al (1984) using the "SIT"-equations. Therefore, the slight disagreement (sometimes by a factor of three, as shown in Figure 5.8) is probably a result of the difference in the activity factor correction method used in the EQ3/6 calculations as compared with the correction method used by Grenthe et al (1984) to obtain the thermodynamic data.

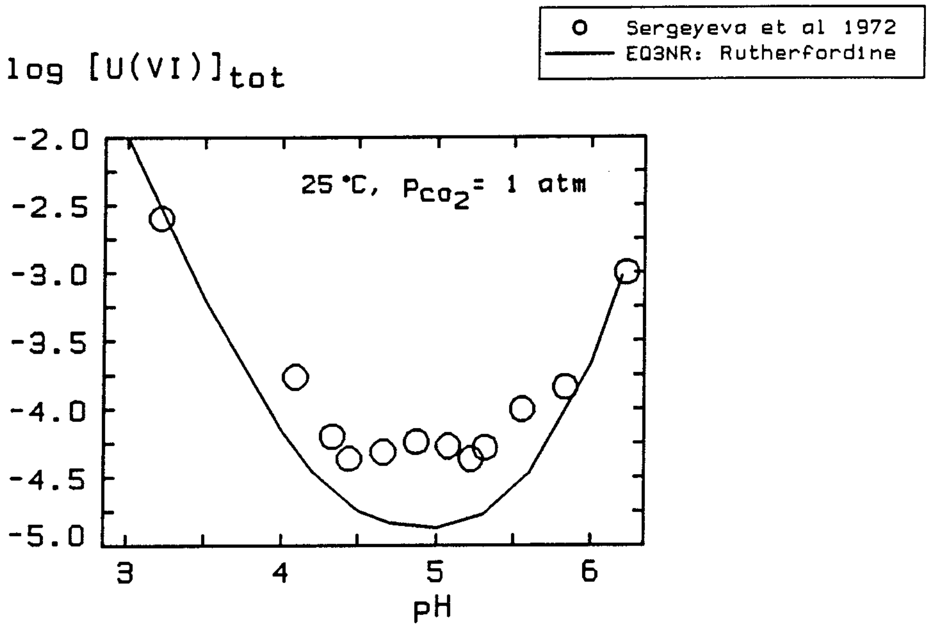

Figure 5.7 The solubility of rutherfordine ($UO_2CO_3(c)$) in dilute solutions as a function of pH at p_{co2}=1 atm and 25°C.

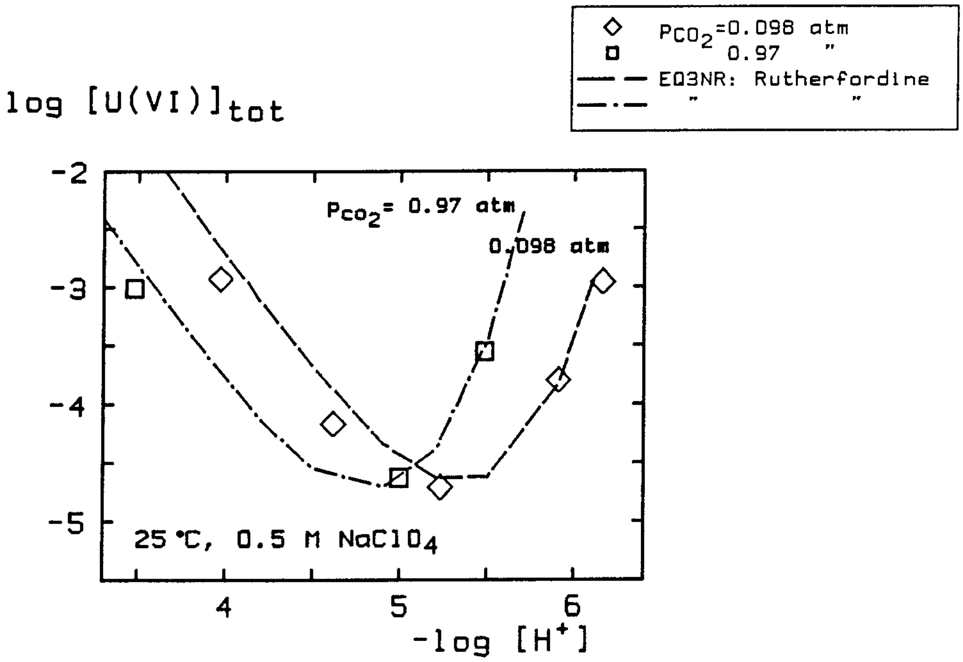

Figure 5.8 The solubility of rutherfordine ($UO_2CO_3(c)$) in 0.5 M $NaClO_4$ solutions as a function of pH at 25°C and p_{co2}=0.97 and 0.098 atm.

The experimental solubility of uranyl carbonate in Na_2CO_3 solutions at 25°C can be reproduced quite well by the EQ3NR calculations. Figure 5.9 compares the data of Blake et al (1956) with the solubilities calculated by using the SKBU1 data base. The EQ3/6 results show that the dominant uranium species in the modeled solutions is $(UO_2)_3(CO_3)_6^{6-}$.

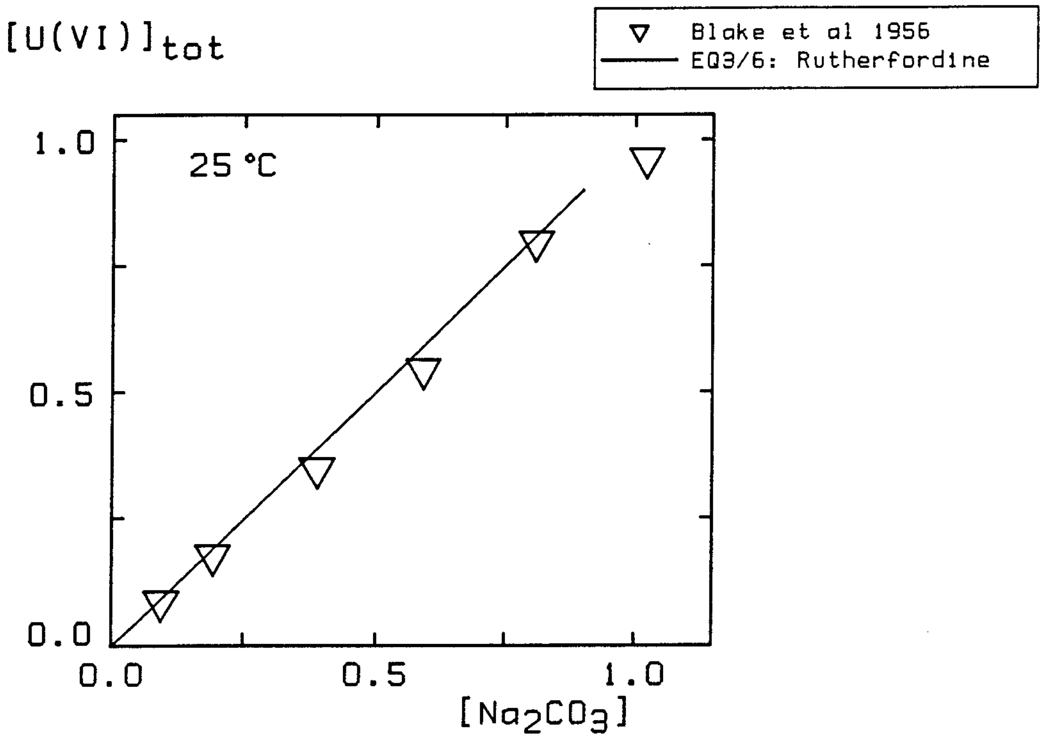

Figure 5.9 The solubility of rutherfordine ($UO_2CO_3(c)$) as a function of $[Na_2CO_3]$ at 25°C.

The solubility of rutherfordine reported by Sergeyeva et al (1972) was also used to test the thermodynamic data base for uranium at high temperature. The agreement at 50°C (see Figure 5.10) is fairly good (as it was at 25°C) in the whole pH range. The agreement at T>50°C is shown in Figure 5.11, which shows the calculated solubility of uranyl carbonate in water at the given partial pressures of $CO_2(g)$, with the results reported by Sergeyeva et al (1972). The disagreement at T>50°C might be either an artifact on the experimental results, or incorrect values for either the standard entropies of some aqueous complexes or for the heat capactiy function of rutherfordine.

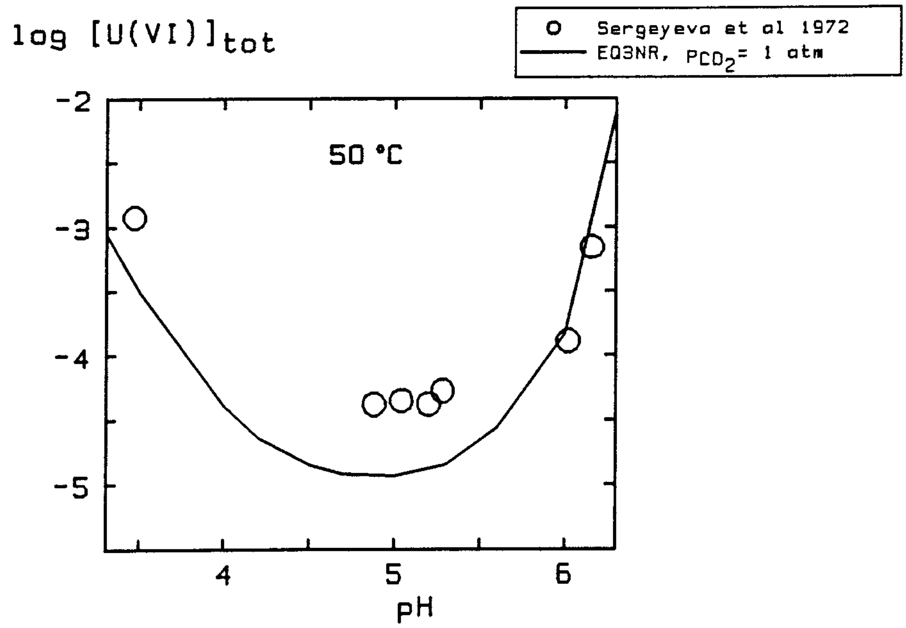

Figure 5.10 The solubility of rutherfordine ($UO_2CO_3(c)$) in dilute solutions as a function of pH at p_{co2}=1 atm and 50°C.

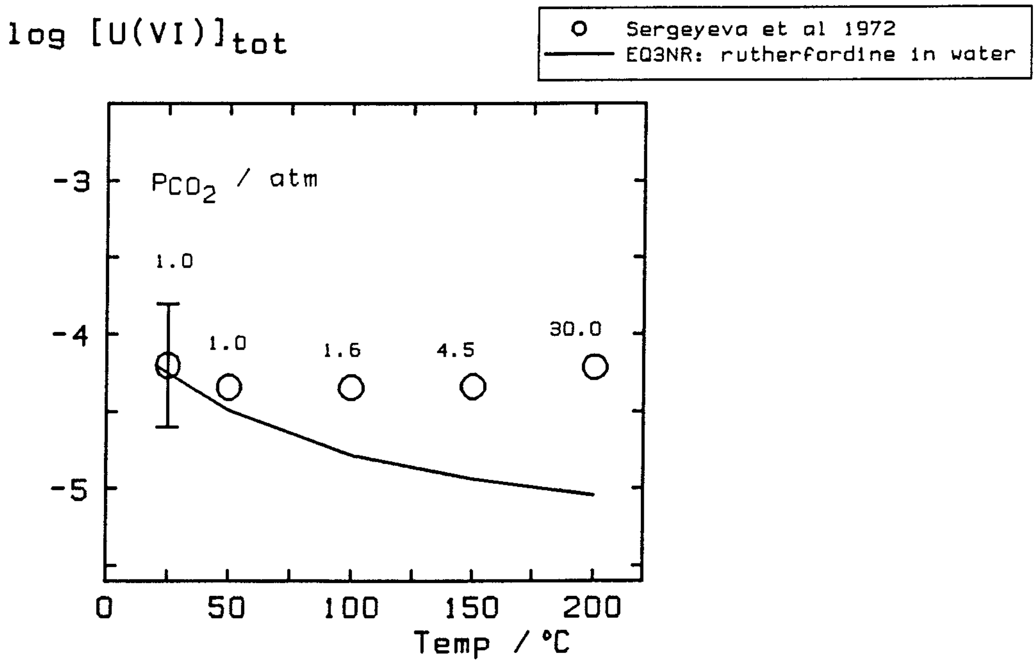

Figure 5.11 The calculated solubility of rutherfordine ($UO_2CO_3(c)$) in water as a function of temperature at the given values for the partial pressure of $CO_2(g)$ (p_{co2}) compared with experimental results of Sergeyeva et al (1972).

5.3 THE SOLUBILITY OF $UO_2(s)$

Solubility data for U^{4+} oxide of various degrees of crystallinity as a function of pH and T have been used to validate the SKBU1 thermodynamic set of data.

The values of $[U(IV)]_T$ versus pH at 25°C and pH<13 for $UO_2(am)$ reported by Gayer and Leider (1957), Galkin and Stepanov (1961), Nikolaeva and Pirozhkov (1978) and Bruno et al (1987), have been compared to EQ3NR results in Figures 5.12 and 5.13. The lack of agreement at pH<6 indicates that the U(IV) solubility cannot be explained in the acid range without taking into account the formation of polynuclear complexes ($U_p(OH)_q^{4p-q}$). Nevertheless, the agreement between model and experiments is good at pH>6.

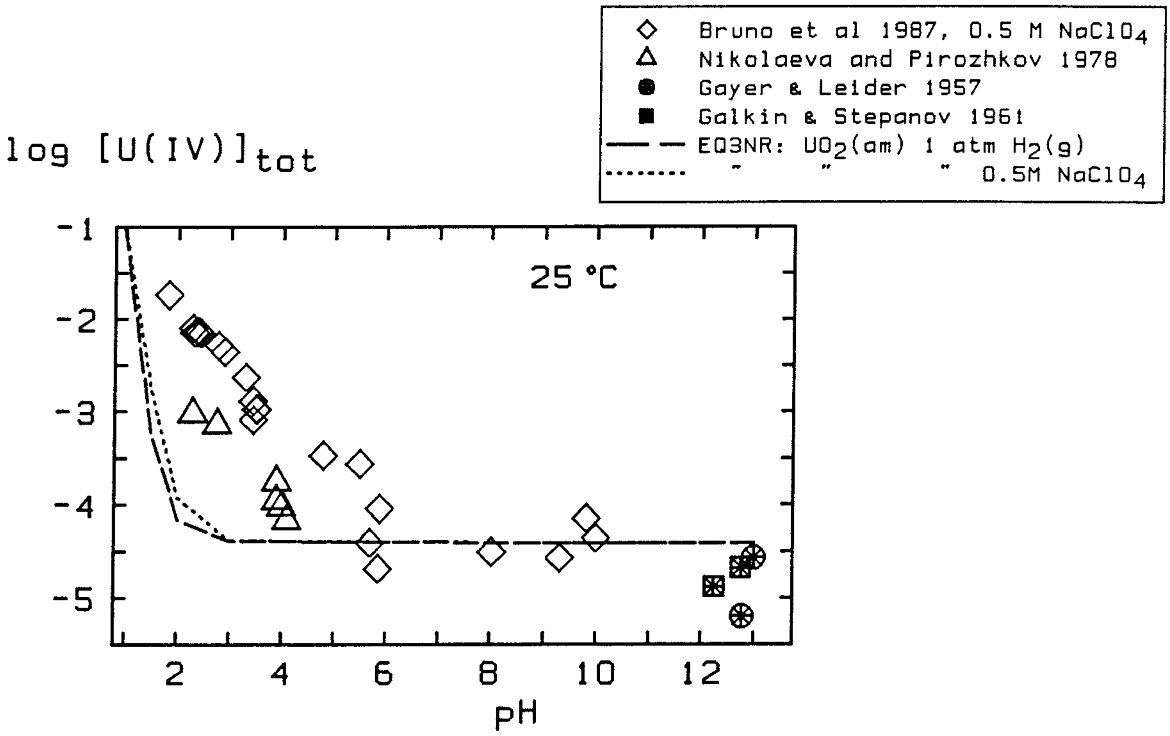

Figure 5.12 The solubility of $UO_2(am)$ (both in diluted solutions and in 0.5 M $NaClO_4$) as a function of pH at 25°C.

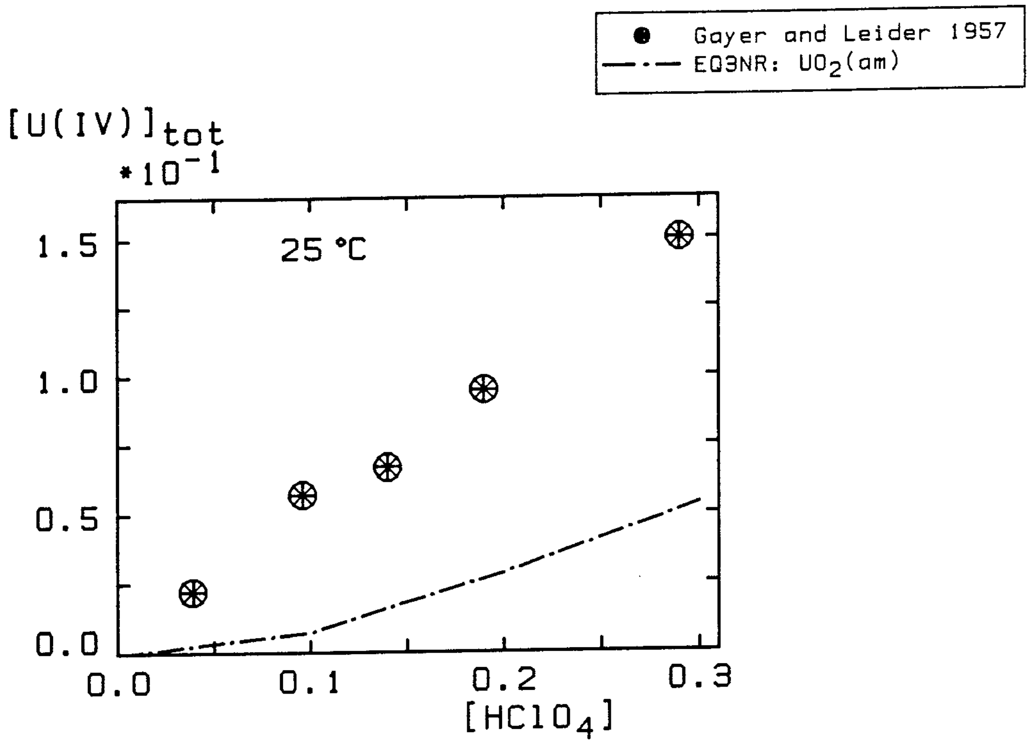

Figure 5.13 The solubility of UO_2(am) in perchloric acid solutions at 25°C.

The solubility data for crystalline U^{4+} oxide (uraninite) has been reported by Nikolaeva and Pirozhkov (1978), Tremaine et al (1981), Nguyen-Trung (1985), Redkin and Omelyanenko (1987) and Parks and Pohl (1988). A comparison between EQ3NR results and experimental data is shown in Figures 5.14 to 5.16. The data of Tremaine et al (1981) were taken with the same solid used by Parks and Pohl (1988). However, the detection limit of the analytical method for uranium used by Tremaine et al (1981) apparently was $2 - 5 \cdot 10^{-8}$ m, which is higher than the experimental uranium concentrations found by Parks and Pohl (1988). Therefore, the data of Tremaine et al (1981) are included in Figures 5.14 to 5.16 only for qualitative comparison.

If we then disregard the values of Tremaine et al (1981), the agreement between the calculated and the experimental solubilities shown in Figures 5.14 to 5.16 is satisfactory for pH>3.

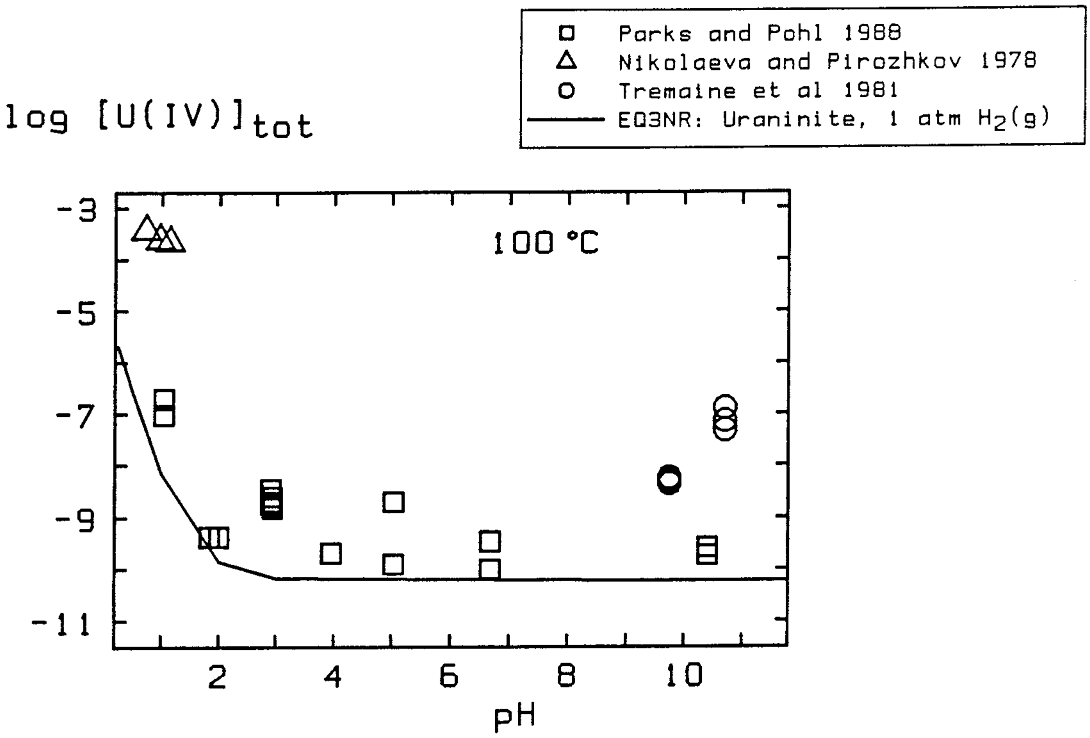

Figure 5.14 The solubility of crystalline $UO_2(s)$ versus pH at 100°C.

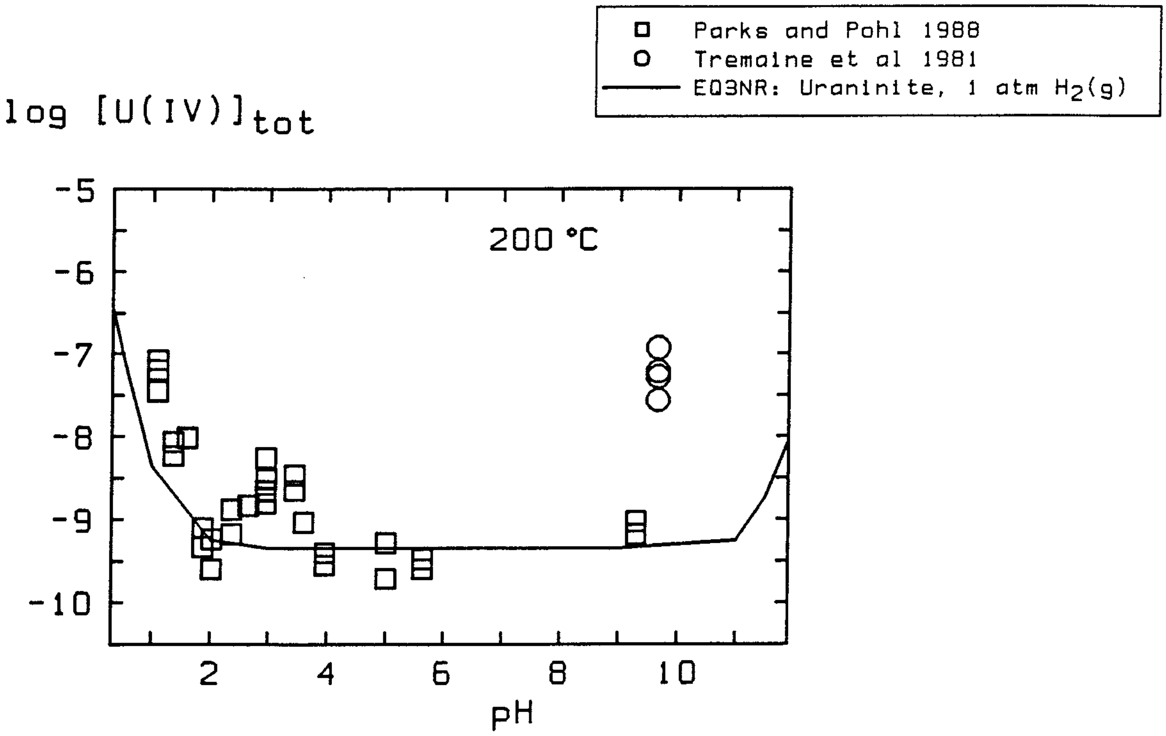

Figure 5.15 The solubility of crystalline $UO_2(s)$ versus pH at 200°C.

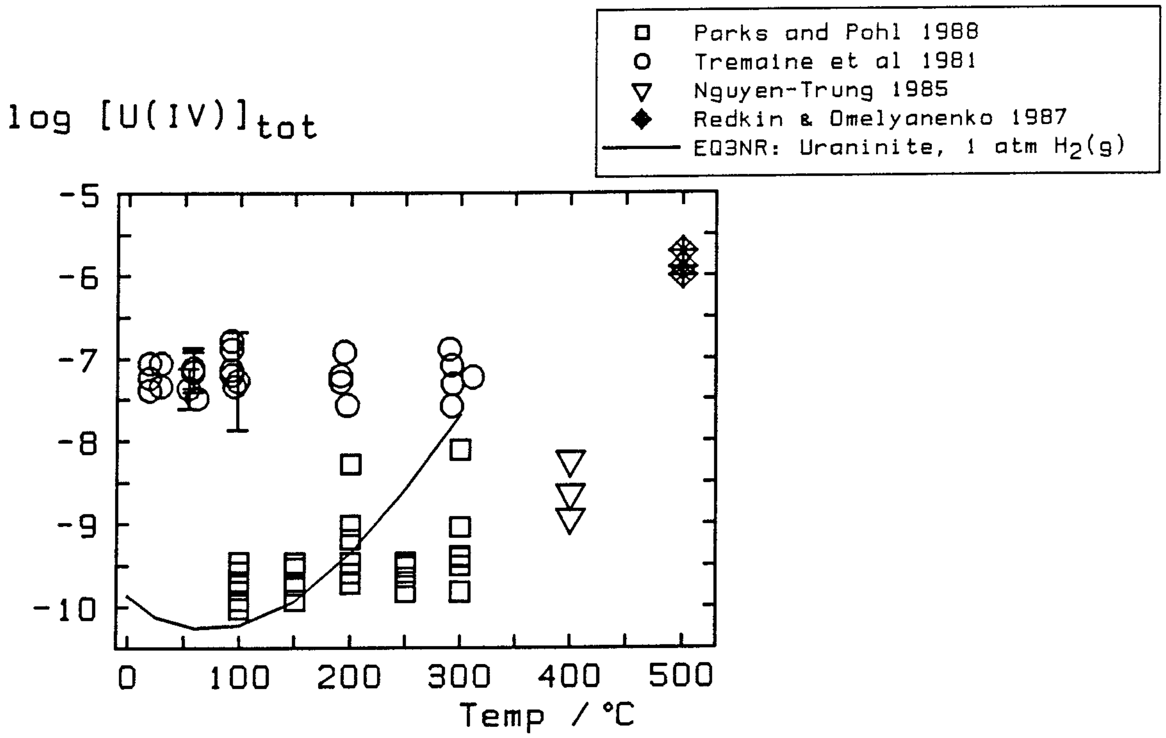

Figure 5.16 The calculated solubility of crystalline $UO_2(s)$ in water at 1 atm $H_2(g)$ versus T, compared with experimental literature values (for $UO_2(c)$ solubility either in water or in diluted solutions of pH>5).

6 CONCLUSIONS

The work that we have presented here has two main aspects: a literature survey of the uranium solubility data, and a verification/validation of a thermodynamic data base for uranium. This allowed us to find out areas where new experimental measurements are needed, as well as small error and mistakes in the data base. The most prominent results are the following:

- The solubility of UO_2^{2+} in acid solutions has been studied by several authors (c.f. Figure 2.2) and there is reasonable consistency between different authors in the literature. However, in alkaline solutions the situation is not as satisfactory. More research is needed on the stability of anionic uranium(VI) hydroxide complexes. Furthermore, studies on the solubility of well characterized alkaline uranates are needed to gain any confidence on the capabilities of geochemical models for uranium in alkaline environments (e.g., in a concrete repository).

- For the UO_2^{2+}/CO_3^{2-} system, the validation exercise has provided encouraging results about the capability of the EQ3/6-SKBU1 combination to predict the behaviour of uranium in quite wide ranges of pH, $[CO_3^{2-}]_T$, p_{co2} and temperature.

- The validation of the data base for the U^{4+}/OH^- system has shown that there is a disagreement between calculated and experimental solubilities in acid solutions (pH<6 at 25°C). This is probably due to lack of thermodynamic data on the stability of polynuclear uranium(IV) hydroxide complexes. In neutral and moderately alkaline solutions, the agreement between calculated and literature solubilities is more satisfactory.

- Another problem that arises in this types of geochemical calculations is the estimation of activity coefficients at moderate to high ionic strength. Extended Debye-Hückel approaches (e.g. Davies and $B^{\bullet}$ eqns.) often do not work satisfactorily. There is the possibility of using the equations proposed by Pitzer within the EQ3/6 code package. This method performs properly up to very high ionic strengths. Nevertheless, the lack of appropiate interaction parameters for most of radionuclides and their complexes, difficults the use of Pitzer's method. Recently, specific interaction equations (called SIT, see for example Biederman et al 1982, and Bruno et al 1985, and references therein) have been adopted within the NEA-TDB effort for this purpose (Grenthe and Wanner, 1988). It would be highly desirable to adapt these equations in some geochemical codes.

We are working on further research in this area. For example, preliminary calculations have shown that the thermodynamic data for the UO_2^{2+}/PO_4^{3-} system in the EQ3/6 data base is erroneous. An experimental program has been started on this system, as well as a verification/validation study will be started.

7 ACKNOWLEDGEMENTS

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and in "Proceedings Conference on Application of Geochemical Models to High-Level Nuclear Waste Repository Assessment", K.G.Jacobs and S.K.Whatley (eds), Oak Ridge, Tennesee, Oct. 1984, NUREG/CP-0062, ORNL/TM-9585, pp. 54-65

APPENDIX A

Data blocks to be used in the MDAS file of the EQ3/6 package. The MDAS file is transformed to a direct access file (MDAR) before it is used as input for the MCRT program. The MCRT program calculates equilibrium constants as a function of temperature, and writes the results in a DFILE. Appendix B contains a list of the DFILE obtained with the data blocks listed beneath.

```
mdas.3245s01     unified mcrt data file
last modified 03Oct88
+----------------------------------------------------------------------
u+++                                                   date= 06-Sep88
 entered by= j. bruno                  quality= good
 keys    = aux             element
 subsets= uranium
     charge=    3.0         titr. factor=    0.0 eq/mol
   ion type=  1                 ion size=    0.0 a     hydr. no.=    0.0
  1 chemical elements
       1.000 u
          temp=    298.150 k          press=      1.013 bars
         delg0=   -114.89  kcal/mol   delh0=    500.000 kcal/mol
            s0=    -41.80  cal/mol/k     se=      4.570 cal/mol/k
            v0=            cc/mol        cp=     -15.3  cal/mol/k
  0 species in reference reaction
  5 species in data0 reaction
     -1.000  u+++                         -1.000  h+
     -0.250  o2(g)                        +1.000  u++++
     +0.500  h2o
 t-p data grid is absent
 source=
* [lemire and tremaine, 1980]
+----------------------------------------------------------------------
u++++                                                  date= 06-Sep88
 entered by= j. bruno                  quality= good
 keys    = basis           element
 subsets= uranium
     charge=    4.0         titr. factor=    0.0 eq/mol
   ion type=  1                 ion size=   11.0 a     hydr. no.=    0.0
  1 chemical elements
       1.000 u
          temp=    298.150 k          press=      1.013 bars
         delg0=   -126.65  kcal/mol   delh0=   -141.30  kcal/mol
            s0=    -99.56  cal/mol/k     se=      4.360 cal/mol/k
            v0=            cc/mol        cp=    -11.47  cal/mol/k
  0 species in reference reaction
  0 species in data0 reaction
 t-p data grid is absent
 source=
* [grenthe et al., 1988a]
* [lemire and tremaine, 1980]  give delg0=-126.89, s0=-98.95
+----------------------------------------------------------------------
uo2+                                                   date= 06-Sep88
 entered by= j. bruno                  quality= fair
 keys    = aux
 subsets= uranium
     charge=    1.0         titr. factor=    0.0 eq/mol
   ion type=  1                 ion size=    0.0 a     hydr. no.=    0.0
  2 chemical elements
       1.000 u              2.000 o
          temp=    298.150 k          press=      1.013 bars
         delg0=   -229.59  kcal/mol   delh0=   -244.91  kcal/mol
            s0=     -5.98  cal/mol/k     se=      0.000 cal/mol/k
            v0=            cc/mol        cp=     23.42  cal/mol/k
  0 species in reference reaction
  5 species in data0 reaction
     -1.000  uo2+                         -3.000  h+
     +1.000  u++++                        +1.500  h2o
     +0.250  o2(g)
 t-p data grid is absent
 source=
* [grenthe et al., 1988a]
* [lemire and tremaine, 1980] give delg0=-231.50, s0=-5.98
+----------------------------------------------------------------------
uo2++                                                  date= 06-Sep88
 entered by= j. bruno                  quality= good
 keys    = aux
 subsets= uranium
     charge=    2.0         titr. factor=    0.0 eq/mol
   ion type=  1                 ion size=    0.0 a     hydr. no.=    0.0
  2 chemical elements
       1.000 u              2.000 o
          temp=    298.150 k          press=      1.013 bars
         delg0=   -227.66  kcal/mol   delh0=   -243.55  kcal/mol
            s0=    -23.47  cal/mol/k     se=      0.00  cal/mol/k
            v0=            cc/mol        cp=       1.2  cal/mol/k
  0 species in reference reaction
  5 species in data0 reaction
     -1.000  uo2++                        -2.000  h+
     +1.000  u++++                        +1.000  h2o
     +0.500  o2(g)
 t-p data grid is absent
 source=
* [grenthe et al., 1988a]
* [lemire and tremaine, 1980] give delg0=-227.70, s0=-23.18
```

```
+-----------------------------------------------------------------------
non-basis aqueous species
+-----------------------------------------------------------------------
(uo2)3(co3)(oh)3+                                           date= 06-Sep88
 entered by= j. bruno                         quality= good
 keys    =
 subsets= uranium
     charge=    1.0          titr. factor=    5.0 eq/mol
   ion type=  1                  ion size=    0.0 a     hydr. no.=    0.0
  4 chemical elements
       3.000 u               12.000 o                3.000 h
       1.000 c
          temp=    298.150 k            press=      1.000 bars
         delg0=   -983.03  kcal/mol     delh0=    500.000 kcal/mol
            s0=     35.85  cal/mol/k       se=      0.000 cal/mol/k
            v0=      0.000 cc/mol          cp=     -3.82  cal/mol/k
  0 species in reference reaction
  5 species in data0 reaction
    -1.000  (uo2)3(co3)(oh)3+                 -4.000  h+
     3.000  uo2++                              3.000  h2o
     1.000  hco3-
 t-p data grid is absent
 source=
* [lemire, 1988]
+-----------------------------------------------------------------------
(uo2)2(oh)2++                                               date= 06-Sep88
 entered by= j. bruno                         quality= good
 keys    =
 subsets= uranium
     charge=    2.0          titr. factor=    0.0 eq/mol
   ion type=  1                  ion size=    0.0 a     hydr. no.=    0.0
  3 chemical elements
       2.000 u                6.000 o                2.000 h
          temp=    298.150 k            press=      1.000 bars
         delg0=   -560.99  kcal/mol     delh0=    500.000 kcal/mol
            s0=     -4.54  cal/mol/k       se=      0.000 cal/mol/k
            v0=      0.000 cc/mol          cp=    -10.04  cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
    -1.000  (uo2)2(oh)2++                     -2.000  h+
     2.000  uo2++                              2.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988]
+-----------------------------------------------------------------------
(uo2)2oh+++                                                 date= 06-Sep88
 entered by= j. bruno                         quality= good
 keys    =
 subsets= uranium
     charge=    3.0          titr. factor=    0.0 eq/mol
   ion type=  1                  ion size=    0.0 a     hydr. no.=    0.0
  3 chemical elements
       2.000 u                5.000 o                1.000 h
          temp=    298.150 k            press=      1.000 bars
         delg0=   -508.37  kcal/mol     delh0=    500.000 kcal/mol
            s0=    -31.07  cal/mol/k       se=      0.000 cal/mol/k
            v0=      0.000 cc/mol          cp=    -24.4   cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
    -1.000  (uo2)2oh+++                       -1.000  h+
     2.000  uo2++                              1.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988]
+-----------------------------------------------------------------------
(uo2)3(co3)6 (6-)                                           date= 06-Sep88
 entered by= j. bruno                         quality= good
 keys    =
 subsets= uranium
     charge=   -6.0          titr. factor=   12.0 eq/mol
   ion type=  3                  ion size=    0.0 a     hydr. no.=    0.0
  3 chemical elements
       3.000 u               24.000 o                6.000 c
          temp=    298.150 k            press=      1.000 bars
         delg0=  -1512.80  kcal/mol     delh0=    500.000 kcal/mol
            s0=     43.02  cal/mol/k       se=      0.000 cal/mol/k
            v0=      0.000 cc/mol          cp=    133.8   cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
    -1.000  (uo2)3(co3)6 (6-)                 -6.000  h+
     3.000  uo2++                              6.000  hco3-
 t-p data grid is absent
 source=
* [lemire, 1988] gives delg0=-1512.978
* delg0 adapted for eq3/6 [puigdomenech and bruno, 1988]
+-----------------------------------------------------------------------
(uo2)3(oh)4++                                               date= 06-Sep88
 entered by= j. bruno                         quality= good
 keys    =
 subsets= uranium
     charge=    2.0          titr. factor=    0.0 eq/mol
   ion type=  1                  ion size=    0.0 a     hydr. no.=    0.0
  3 chemical elements
       3.000 u               10.000 o                4.000 h
          temp=    298.150 k            press=      1.000 bars
         delg0=   -893.50  kcal/mol     delh0=    500.000 kcal/mol
            s0=      9.56  cal/mol/k       se=      0.000 cal/mol/k
            v0=      0.000 cc/mol          cp=    -18.4   cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
    -1.000  (uo2)3(oh)4++                     -4.000  h+
     3.000  uo2++                              4.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988]
```

```
+-----------------------------------------------------------------------
(uo2)3(oh)5+                                                date= 06-Sep88
 entered by= j. bruno                     quality= good
 keys    =
 subsets= uranium
      charge=    1.0          titr. factor=     0.0 eq/mol
   ion type=  1                    ion size=     0.0 a      hydr. no.=    0.0
  3 chemical elements
       3.000 u                11.000 o               5.000 h
           temp=    298.150 k               press=      1.000 bars
          delg0=   -945.27  kcal/mol        delh0=    500.000 kcal/mol
             s0=     26.53  cal/mol/k          se=      0.000 cal/mol/k
             v0=      0.000 cc/mol             cp=      1.91  cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  (uo2)3(oh)5+                 -5.000  h+
      3.000  uo2++                         5.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988]
+-----------------------------------------------------------------------
(uo2)3(oh)7-                                                date= 06-Sep88
 entered by= j. bruno                     quality= good
 keys    =
 subsets= uranium
      charge=   -1.0          titr. factor=     7.0 eq/mol
   ion type=  3                    ion size=     0.0 a      hydr. no.=    0.0
  3 chemical elements
       3.000 u                13.000 o               7.000 h
           temp=    298.150 k               press=      1.000 bars
          delg0=  -1035.6   kcal/mol        delh0=    500.000 kcal/mol
             s0=     81.26  cal/mol/k          se=      0.000 cal/mol/k
             v0=      0.000 cc/mol             cp=    241.4   cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  (uo2)3(oh)7-                 -7.000  h+
      3.000  uo2++                        +7.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988] gives delg0=-1034.895
* delg0 adapted for eq3/6 [puigdomenech and bruno 1988]
+-----------------------------------------------------------------------
(uo2)4(oh)7+                                                date= 06-Sep88
 entered by= j. bruno                     quality= good
 keys    =
 subsets= uranium
      charge=    1.0          titr. factor=     7.0 eq/mol
   ion type=  1                    ion size=     0.0 a      hydr. no.=    0.0
  3 chemical elements
       4.000 u                15.000 o               7.000 h
           temp=    298.150 k               press=      1.000 bars
          delg0=  -1277.72  kcal/mol        delh0=    500.000 kcal/mol
             s0=     50.19  cal/mol/k          se=      0.000 cal/mol/k
             v0=      0.000 cc/mol             cp=    -12.43  cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  (uo2)4(oh)7+                 -7.000  h+
      4.000  uo2++                        +7.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988]
+-----------------------------------------------------------------------
uo2oh+                                                      date= 06-Sep88
 entered by= j. bruno                     quality= good
 keys    =
 subsets= uranium
      charge=    1.0          titr. factor=     1.0 eq/mol
   ion type=  1                    ion size=     0.0 a      hydr. no.=    0.0
  3 chemical elements
       1.000 u                 3.000 o               1.000 h
           temp=    298.150 k               press=      1.000 bars
          delg0=   -277.151 kcal/mol        delh0=    500.000 kcal/mol
             s0=      3.82  cal/mol/k          se=      0.000 cal/mol/k
             v0=      0.000 cc/mol             cp=     15.54  cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  uo2oh+                       -1.000  h+
      1.000  uo2++                         1.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988]
+-----------------------------------------------------------------------
uo2co3                                                      date= 03-Oct88
 entered by= j. bruno                     quality= good
 keys    =
 subsets= uranium
      charge=    0.0          titr. factor=     2.0 eq/mol
   ion type=  0                    ion size=     0.0 a      hydr. no.=    0.0
  3 chemical elements
       1.000 u                 1.000 c               5.000 o
           temp=    298.150 k               press=      1.000 bars
          delg0=   -366.83  kcal/mol        delh0=   -401.38  kcal/mol
             s0=    500.000 cal/mol/k          se=      0.000 cal/mol/k
             v0=      0.000 cc/mol             cp=            cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  uo2co3                       -1.000  h+
      1.000  uo2++                         1.000  hco3-
 t-p data grid is absent
 source=
* delg0 from [lemire, 1988], delh0 calculated from s0=+20.
* [lemire and tremaine, 1980] report s0=0 +-14
```

```
+-----------------------------------------------------------------------
uo2(co3)2--                                                   date= 06-Sep88
 entered by= j. bruno                        quality= good
 keys   =
 subsets= uranium
     charge=    -2.0           titr. factor=    4.0 eq/mol
  ion type=  3                      ion size=    0.0 a      hydr. no.=     0.0
  3 chemical elements
       1.000 u                   8.000 o               2.000 c
           temp=    298.150 k               press=      1.000 bars
         delg0=    -502.70  kcal/mol       delh0=    500.000 kcal/mol
             s0=      37.28  cal/mol/k        se=      0.000 cal/mol/k
             v0=      0.000 cc/mol            cp=      2.39  cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  uo2(co3)2--                      -2.000  h+
      1.000  uo2++                             2.000  hco3-
 t-p data grid is absent
 source=
* [lemire, 1988]
+-----------------------------------------------------------------------
uo2(co3)3----                                                 date= 06-Sep88
 entered by= j. bruno                        quality= good
 keys   =
 subsets= uranium
     charge=    -4.0           titr. factor=    6.0 eq/mol
  ion type=  3                      ion size=    0.0 a      hydr. no.=     0.0
  3 chemical elements
       1.000 u                  11.000 o               3.000 c
           temp=    298.150 k               press=      1.000 bars
         delg0=    -635.28  kcal/mol       delh0=    500.000 kcal/mol
             s0=       4.78  cal/mol/k        se=      0.000 cal/mol/k
             v0=      0.000 cc/mol            cp=     69.31  cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  uo2(co3)3----                    -3.000  h+
      1.000  uo2++                             3.000  hco3-
 t-p data grid is absent
 source=
* [lemire, 1988]
+-----------------------------------------------------------------------
uo2(oh)2                                                      date= 06-Sep88
 entered by= j. bruno                        quality= fair
 keys   =
 subsets= uranium
     charge=     0.0           titr. factor=    2.0 eq/mol
  ion type=  0                      ion size=    0.0 a      hydr. no.=     0.0
  3 chemical elements
       1.000 u                   4.000 o               2.000 h
           temp=    298.150 k               press=      1.000 bars
         delg0=    -324.81  kcal/mol       delh0=   -362.71  kcal/mol
             s0=    500.000 cal/mol/k         se=      0.000 cal/mol/k
             v0=      0.000 cc/mol            cp=            cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  uo2(oh)2                         -2.000  h+
      1.000  uo2++                             2.000  h2o
 t-p data grid is absent
 source=
* [lemire and tremaine, 1980], delh0 calculated from s0=14.1
+-----------------------------------------------------------------------
uo2(oh)4--                                                    date= 06-Sep88
 entered by= j. bruno                        quality= good
 keys   =
 subsets= uranium
     charge=    -2.0           titr. factor=    4.0 eq/mol
  ion type=  3                      ion size=    0.0 a      hydr. no.=     0.0
  3 chemical elements
       1.000 u                   6.000 o               4.000 h
           temp=    298.150 k               press=      1.000 bars
         delg0=    -411.09  kcal/mol       delh0=    500.000 kcal/mol
             s0=      20.32  cal/mol/k        se=      0.000 cal/mol/k
             v0=      0.000 cc/mol            cp=     44.93  cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  uo2(oh)4--                       -4.000  h+
      1.000  uo2++                             4.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988]
+-----------------------------------------------------------------------
uoh+++                                                        date= 06-Sep88
 entered by= j. bruno                        quality= good
 keys   =
 subsets= uranium
     charge=     3.0           titr. factor=    1.0 eq/mol
  ion type=  1                      ion size=    0.0 a      hydr. no.=     0.0
  3 chemical elements
       1.000 u                   1.000 o               1.000 h
           temp=    298.150 k               press=      1.000 bars
         delg0=    -182.64  kcal/mol       delh0=    500.000 kcal/mol
             s0=     -45.89  cal/mol/k        se=      4.36  cal/mol/k
             v0=      0.000 cc/mol            cp=    -12.91  cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  uoh+++                           -1.000  h+
      1.000  u++++                             1.000  h2o
 t-p data grid is absent
 source=
* delg0 from data of [grenthe et al., 1988b]; delg0 from log K4=-0.5
* [lemire and tremaine, 1980] give delg0=-182.60
* s0 from [lemire and tremaine, 1980]  uncertainty in s0 is +- 10
```

```
+-------------------------------------------------------------
u(oh)2++                                        date= 06-Sep88
 entered by= j.bruno                 quality= fair
 keys    =
 subsets= uranium
     charge=    2.0         titr. factor=    0.0 eq/mol
   ion type=  1                 ion size=    0.0 a     hydr. no.=    0.0
  3 chemical elements
      1.000 u             2.000 o               2.000 h
         temp=    298.150 k          press=      1.000 bars
        delg0=   -237.16  kcal/mol   delh0=    500.000 kcal/mol
           s0=    -16.5   cal/mol/k     se=      4.360 cal/mol/k
           v0=      0.000 cc/mol        cp=     -0.2   cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  u(oh)2++                      -2.000  h+
      1.000  u++++                          2.000  h2o
 t-p data grid is absent
 source=
* [puigdomenech and bruno, 1988]: delg0 from log K4=-2.1
* [lemire and tremaine, 1980] give delg0=-237.09
* s0 from [lemire and tremaine, 1980], uncertainty in s0 is +- 10
+-------------------------------------------------------------
u(oh)3+                                         date= 06-Sep88
 entered by= j.bruno                 quality= fair
 keys    =
 subsets= uranium
     charge=    1.0         titr. factor=    0.0 eq/mol
   ion type=  1                 ion size=    0.0 a     hydr. no.=    0.0
  3 chemical elements
      1.000 u             3.000 o               3.000 h
         temp=    298.150 k          press=      1.000 bars
        delg0=   -291.66  kcal/mol   delh0=    500.000 kcal/mol
           s0=      4.5   cal/mol/k     se=      4.360 cal/mol/k
           v0=      0.000 cc/mol        cp=     17.7   cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  u(oh)3+                       -3.000  h+
      1.000  u++++                          3.000  h2o
 t-p data grid is absent
 source=
* [puigdomenech and bruno, 1988]: delg0 from log K4=-3.7
* [lemire and tremaine, 1980] give delg0=-290.15
* s0 from [lemire and tremaine, 1980], uncertainty in s0 is +- 10
+-------------------------------------------------------------
u(oh)4                                          date= 29-Sep88
 entered by= i.puigdomenech          quality= good
 keys    =
 subsets= uranium
     charge=    0.0         titr. factor=    4.0 eq/mol
   ion type=  0                 ion size=    0.0 a     hydr. no.=    0.0
  3 chemical elements
      1.000 u             4.000 o               4.000 h
         temp=    298.150 k          press=      1.000 bars
        delg0=   -346.19  kcal/mol   delh0=   -398.80  kcal/mol
           s0=    500.000 cal/mol/k     se=      4.36  cal/mol/k
           v0=      0.000 cc/mol        cp=            cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  u(oh)4                        -4.000  h+
      1.000  u++++                          4.000  h2o
 t-p data grid is absent
 source=
* [puigdomenech and bruno, 1988]:
*       delg0 from log K4=-5.28; log K4 calculated from [lemire, 1988]
*       for reaction:   U++++  +  4H2O  ==  U(OH)4  +  4H+
*       s0 must be -4 to give UO2(s) solubility independent on T
*       delh0 calculated from s0=-4
* [lemire, 1988] gives delg0=-346.44, s0=+12.
+-------------------------------------------------------------
u(co3)5 (6-)                                    date= 06-Sep88
 entered by= j. bruno                quality= good
 keys    =
 subsets= uranium
     charge=   -6.0         titr. factor=   10.0 eq/mol
   ion type=  3                 ion size=    0.0 a     hydr. no.=    0.0
  3 chemical elements
      1.000 u            15.000 o               5.000 c
         temp=    298.150 k          press=      1.000 bars
        delg0=   -811.42  kcal/mol   delh0=    500.000 kcal/mol
           s0=     71.70  cal/mol/k     se=      4.36  cal/mol/k
           v0=      0.000 cc/mol        cp=    133.8   cal/mol/k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  u(co3)5 (6-)                  -5.000  h+
      1.000  u++++                          5.000  hco3-
 t-p data grid is absent
 source=
* [lemire, 1988]
+-------------------------------------------------------------
solids
+-------------------------------------------------------------
cauo4(c)                                        date= 06-Sep88
 entered by= i.puigdomench           quality= good
 keys    =
 subsets= uranium
  3 chemical elements
      1.000 ca            1.000 u               4.000 o
         temp=    298.150 k          press=      1.000 bars
        delg0=   -452.892 kcal/mol   delh0=    500.000 kcal/mol
           s0=     34.49  cal/mol/k     v0=      0.0   cc/mol
  1 heat capacity range(s) follow
      equation=  0                  cp or a=     27.629 cal/mol/k
            b=     11.19                 c=      0.000
            d=      0.000                e=      0.000
        limit=    600.000 k
  0 species in reference reaction
  5 species in data0 reaction
     -1.000  cauo4(c)                      -4.000  h+
     +1.000  ca++                          +1.000  uo2++
     +2.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988]
```

```
+---------------------------------------------------------------------
coffinite                     usio4(c)                 date= 06-Sep88
 entered by= i.puigdomenech            quality= good
 keys    =
 subsets= uranium          uo2sio4
  3 chemical elements
       1.000 u               1.000 si              4.000 o
          temp=    298.150 k          press=      1.000 bars
         delg0=   -450.048 kcal/mol   delh0=    500.000 kcal/mol
            s0=     27.96  cal/mol/k     v0=     46.120 cc/mol
  1 heat capacity range(s) follow
      equation=  0                  cp or a=     26.052 cal/mol/k
            b=      0.000                c=       0.000
            d=      0.000                e=       0.000
         limit=    600.000 k
  0 species in reference reaction
  5 species in data0 reaction
     -1.000  coffinite                    -4.000  h+
     +1.000  u++++                        +1.000  sio2(aq)
     +2.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988]
+---------------------------------------------------------------------
k2uo4(c)                                               date= 06-Sep88
 entered by= i.puigdomenech            quality= good
 keys    =
 subsets= uranium
  3 chemical elements
       2.000 k               1.000 u               4.000 o
          temp=    298.150 k          press=      1.000 bars
         delg0=   -427.605 kcal/mol   delh0=    500.000 kcal/mol
            s0=     43.093 cal/mol/k     v0=    0.0     cc/mol
  1 heat capacity range(s) follow
      equation=  0                  cp or a=     39.197 cal/mol/k
            b=      5.115                c=      -4.326
            d=      0.000                e=       0.000
         limit=    600.000 k
  0 species in reference reaction
  5 species in data0 reaction
     -1.000  k2uo4(c)                     -4.000  h+
     +2.000  k+                           +1.000  uo2++
     +2.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988]
+---------------------------------------------------------------------
mguo4(c)                                               date= 06-Sep88
 entered by= i.puigdomench             quality= good
 keys    =
 subsets= uranium
  3 chemical elements
       1.000 mg              1.000 u               4.000 o
          temp=    298.150 k          press=      1.000 bars
         delg0=   -418.093 kcal/mol   delh0=    500.000 kcal/mol
            s0=     31.53  cal/mol/k     v0=    0.0     cc/mol
  1 heat capacity range(s) follow
      equation=  0                  cp or a=     31.07  cal/mol/k
            b=      0.000                c=       0.000
            d=      0.000                e=       0.000
         limit=    600.000 k
  0 species in reference reaction
  5 species in data0 reaction
     -1.000  mguo4(c)                     -4.000  h+
     +1.000  mg++                         +1.000  uo2++
     +2.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988]
+---------------------------------------------------------------------
na2u2o7(c)                                             date= 06-Sep88
 entered by= j. bruno                  quality= good
 keys    =
 subsets= uranium
  3 chemical elements
       2.000 na              2.000 u               7.000 o
          temp=   298.1500 k          press=      1.000 bars
         delg0=   -716.54  kcal/mol   delh0=    500.000 kcal/mol
            s0=     62.38  cal/mol/k     v0=      0.000 cc/mol
  1 heat capacity range(s) follow
      equation=  0                  cp or a=     54.97  cal/mol/k
            b=      0.000                c=       0.000
            d=      0.000                e=       0.000
         limit=    600.000 k
  0 species in reference reaction
  5 species in data0 reaction
     -1.000  na2u2o7(c)                   -6.000  h+
     +2.000  na+                          +2.000  uo2++
     +3.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988]
+---------------------------------------------------------------------
na2uo4(c)                                              date= 06-Sep88
 entered by= j. bruno                  quality= good
 keys    =
 subsets= uranium
  3 chemical elements
       2.000 na              1.000 u               4.000 o
          temp=    298.150 k          press=      1.000 bars
         delg0=   -423.207 kcal/mol   delh0=    500.000 kcal/mol
            s0=     39.68  cal/mol/k     v0=     58.6   cc/mol
  1 heat capacity range(s) follow
      equation=  0                  cp or a=     38.848 cal/mol/k
            b=      6.188                c=      -5.012
            d=      0.000                e=       0.000
         limit=    600.000 k
  0 species in reference reaction
  5 species in data0 reaction
     -1.000  na2uo4(c)                    -4.000  h+
     +2.000  na+                          +1.000  uo2++
     +2.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988]
```

```
+----------------------------------------------------------------------
na3uo4(c)                                              date= 06-Sep88
 entered by= j. bruno                  quality= good
 keys   =
 subsets= uranium
  3 chemical elements
       3.000 na              1.000 u               4.000 o
          temp=    298.150 k          press=      1.000 bars
         delg0=   -453.609 kcal/mol   delh0=    500.000 kcal/mol
            s0=     47.371 cal/mol/k     v0=      0.000 cc/mol
  1 heat capacity range(s) follow
      equation=  0                  cp or a=     45.148 cal/mol/k
             b=      6.018                c=     -4.971
             d=      0.000                e=      0.000
         limit=    600.000 k
  0 species in reference reaction
  5 species in data0 reaction
     -1.000  na3uo4(c)                    -4.000  h+
     +3.000  na+                          +1.000  uo2+
     +2.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988]
+----------------------------------------------------------------------
nauo3(c)                                               date= 06-Sep88
 entered by= i.puigdomenech            quality= good
 keys   =
 subsets= uranium
  3 chemical elements
       1.000 na              1.000 u               3.000 o
          temp=   298.1500 k          press=      1.000 bars
         delg0=   -337.476 kcal/mol   delh0=    500.000 kcal/mol
            s0=     31.750 cal/mol/k     v0=      0.000 cc/mol
  1 heat capacity range(s) follow
      equation=  0                  cp or a=     26.052 cal/mol/k
             b=      0.000                c=      0.000
             d=      0.000                e=      0.000
         limit=    600.000 k
  0 species in reference reaction
  6 species in data0 reaction
     -1.000  nauo3(c)                     -3.000  h+
     -0.250  o2(g)                        +1.000  uo2++
     +1.500  h2o                          +1.000  na+
 t-p data grid is absent
 source=
* [lemire, 1988]
+----------------------------------------------------------------------
rutherfordine                 uo2co3(c)                date= 06-Sep88
 entered by= j. bruno                  quality= good
 keys   =
 subsets= uranium
  3 chemical elements
       1.000 u               1.000 c               5.000 o
          temp=    298.150 k          press=      1.000 bars
         delg0=   -373.566 kcal/mol   delh0=    500.000 kcal/mol
            s0=     46.367 cal/mol/k     v0=     57.7   cc/mol
  1 heat capacity range(s) follow
      equation=  0                  cp or a=     14.603 cal/mol/k
             b=     37.046                c=      4.517
             d=      0.000                e=      0.000
         limit=    600.000 k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  rutherfordine                -1.000  h+
     +1.000  uo2++                         1.000  hco3-
 t-p data grid is absent
 source=
* [lemire, 1988]
+----------------------------------------------------------------------
schoepite                     uo2(oh)2.h2o             date= 06-Sep88
 entered by= j. bruno                  quality= good
 keys   =
 subsets= uranium
  3 chemical elements
       1.000 u               5.000 o               4.000 h
          temp=    298.150 k          press=      1.000 bars
         delg0=   -390.06  kcal/mol   delh0=    500.000 kcal/mol
            s0=      40.87 cal/mol/k     v0=     66.7   cc/mol
  1 heat capacity range(s) follow
      equation=  0                  cp or a=     22.658 cal/mol/k
             b=     59.273                c=      6.692
             d=      0.000                e=      0.000
         limit=    600.000 k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  schoepite                    -2.000  h+
      3.000  h2o                           1.000  uo2++
 t-p data grid is absent
 source=
* [lemire and tremaine, 1980]
+----------------------------------------------------------------------
u3o7(c)                                                date= 06-Sep88
 entered by= j. bruno                  quality= good
 keys   =
 subsets= uranium
  2 chemical elements
       3.000 u               7.000 o
          temp=    298.150 k          press=      1.000 bars
         delg0=   -775.096 kcal/mol   delh0=    500.000 kcal/mol
            s0=      59.88 cal/mol/k     v0=     71.9   cc/mol
  1 heat capacity range(s) follow
      equation=  0                  cp or a=     46.131 cal/mol/k
             b=     35.700                c=     -4.696
             d=      0.000                e=      0.000
         limit=    600.000 k
  0 species in reference reaction
  5 species in data0 reaction
     -1.000  u3o7(c)                     -12.000  h+
      0.500  o2(g)                         3.000  u++++
      6.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988]
```

```
+-----------------------------------------------------------------------
u3o8(c,alph)                                           date= 06-Sep88
 entered by= j. bruno                  quality= good
 keys    =
 subsets= uranium
  2 chemical elements
       3.000 u               8.000 o
          temp=    298.150 k          press=       1.000 bars
         delg0=   -805.45  kcal/mol   delh0=     500.000 kcal/mol
            s0=     67.54  cal/mol/k     v0=     100.3   cc/mol
  1 heat capacity range(s) follow
      equation=  0                  cp or a=      79.565 cal/mol/k
            b=   -15.041                 c=     -16.276
            d=     0.000                 e=       0.000
         limit=    600.000 k
  0 species in reference reaction
  5 species in data0 reaction
     -1.000  u3o8(c,alph)                  -12.000  h+
      1.000  o2(g)                           3.000  u++++
      6.000  h2o
 t-p data grid is absent
 source=
* [lemire and tremaine, 1980]
* reaction changed from:
*          U3O8   +  6 H+   +  0.5 O2   =   3 UO2++   +  3 H2O
* to:      U3O8   + 12 H+   =  O2   +  3 U++++   +  6 H2O
* by I.Puigdomenech at Studsvik
+-----------------------------------------------------------------------
u4o9(c)                                                date= 06-Sep88
 entered by= j. bruno                  quality= fair
 keys    =
 subsets= uranium
  2 chemical elements
       4.000 u               9.000 o
          temp=    298.150 k          press=       1.000 bars
         delg0=  -1022.47  kcal/mol   delh0=     500.000 kcal/mol
            s0=     81.96  cal/mol/k     v0=      96.6   cc/mol
  1 heat capacity range(s) follow
      equation=  0                  cp or a=      60.492 cal/mol/k
            b=    33.819                 c=       1.230
            d=     0.000                 e=       0.000
         limit=    600.000 k
  0 species in reference reaction
  5 species in data0 reaction
     -1.000  u4o9(c)                       -16.000  h+
      4.000  u++++                           8.000  h2o
      0.500  o2(g)
 t-p data grid is absent
 source=
* [lemire and tremaine, 1980]
* [lemire and tremaine, 1980] note that u4o9 undergoes a lambda
* transition near 373 k. for the transition, they give values of
* delh = 0.67 kcal/mol and dels = 2.08 cal/mol/k .
+-----------------------------------------------------------------------
uo2(am)                                                date= 06-Sep88
 entered by= j. bruno                  quality= good
 keys    =
 subsets= uranium
  2 chemical elements
       1.000 u               2.000 o
          temp=    298.150 k          press=       1.000 bars
         delg0=   -238.82  kcal/mol   delh0=     500.000 kcal/mol
            s0=     18.4   cal/mol/k     v0=       0.000 cc/mol
  1 heat capacity range(s) follow
      equation=  0                  cp or a=      15.28  cal/mol/k
            b=     0.000                 c=       0.000
            d=     0.000                 e=       0.000
         limit=    600.000 k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  uo2(am)                        -4.000  h+
     +1.000  u++++                          +2.000  h2o
 t-p data grid is absent
 source=
* delg0 from solubility constant of [bruno et al., 1987]
*          and log K4 of [lemire, 1988]
* log Ks0= log Ks4 - log K4= -4.4 - (-5.28) = 0.88
* delgr=-(-0.88*1.3642)=1.2005
* delg0(uo2(am)) = delg0(u++++)+2(delg0(h2o))+degr
*                = -126.65+2(-56.686)+1.2005
* s0 = s0 for uraninite
* cp(T=298) = cp(T=298) for uraninite
+-----------------------------------------------------------------------
uo2(fuel)                                              date= 15-Sep88
 entered by= j. bruno                  quality= good
 keys    =
 subsets= uranium
  2 chemical elements
       1.000 u               2.000 o
          temp=    298.150 k          press=       1.000 bars
         delg0=   -242.64  kcal/mol   delh0=     500.000 kcal/mol
            s0=     18.4   cal/mol/k     v0=       0.000 cc/mol
  1 heat capacity range(s) follow
      equation=  0                  cp or a=      15.28  cal/mol/k
            b=     0.000                 c=       0.000
            d=     0.000                 e=       0.000
         limit=    600.000 k
  0 species in reference reaction
  4 species in data0 reaction
     -1.000  uo2(fuel)                      -4.000  h+
     +1.000  u++++                          +2.000  h2o
 t-p data grid is absent
 source=
* delg0 from solubility constant of [bruno et al., 1988]
*          and log K4 of [lemire, 1988]
* log Ks0= log Ks4 - log K4= -7.2 - (-5.28) = -1.92
* delgr=-(+1.92*1.3642)=-2.6193
* delg0(uo2(am)) = delg0(u++++)+2(delg0(h2o))+degr
*                = -126.65+2(-56.686)-2.6193
* s0 = s0 for uraninite
* cp(T=298) = cp(T=298) for uraninite
```

```
+-----------------------------------------------------------------------
uo2(oh)2(c,bet)                                         date= 06-Sep88
 entered by= j. bruno                     quality= good
 keys    =
 subsets= uranium
  3 chemical elements
       1.000 u            4.000 o                2.000 h
          temp=    298.150 k          press=      1.000 bars
         delg0=   -333.17  kcal/mol   delh0=    500.000 kcal/mol
            s0=     29.4   cal/mol/k     v0=      0.000 cc/mol
  1 heat capacity range(s) follow
     equation= 0                  cp or a=      9.990 cal/mol/k
            b=     47.801                c=     -8.437
            d=      0.000                e=      0.000
        limit=    600.000 k
  0 species in reference reaction
  4 species in data0 reaction
    -1.000  uo2(oh)2(c,bet)            -2.000  h+
     2.000  h2o                         1.000  uo2++
 t-p data grid is absent
 source=see notes
* [lemire and tremaine, 1980]
+-----------------------------------------------------------------------
uo3(c,gamma)                                            date= 06-Sep88
 entered by= j. bruno                     quality= good
 keys    =
 subsets= uranium
  2 chemical elements
       1.000 u            3.000 o
          temp=    298.150 k          press=      1.000 bars
         delg0=   -273.90  kcal/mol   delh0=    500.000 kcal/mol
            s0=     22.97  cal/mol/k     v0=     35.560 cc/mol
  1 heat capacity range(s) follow
     equation= 0                  cp or a=     22.108 cal/mol/k
            b=      2.641                c=     -2.653
            d=      0.000                e=      0.000
        limit=    600.000 k
  0 species in reference reaction
  4 species in data0 reaction
    -1.000  uo3(c,gamma)               -2.000  h+
    +1.000  uo2++                      +1.000  h2o
 t-p data grid is absent
 source=see notes
* [lemire and tremaine, 1980]
+-----------------------------------------------------------------------
uraninite                    uo2(c)                     date= 06-Sep88
 entered by= j. bruno                     quality= good
 keys    =
 subsets= uranium
  2 chemical elements
       1.000 u            2.000 o
          temp=    298.150 k          press=      1.000 bars
         delg0=   -246.61  kcal/mol   delh0=    500.000 kcal/mol
            s0=     18.411 cal/mol/k     v0=     24.618 cc/mol
  1 heat capacity range(s) follow
     equation= 0                  cp or a=     16.467 cal/mol/k
            b=      5.406                c=     -2.486
            d=      0.000                e=      0.000
        limit=    600.000 k
  0 species in reference reaction
  4 species in data0 reaction
    -1.000  uraninite                  -4.000  h+
     2.000  h2o                         1.000  u++++
 t-p data grid is absent
 source=see notes
* [lemire and tremaine, 1980]
+-----------------------------------------------------------------------
uranophane                   ca(uo2)2(sio3oh)2          date= 06-Sep88
 entered by= i.puigdomenech               quality= good
 keys    =
 subsets= uranium            uo2sio4
  5 chemical elements
       2.000 u            1.000 ca               2.000 si
      12.000 o            2.000 h
          temp=    298.150 k          press=      1.000 bars
         delg0=  -1189.054 kcal/mol   delh0=    500.000 kcal/mol
            s0=     83.652 cal/mol/k     v0=      0.000 cc/mol
  1 heat capacity range(s) follow
     equation= 0                  cp or a=     78.872 cal/mol/k
            b=      0.000                c=      0.000
            d=      0.000                e=      0.000
        limit=    600.000 k
  0 species in reference reaction
  6 species in data0 reaction
    -1.000  uranophane                 -6.000  h+
    +2.000  uo2++                      +1.000  ca++
    +2.000  sio2(aq)                   +4.000  h2o
 t-p data grid is absent
 source=
* [lemire, 1988]
+-----------------------------------------------------------------------
gases
+-----------------------------------------------------------------------
stop.
```

notation--
-- delg0 = gibbs energy of formation
-- delg0r = gibbs energy of reaction
-- delh0 = enthalpy of formation
-- delh0r = enthalpy of reaction
-- s0 = conventional entropy
-- se = internal electronic entropy (applies only to actinides and lanthanides)
-- dels0r = entropy of reaction

note that "500." is entered to mean "no data" for delg0, delh0, s0, log k, delh0, and dels0r. never enter zero or leave blank for these parameters unless you mean to use an actual value of zero.

note also that citations in comment lines in the data blocks must be enclosed in brackets, must begin and end in the same line, and must match exactly the citation patterns given in the references section.

ionic strength corrections noted in the comments for specific species of americium, barium and radium were made using the davies equation in the form:

```
log(gamma) = 0.51*(z**2)((sqrt(i)/(1+sqrt(i)))-(0.2*i))
```

where gamma is the activity coefficient, z is the charge, and i is the ionic strength.

data for aqueous species=

name= an alphabetic or alphanumeric string (maximum 18 characters)
entered by= name of person who entered data or last revised it
date= data of entering or revising data
source= alphanumeric string identifying a source of data. use "see below" or blank if more than one source is used or the pattern exceeds twelve characters. any other input must match one of the citation patterns in the references section. do not enclose the pattern here in brackets.
quality= alphanumeric string describing the probable error in gibbs energy or its equivalent, based on such information as might be available in individual cases. the quality parameter is intended primarily for flagging particularly good or bad data. it is not intended to be used for the purpose of certifying data for any use. the strings used must be chosen from among the following (in order of increasing value)

bad - demonstrably bad data- do not use except in illustrative calculations
poor - likely error exceeds a couple of kcal
restricted - ad hoc fit to a particular situation; see notes
speculative - species not shown conclusively to exist
uncertain - chosen rather arbitrarily from divergent reported values
unspecified - no quality information
(blank) - equivalent to unspecified
fair - likely error less than a couple of kcal
good - likely error less than about one kcal

the code must be modified to allow other quality parameter inputs. unrecognized inputs are set to "error". data for which the quality is "restricted", "speculative", or "uncertain" may numerically be "fair" to "good". even "good" data may not accurately predict solubilities in all cases, and "poor" data may be good enough for some applications. the user must always bear in mind the sensitivity of the results to the data, which may vary from one application to another. "unspecified" or blank is a perfectly acceptable input.

charge= electrical charge
titration factor= alkalin   weighting factor for ph 4.5 (methyl orange) alkalinity
ion type= criss-cobble ion type

0 neutral complex (may be left blank)
1 cation (may be left blank)
2 simple anion (may be left blank)
3 oxyanion
4 acid oxyanion

ion size= debye-huckel hydrated ion size (for activity coefficient estimation)
hydration number= number of bound water molecules not explicitly shown in the assumed molecular formula (for activity coefficient estimation)
temp= the base point temperature (k), usually 298.15 k
press= the base point pressure (bars), usually 1 bar
delg0= the apparent standard partial molal gibbs energy at the base point
delh0= the apparent standard partial molal enthalpy at the base point
s0= the conventional entropy at the base point
se= the internal electronic entropy; this function is zero except for some actinide and rare earth species (it is used in criss-cobble method temperature extrapolations)
v0= the partial molal volume at the base point; there is no current provision for using this input in the current mcrt (as of 12/16/82)
cp= the partial molal heat capacity at the base point; there is no current provision for using this input in the current mcrt (as of 12/16/82)
log k= log of the equilibrium constant of the given reaction at the base point
delh0r= enthalpy of the given reaction at the base point
dels0r= entropy of the given reaction at the base point

data for solid/gas species=

name= an alphabetic or alphanumeric string; e.g., calcite, bischofite. the formula may also be used as the name. (maximum 18 characters)
formula= an alphanumeric string denoting the formula of the species; e.g., caco3, mgcl2.6h2o. (maximum 18 characters)
entered by= name of person who entered data or last revised it
date= data of entering or revising data
source= alphanumeric string identifying a source of data. use "see below" or blank if more than one source is used or the pattern exceeds twelve characters. any other input must match one of the citation patterns in the references section. do not enclose the pattern here in brackets.
quality= alphanumeric string describing the quality of the data. not intended for use in "certifying" data. see remarks given above under "data for aqueous species".
number of chemical elements per mole
  number of moles of element per mole of substance
  symbol of chemical element
temp= the base point temperature (k), usually 298.15 k
press= the base point pressure (bars), usually 1 bar
delg0= the apparent standard partial molal gibbs energy at the base point
delh0= the apparent standard partial molal enthalpy at the base point
s0= the conventional entropy at the base point
v0= the molar volume at the base point
number of heat capacity ranges that follow
  equation= heat capacity equation code

```
0-  cp = a + 1.e-3*b*t + 1.e+5*c*t**(-2) + 1.e-6*d*t**2
1-  cp = a + 1.e-3*b*t + 1.e+5*c*t**(-2) + 1.e+8*d*t**(-3)
```

  cp= the molar heat capacity, assumed to be a constant
  a= first heat capacity coefficient
  b= second heat capacity coefficient
  c= third heat capacity coefficient

```
      d= fourth heat capacity coefficient
      e= fifth heat capacity coefficient
      limit= upper limit (k) of the preceding set of heat capacity parameters;
         no t-p data grid
        this is either the upper limit of use based on the range of measurement
        or the temperature of a phase transition
      delhtr= enthalpy of a phase transition, if any
      delstr= entropy of a phase transition, if any
    t-p data grid indication line, followed by t-p data grid, if any
      temp, press, delg0, delh0, s0, v0
    number of species appearing in a reaction, if a reaction follows
      number of moles of a species appearing in the reaction
      the name of the corresponding species
    log k= log of the equilibrium constant of the given reaction at the
      base point
    delh0r= enthalpy of the given reaction at the base point
    dels0r= entropy of the given reaction at the base point

note- "supcrt" refers to the supcrt code and data base (helgeson and
    kirkham, 1974, 1974b, 1976; helgeson et al., 1978; helgeson et al.,
    1982; and subsequent "supcrt update notices" from helgeson).

stop.
```

references

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stop.

APPENDIX B

Data blocks to be used in the DATA0 file of the EQ3/6 package. This data blocks were produced by the MCRT program with the thermodynamic data listed in Appendix A. The DATA0 file is reformatted into the DATA1, DATA2 and DATA3 files before its equilibrium constants may be used as input for the EQ3NR and EQ6 programs.

```
dfile
 mcrt.3245R65
+------------------------------------------------------------------------
u+++
    entered by= mcrt (see below)         date= 29Sep88
        source= mcrt.3245R65          quality= good
        charge=   3.0          titr. factor=   0.0 eq/mol
      ion size=   8.0 a        hydr. number=   0.0
     1 chemical elements=
       1.000 u
     5 species in reaction=
      -1.000 u+++            -1.000 h+              -0.250 o2(g)
       1.000 u++++            0.500 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
        33.3278   29.3961   24.9054   20.8326
        16.8620   13.7642   11.2796    9.2470
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000  500.0000  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*   (quality of u+++                data = good        )
+------------------------------------------------------------------------
uo2+
    entered by= mcrt (see below)         date= 29Sep88
        source= mcrt.3245R65          quality= fair
        charge=   1.0          titr. factor=   0.0 eq/mol
      ion size=   4.0 a        hydr. number=   0.0
     2 chemical elements=
       1.000 u                2.000 o
     5 species in reaction=
      -1.000 uo2+            -3.000 h+               1.000 u++++
       1.500 h2o              0.250 o2(g)
*    log k grid (0-25-60-100/150-200-250-300 c) =
       -13.1843  -13.1249  -13.0285  -12.9471
       -12.8892  -12.8649  -12.8916  -12.9605
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000  500.0000  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*   (quality of uo2+                data = fair        )
+------------------------------------------------------------------------
uo2++
    entered by= mcrt (see below)         date= 29Sep88
        source= mcrt.3245R65          quality= good
        charge=   2.0          titr. factor=   0.0 eq/mol
      ion size=   5.0 a        hydr. number=   0.0
     2 chemical elements=
       1.000 u                2.000 o
     5 species in reaction=
      -1.000 uo2++           -2.000 h+               1.000 u++++
       1.000 h2o              0.500 o2(g)
*    log k grid (0-25-60-100/150-200-250-300 c) =
       -34.7464  -32.4864  -29.8570  -27.4320
       -25.0208  -23.0980  -21.5425  -20.2615
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000  500.0000  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*   (quality of uo2++               data = good        )
+------------------------------------------------------------------------
u(co3)5 (6-)
    entered by= mcrt (see below)         date= 29Sep88
        source= mcrt.3245R65          quality= good
        charge=  -6.0          titr. factor=  10.0 eq/mol
      ion size=   4.0 a        hydr. number=   0.0
     3 chemical elements=
       1.000 u               15.000 o                5.000 c
     4 species in reaction=
      -1.000 u(co3)5 (6-)    -5.000 h+               1.000 u++++
       5.000 hco3-
*    log k grid (0-25-60-100/150-200-250-300 c) =
        14.2269   12.1965    9.3475    5.9097
         1.4007   -3.3301   -8.5497  -14.6916
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000  121.6304  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*   (quality of u(co3)5 (6-)        data = good        )
```

```
+----------------------------------------------------------------------
(uo2)2oh+++
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        charge=   3.0          titr. factor=   0.0 eq/mol
      ion size=   8.0 a        hydr. number=   0.0
     3 chemical elements=
       2.000 u                 5.000 o                  1.000 h
     4 species in reaction=
      -1.000 (uo2)2oh+++     -1.000 h+                  2.000 uo2++
       1.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
          2.9224     2.6675     2.4574     2.3823
          2.4502     2.6378     2.9070     3.2293
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000    18.0684   500.0000   500.0000
       500.0000   500.0000   500.0000   500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*    (quality of (uo2)2oh+++        data = good          )
+----------------------------------------------------------------------
(uo2)2(oh)2++
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        charge=   2.0          titr. factor=   0.0 eq/mol
      ion size=   5.0 a        hydr. number=   0.0
     3 chemical elements=
       2.000 u                 6.000 o                  2.000 h
     4 species in reaction=
      -1.000 (uo2)2(oh)2+    -2.000 h+                  2.000 uo2++
       2.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
          6.3860     5.6502     4.9122     4.3660
          3.9758     3.8063     3.7792     3.8469
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000    36.1368   500.0000   500.0000
       500.0000   500.0000   500.0000   500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*    (quality of (uo2)2(oh)2++      data = good          )
+----------------------------------------------------------------------
(uo2)3(co3)6 (6-)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        charge=  -6.0          titr. factor=  12.0 eq/mol
      ion size=   4.0 a        hydr. number=   0.0
     3 chemical elements=
       3.000 u                24.000 o                  6.000 c
     4 species in reaction=
      -1.000 (uo2)3(co3)6    -6.000 h+                  3.000 uo2++
       6.000 hco3-
*    log k grid (0-25-60-100/150-200-250-300 c) =
          8.8223     8.7015     8.1554     6.9727
          4.8237     2.0903    -1.4114    -6.1736
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000   145.9564   500.0000   500.0000
       500.0000   500.0000   500.0000   500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*    (quality of (uo2)3(co3)6 (6-)  data = good          )
+----------------------------------------------------------------------
(uo2)3(co3)(oh)3+
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        charge=   1.0          titr. factor=   5.0 eq/mol
      ion size=   4.0 a        hydr. number=   0.0
     4 chemical elements=
       3.000 u                12.000 o                  3.000 h
       1.000 c
     5 species in reaction=
      -1.000 (uo2)3(co3)(    -4.000 h+                  3.000 uo2++
       3.000 h2o              1.000 hco3-
*    log k grid (0-25-60-100/150-200-250-300 c) =
          8.9399     7.5494     6.0782     4.8720
          3.8216     3.1031     2.5431     1.9671
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000    78.5312   500.0000   500.0000
       500.0000   500.0000   500.0000   500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*    (quality of (uo2)3(co3)(oh)3+  data = good          )
+----------------------------------------------------------------------
(uo2)3(oh)4++
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        charge=   2.0          titr. factor=   0.0 eq/mol
      ion size=   5.0 a        hydr. number=   0.0
     3 chemical elements=
       3.000 u                10.000 o                  4.000 h
     4 species in reaction=
      -1.000 (uo2)3(oh)4+    -4.000 h+                  3.000 uo2++
       4.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
         13.3307    11.9014    10.4707     9.4125
          8.6557     8.3260     8.2711     8.3984
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000    72.2736   500.0000   500.0000
       500.0000   500.0000   500.0000   500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*    (quality of (uo2)3(oh)4++      data = good          )
+----------------------------------------------------------------------
(uo2)3(oh)5+
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        charge=   1.0          titr. factor=   0.0 eq/mol
      ion size=   4.0 a        hydr. number=   0.0
     3 chemical elements=
       3.000 u                11.000 o                  5.000 h
     4 species in reaction=
      -1.000 (uo2)3(oh)5+    -5.000 h+                  3.000 uo2++
       5.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
         17.2815    15.5071    13.6975    12.2903
         11.1755    10.5493    10.2311    10.1161
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000    90.3419   500.0000   500.0000
       500.0000   500.0000   500.0000   500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*    (quality of (uo2)3(oh)5+       data = good          )
```

```
+-----------------------------------------------------------------------
(uo2)3(oh)7-
    entered by= mcrt (see below)           date= 29Sep88
        source= mcrt.3245R65            quality= good
        charge=  -1.0           titr. factor=   7.0 eq/mol
      ion size=   4.0 a         hydr. number=   0.0
     3 chemical elements=
       3.000 u                13.000 o                 7.000 h
     4 species in reaction=
      -1.000 (uo2)3(oh)7-     -7.000 h+                3.000 uo2++
       7.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
        36.1404   32.4012   28.2953   24.6929
        21.3045   18.7558   16.7165   15.0361
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000  126.4787  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*    (quality of (uo2)3(oh)7-       data = good         )
+-----------------------------------------------------------------------
(uo2)4(oh)7+
    entered by= mcrt (see below)           date= 29Sep88
        source= mcrt.3245R65            quality= good
        charge=   1.0           titr. factor=   7.0 eq/mol
      ion size=   4.0 a         hydr. number=   0.0
     3 chemical elements=
       4.000 u                15.000 o                 7.000 h
     4 species in reaction=
      -1.000 (uo2)4(oh)7+     -7.000 h+                4.000 uo2++
       7.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
        24.4670   21.8023   19.0809   16.9756
        15.3311   14.4345   14.0154   13.9146
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000  126.4787  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*    (quality of (uo2)4(oh)7+       data = good         )
+-----------------------------------------------------------------------
uo2co3
    entered by= mcrt (see below)           date= 03Oct88
        source= mcrt.3245R65            quality= good
        charge=   0.0           titr. factor=   2.0 eq/mol
      ion size=   4.0 a         hydr. number=   0.0
     3 chemical elements=
       1.000 u                 1.000 c                 5.000 o
     4 species in reaction=
      -1.000 uo2co3           -1.000 h+                1.000 uo2++
       1.000 hco3-
*    log k grid (0-25-60-100/150-200-250-300 c) =
         1.2678    0.8150    0.2317   -0.4002
        -1.1886   -2.0418  500.0000  500.0000
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000   24.3261  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   03Oct88 , dquant method
*    (quality of uo2co3             data = good         )
+-----------------------------------------------------------------------
uo2(co3)2--
    entered by= mcrt (see below)           date= 29Sep88
        source= mcrt.3245R65            quality= good
        charge=  -2.0           titr. factor=   4.0 eq/mol
      ion size=   4.0 a         hydr. number=   0.0
     3 chemical elements=
       1.000 u                 8.000 o                 2.000 c
     4 species in reaction=
      -1.000 uo2(co3)2--      -2.000 h+                1.000 uo2++
       2.000 hco3-
*    log k grid (0-25-60-100/150-200-250-300 c) =
         4.6817    4.0488    3.2704    2.4460
         1.4461    0.4444   -0.6687   -2.0944
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000   48.6521  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*    (quality of uo2(co3)2--        data = good         )
+-----------------------------------------------------------------------
uo2(co3)3----
    entered by= mcrt (see below)           date= 29Sep88
        source= mcrt.3245R65            quality= good
        charge=  -4.0           titr. factor=   6.0 eq/mol
      ion size=   4.0 a         hydr. number=   0.0
     3 chemical elements=
       1.000 u                11.000 o                 3.000 c
     4 species in reaction=
      -1.000 uo2(co3)3---     -3.000 h+                1.000 uo2++
       3.000 hco3-
*    log k grid (0-25-60-100/150-200-250-300 c) =
         9.7265    9.6942    9.5933    9.3216
         8.7272    7.9143    6.7982    5.1043
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000   72.9782  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*    (quality of uo2(co3)3----      data = good         )
+-----------------------------------------------------------------------
uo2oh+
    entered by= mcrt (see below)           date= 29Sep88
        source= mcrt.3245R65            quality= good
        charge=   1.0           titr. factor=   1.0 eq/mol
      ion size=   4.0 a         hydr. number=   0.0
     3 chemical elements=
       1.000 u                 3.000 o                 1.000 h
     4 species in reaction=
      -1.000 uo2oh+           -1.000 h+                1.000 uo2++
       1.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
         5.9824    5.2762    4.4903    3.7847
         3.0975    2.5611    2.1211    1.7490
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000   18.0684  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*    (quality of uo2oh+             data = good         )
```

```
+-----------------------------------------------------------------
uo2(oh)2
    entered by= mcrt (see below)        date= 29Sep88
       source= mcrt.3245R65          quality= fair
       charge=   0.0              titr. factor=   2.0 eq/mol
     ion size=   4.0 a            hydr. number=   0.0
    3 chemical elements=
      1.000 u                    4.000 o                   2.000 h
    4 species in reaction=
     -1.000 uo2(oh)2        -2.000 h+                    1.000 uo2++
      2.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
       13.0690    11.8952    10.5482     9.2919
        8.0211     6.9710   500.0000   500.0000
*    delvr grid (0-25-60-100/150-200-250-300 c) =
      500.0000    36.1368   500.0000   500.0000
      500.0000   500.0000   500.0000   500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , dquant method
*   (quality of uo2(oh)2             data = fair        )
+-----------------------------------------------------------------
uo2(oh)4--
    entered by= mcrt (see below)        date= 29Sep88
       source= mcrt.3245R65          quality= good
       charge=  -2.0              titr. factor=   4.0 eq/mol
     ion size=   4.0 a            hydr. number=   0.0
    3 chemical elements=
      1.000 u                    6.000 o                   4.000 h
    4 species in reaction=
     -1.000 uo2(oh)4--      -4.000 h+                    1.000 uo2++
      4.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
       34.3000    31.7579    29.0780    26.8552
       24.8878    23.5519    22.6461    22.0398
*    delvr grid (0-25-60-100/150-200-250-300 c) =
      500.0000    72.2736   500.0000   500.0000
      500.0000   500.0000   500.0000   500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*   (quality of uo2(oh)4--           data = good        )
+-----------------------------------------------------------------
uoh+++
    entered by= mcrt (see below)        date= 29Sep88
       source= mcrt.3245R65          quality= good
       charge=   3.0              titr. factor=   1.0 eq/mol
     ion size=   8.0 a            hydr. number=   0.0
    3 chemical elements=
      1.000 u                    1.000 o                   1.000 h
    4 species in reaction=
     -1.000 uoh+++          -1.000 h+                    1.000 u++++
      1.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
        1.3152     0.5125    -0.3647    -1.1151
       -1.7909    -2.2681    -2.6157    -2.8705
*    delvr grid (0-25-60-100/150-200-250-300 c) =
      500.0000    18.0684   500.0000   500.0000
      500.0000   500.0000   500.0000   500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*   (quality of uoh+++               data = good        )
+-----------------------------------------------------------------
u(oh)2++
    entered by= mcrt (see below)        date= 29Sep88
       source= mcrt.3245R65          quality= fair
       charge=   2.0              titr. factor=   0.0 eq/mol
     ion size=   5.0 a            hydr. number=   0.0
    3 chemical elements=
      1.000 u                    2.000 o                   2.000 h
    4 species in reaction=
     -1.000 u(oh)2++        -2.000 h+                    1.000 u++++
      2.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
        3.3165     2.1025     0.7796    -0.3627
       -1.4126    -2.1753    -2.7563    -3.2095
*    delvr grid (0-25-60-100/150-200-250-300 c) =
      500.0000    36.1368   500.0000   500.0000
      500.0000   500.0000   500.0000   500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*   (quality of u(oh)2++             data = fair        )
+-----------------------------------------------------------------
u(oh)3+
    entered by= mcrt (see below)        date= 29Sep88
       source= mcrt.3245R65          quality= fair
       charge=   1.0              titr. factor=   0.0 eq/mol
     ion size=   4.0 a            hydr. number=   0.0
    3 chemical elements=
      1.000 u                    3.000 o                   3.000 h
    4 species in reaction=
     -1.000 u(oh)3+         -3.000 h+                    1.000 u++++
      3.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
        5.1644     3.7072     2.1252     0.7492
       -0.5368    -1.4921    -2.2456    -2.8599
*    delvr grid (0-25-60-100/150-200-250-300 c) =
      500.0000    54.2052   500.0000   500.0000
      500.0000   500.0000   500.0000   500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , criss-cobble method
*   (quality of u(oh)3+              data = fair        )
+-----------------------------------------------------------------
u(oh)4
    entered by= mcrt (see below)        date= 29Sep88
       source= mcrt.3245R65          quality= good
       charge=   0.0              titr. factor=   4.0 eq/mol
     ion size=   4.0 a            hydr. number=   0.0
    3 chemical elements=
      1.000 u                    4.000 o                   4.000 h
    4 species in reaction=
     -1.000 u(oh)4          -4.000 h+                    1.000 u++++
      4.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
        6.3141     5.2899     4.0185     2.7170
        1.1976    -0.3328   500.0000   500.0000
*    delvr grid (0-25-60-100/150-200-250-300 c) =
      500.0000    72.2736   500.0000   500.0000
      500.0000   500.0000   500.0000   500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , dquant method
*   (quality of u(oh)4               data = good        )
```

```
+-----------------------------------------------------------------------
cauo4(c)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=     0.000 cc/mol
     3 chemical elements=
       1.000 ca              1.000 u                 4.000 o
     5 species in reaction=
      -1.000 cauo4(c)       -4.000 h+                1.000 ca++
       1.000 uo2++           2.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
        16.9959   14.8806   12.4546   10.2694
         8.1446    6.4783    5.0866    3.7822
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000   17.6505  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of cauo4(c)           data = good          )
+-----------------------------------------------------------------------
coffinite           usio4(c)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=    46.120 cc/mol
     3 chemical elements=
       1.000 u               1.000 si                4.000 o
     5 species in reaction=
      -1.000 coffinite      -4.000 h+                1.000 u++++
       1.000 sio2(aq)        2.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
        -7.0971   -7.9379   -8.9396   -9.8715
       -10.7879  -11.4870  -12.0480  -12.5178
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000    3.6660  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of coffinite          data = good          )
+-----------------------------------------------------------------------
k2uo4(c)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=     0.000 cc/mol
     3 chemical elements=
       2.000 k               1.000 u                 4.000 o
     5 species in reaction=
      -1.000 k2uo4(c)       -4.000 h+                2.000 k+
       1.000 uo2++           2.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
        38.5739   35.6123   32.2376   29.1919
        26.2306   23.9293   22.0755   20.5098
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000   53.6002  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of k2uo4(c)           data = good          )
+-----------------------------------------------------------------------
mguo4(c)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=     0.000 cc/mol
     3 chemical elements=
       1.000 mg              1.000 u                 4.000 o
     5 species in reaction=
      -1.000 mguo4(c)       -4.000 h+                1.000 mg++
       1.000 uo2++           2.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
        26.4006   23.1955   19.5196   16.1989
        12.9601   10.4126    8.2948    6.3525
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000   14.8704  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of mguo4(c)           data = good          )
+-----------------------------------------------------------------------
na2u2o7(c)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=     0.000 cc/mol
     3 chemical elements=
       2.000 na              2.000 u                 7.000 o
     5 species in reaction=
      -1.000 na2u2o7(c)     -6.000 h+                2.000 na+
       2.000 uo2++           3.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
        27.9223   24.9886   21.6866   18.7535
        15.9538   13.8328   12.1569   10.7386
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000   51.6746  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of na2u2o7(c)         data = good          )
+-----------------------------------------------------------------------
na2uo4(c)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=    58.600 cc/mol
     3 chemical elements=
       2.000 na              1.000 u                 4.000 o
     5 species in reaction=
      -1.000 na2uo4(c)      -4.000 h+                2.000 na+
       1.000 uo2++           2.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
        34.5179   31.5736   28.2531   25.2993
        22.4837   20.3463   18.6607   17.2448
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000  -24.9938  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of na2uo4(c)          data = good          )
```

```
+----------------------------------------------------------------------
na3uo4(c)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= fair
        volume=      0.000 cc/mol
     3 chemical elements=
       3.000 na              1.000 u               4.000 o
     5 species in reaction=
      -1.000 na3uo4(c)      -4.000 h+              3.000 na+
       1.000 uo2+            2.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
        61.3714   56.6050   51.2256   46.4620
        41.9606   38.5754   35.9435   33.7679
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000   32.3409  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of na3uo4(c)          data = good        )
+----------------------------------------------------------------------
nauo3(c)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=      0.000 cc/mol
     3 chemical elements=
       1.000 na              1.000 u               3.000 o
     6 species in reaction=
      -1.000 nauo3(c)       -3.000 h+             -0.250 o2(g)
       1.000 uo2++           1.500 h2o             1.000 na+
*    log k grid (0-25-60-100/150-200-250-300 c) =
        30.8538   27.7360   24.1892   20.9905
        17.8849   15.4780   13.5501   11.9404
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000  500.0000  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of nauo3(c)           data = good        )
+----------------------------------------------------------------------
rutherfordine    uo2co3(c)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=     57.700 cc/mol
     3 chemical elements=
       1.000 u               1.000 c               5.000 o
     4 species in reaction=
      -1.000 rutherfordin   -1.000 h+              1.000 uo2++
       1.000 hco3-
*    log k grid (0-25-60-100/150-200-250-300 c) =
        -3.6057   -4.1224   -4.8021   -5.5260
        -6.3794   -7.1965   -8.0300   -8.9867
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000  -33.3739  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of rutherfordine      data = good        )
+----------------------------------------------------------------------
schoepite          uo2(oh)2.h2o
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=     66.700 cc/mol
     3 chemical elements=
       1.000 u               5.000 o               4.000 h
     4 species in reaction=
      -1.000 schoepite      -2.000 h+              3.000 h2o
       1.000 uo2++
*    log k grid (0-25-60-100/150-200-250-300 c) =
         6.4288    5.6203    4.7167    3.9115
         3.1312    2.5316    2.0543    1.6644
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000  -12.4948  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of schoepite          data = good        )
+----------------------------------------------------------------------
u3o7(c)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=     71.900 cc/mol
     2 chemical elements=
       3.000 u               7.000 o
     5 species in reaction=
      -1.000 u3o7(c)       -12.000 h+              0.500 o2(g)
       3.000 u++++           6.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
       -39.2777  -40.3220  -41.3939  -42.3035
       -43.1613  -43.7895  -44.3231  -44.8024
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000  500.0000  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of u3o7(c)            data = good        )
+----------------------------------------------------------------------
u3o8(c,alph)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=    100.300 cc/mol
     2 chemical elements=
       3.000 u               8.000 o
     5 species in reaction=
      -1.000 u3o8(c,alph)  -12.000 h+              1.000 o2(g)
       3.000 u++++           6.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
       -63.9011  -62.5710  -60.9215  -59.3526
       -57.7776  -56.4898  -55.4710  -54.6611
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000  500.0000  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of u3o8(c,alph)       data = good        )
```

```
+-----------------------------------------------------------------------
u4o9(c)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= fair
        volume=    96.600 cc/mol
     2 chemical elements=
       4.000 u               9.000 o
     5 species in reaction=
      -1.000 u4o9(c)       -16.000 h+              4.000 u++++
       8.000 h2o             0.500 o2(g)
*    log k grid (0-25-60-100/150-200-250-300 c) =
       -43.3751  -45.7061  -48.2228  -50.4346
       -52.5628  -54.1728  -55.5026  -56.6443
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000  500.0000  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of u4o9(c)            data = fair          )
+-----------------------------------------------------------------------
uo2(am)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=     0.000 cc/mol
     2 chemical elements=
       1.000 u               2.000 o
     4 species in reaction=
      -1.000 uo2(am)        -4.000 h+              1.000 u++++
       2.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
         2.6777    0.8858   -1.1271   -2.9382
        -4.7023   -6.0710   -7.1815   -8.1078
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000   36.1368  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of uo2(am)            data = good          )
+-----------------------------------------------------------------------
uo2(fuel)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=     0.000 cc/mol
     2 chemical elements=
       1.000 u               2.000 o
     4 species in reaction=
      -1.000 uo2(fuel)      -4.000 h+              1.000 u++++
       2.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
        -0.3786   -1.9142   -3.6329   -5.1754
        -6.6751   -7.8354   -8.7773   -9.5644
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000   36.1368  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of uo2(fuel)          data = good          )
+-----------------------------------------------------------------------
uo2(oh)2(c,bet)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=     0.000 cc/mol
     3 chemical elements=
       1.000 u               4.000 o               2.000 h
     4 species in reaction=
      -1.000 uo2(oh)2(c,b   -2.000 h+              2.000 h2o
       1.000 uo2++
*    log k grid (0-25-60-100/150-200-250-300 c) =
         6.7082    5.7674    4.7461    3.8652
         3.0379    2.4182    1.9314    1.5320
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000   36.1368  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of uo2(oh)2(c,bet)    data = good          )
+-----------------------------------------------------------------------
uo3(c,gamma)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=    35.560 cc/mol
     2 chemical elements=
       1.000 u               3.000 o
     4 species in reaction=
      -1.000 uo3(c,gamma)   -2.000 h+              1.000 uo2++
       1.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
         8.9581    7.6591    6.1741    4.8164
         3.4684    2.4017    1.5332    0.8105
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000  -17.4916  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of uo3(c,gamma)       data = good          )
+-----------------------------------------------------------------------
uraninite           uo2(c)
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=    24.618 cc/mol
     2 chemical elements=
       1.000 u               2.000 o
     4 species in reaction=
      -1.000 uraninite      -4.000 h+              2.000 h2o
       1.000 u++++
*    log k grid (0-25-60-100/150-200-250-300 c) =
        -3.5545   -4.8242   -6.2378   -7.5036
        -8.7344   -9.6863  -10.4621  -11.1129
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000   11.5188  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of uraninite          data = good          )
```

```
+-----------------------------------------------------------------------
uranophane        ca(uo2)2(sio3oh)2
    entered by= mcrt (see below)        date= 29Sep88
        source= mcrt.3245R65         quality= good
        volume=     0.000 cc/mol
     5 chemical elements=
       2.000 u               1.000 ca              2.000 si
      12.000 o               2.000 h
     6 species in reaction=
      -1.000 uranophane     -6.000 h+              2.000 uo2++
       1.000 ca++            2.000 sio2(aq)        4.000 h2o
*    log k grid (0-25-60-100/150-200-250-300 c) =
        19.7289   17.2675   14.3156   11.5936
         8.9297    6.8671    5.1823    3.6405
*    delvr grid (0-25-60-100/150-200-250-300 c) =
       500.0000   81.0857  500.0000  500.0000
       500.0000  500.0000  500.0000  500.0000
* mcrt file mdas.3245s01, rev.   29Sep88 , heat capacity integration
*   (quality of uranophane        data = good          )
+-----------------------------------------------------------------------
stop.

 there were
       0 errors encountered and
       9 warnings about g-h-s inconsistencies
```

List of SKB reports

Annual Reports

1977–78
TR 121
KBS Technical Reports 1 – 120.
Summaries. Stockholm, May 1979.

1979
TR 79–28
The KBS Annual Report 1979.
KBS Technical Reports 79-01 – 79-27.
Summaries. Stockholm, March 1980.

1980
TR 80–26
The KBS Annual Report 1980.
KBS Technical Reports 80-01 – 80-25.
Summaries. Stockholm, March 1981.

1981
TR 81–17
The KBS Annual Report 1981.
KBS Technical Reports 81-01 – 81-16.
Summaries. Stockholm, April 1982.

1982
TR 82–28
The KBS Annual Report 1982.
KBS Technical Reports 82-01 – 82-27.
Summaries. Stockholm, July 1983.

1983
TR 83–77
The KBS Annual Report 1983.
KBS Technical Reports 83-01 – 83-76
Summaries. Stockholm, June 1984.

1984
TR 85–01
Annual Research and Development Report 1984
Including Summaries of Technical Reports Issued during 1984. (Technical Reports 84-01 – 84-19)
Stockholm June 1985.

1985
TR 85-20
Annual Research and Development Report 1985
Including Summaries of Technical Reports Issued during 1985. (Technical Reports 85-01-85-19)
Stockholm May 1986.

1986
TR 86-31
SKB Annual Report 1986
Including Summaries of Technical Reports Issued during 1986
Stockholm, May 1987

1987
TR 87-33
SKB Annual Report 1987
Including Summaries of Technical Reports Issued during 1987
Stockholm, May 1988

Technical Reports

1988

TR 88-01
Preliminary investigations of deep ground water microbiology in Swedish granitic rocks
Karsten Pedersen
University of Göteborg
December 1987

TR 88-02
Migration of the fission products strontium, technetium, iodine, cesium and the actinides neptunium, plutonium, americium in granitic rock
Thomas Ittner[1], Börje Torstenfelt[1], Bert Allard[2]
[1]Chalmers University of Technology
[2]University of Linköping
January 1988

TR 88-03
Flow and solute transport in a single fracture. A two-dimensional statistical model
Luis Moreno[1], Yvonne Tsang[2], Chin Fu Tsang[2], Ivars Neretnieks[1]
[1]Royal Institute of Technology, Stockholm, Sweden
[2]Lawrence Berkeley Laboratory, Berkeley, CA, USA
January 1988

TR 88-04
Ion binding by humic and fulvic acids: A computational procedure based on functional site heterogeneity and the physical chemistry of polyelectrolyte solutions
J A Marinsky, M M Reddy, J Ephraim, A Mathuthu
US Geological Survey, Lakewood, CA, USA
Linköping University, Linköping
State University of New York at Buffalo, Buffalo, NY, USA
April 1987

TR 88-05
Description of geophysical data on the SKB database GEOTAB
Stefan Sehlstedt
Swedish Geological Co, Luleå
February 1988

TR 88-06
Description of geological data in SKBs database GEOTAB
Tomas Stark
Swedish Geological Co, Luleå
April 1988

TR 88-07
Tectonic studies in the Lansjärv region
Herbert Henkel
Swedish Geological Survey, Uppsala
October 1987

TR 88-08
Diffusion in the matrix of granitic rock.
Field test in the Stripa mine. Final report.
Lars Birgersson, Ivars Neretnieks
Royal Institute of Technology, Stockholm
April 1988

TR 88-09
The kinetics of pitting corrosion of carbon steel. Progress report to June 1987
G P Marsh, K J Taylor, Z Sooi
Materials Development Division
Harwell Laboratory
February 1988

TR 88-10
GWHRT – A flow model for coupled groundwater and heat flow
Version 1.0
Roger Thunvik[1], Carol Braester[2]
[1] Royal Institute of Technology, Stockholm
[2] Israel Institute of Technology, Haifa
April 1988

TR 88-11
Groundwater numerical modelling of the Fjällveden study site – Evaluation of parameter variations
A hydrocoin study – Level 3, case 5A
Nils-Åke Larsson[1], Anders Markström[2]
[1] Swedish Geological Company, Uppsala
[2] Kemakta Consultants Co, Stockholm
October 1987

TR 88-12
Near-distance seismological monitoring of the Lansjärv neotectonic fault region
Rutger Wahlström, Sven-Olof Linder,
Conny Holmqvist
Seismological Depertment, Uppsala University,
Uppsala
May 1988

TR 88-13
Validation of the rock mechanics HNFEMP code against Colorado school of mines block test data
Ove Stephansson, Tomas Savilahti
University of Luleå, Luleå
May 1988

TR 88-14
Validation of MUDEC against Colorado school of mines block test data
Nick Barton, Panayiotis Chryssanthakis,
Karstein Monsen
Norges Geotekniske Institutt, Oslo, Norge
April 1988

TR 88-15
Hydrothermal effects on montmorillonite.
A preliminary study
Roland Pusch
Ola Karnland
June 1988

TR 88-16
Swedish Hard Rock Laboratory
First evaluation of preinvestigations 1986-87 and target area characterization
Gunnar Gustafson
Roy Stanfors
Peter Wikberg
June 1988

TR 88-17
On the corrosion of copper in pure water
T E Eriksen[1], P Ndalamba[1], I Grenthe[2]
[1] The Royal Institute of Technology, Stockholm
Department of nuclear chemistry
[2] The Royal Institute of Technology, Stockholm
Department of inorganic chemistry
March 1988

TR 88-18
Geochemical modelling of the evolution of a granite-concrete-water system around a repository for spent nuclear fuel
Bertrand Fritz, Benoit Madé, Yves Tardy
Université Louis Pasteur de Strasbourg
April 1988

TR 88-19
A Bayesian nonparametric estimation of distributions and quantiles
Kurt Pörn
Studsvik AB
November 1988

TR 88-20

Creep properties of welded joints in OFHC copper for nuclear waste containment

Bo Ivarsson, Jan-Olof Österberg
Swedish Institute for Metals Research
August 1988