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Precipitation and Hydrolysis of Uranium(VI) in Aqueous Solutions. III. Uranyl Nitrate — Sodium Carbonate — Strontium Chloride*

H. Füredi**

Department of Physical Chemistry, Institute »Ruđer Bošković«, Zagreb, Croatia, Yugoslavia

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In the precipitation system uranyl nitrate — sodium carbonate — strontium chloride the influence of changes in concentration of uranyl nitrate on the precipitation of uranium was studied. The »concentration maximum« was found to occur when the equivalent concentrations of the precipitating components are satisfying the following expressions:

$[Na_2CO_3]$		1.5	$[UO_2(NO_3)_2]$	(1)
$[SrCl_2]$	>	0.7	$[UO_2(NO_3)_2] - 1.36$	(2)

The final pH values of the systems were about 6. It was shown that expression (1) is valid in the range of $1 \times 10^{-1} N$ to $1 \times 10^{-4} N$ of uranyl nitrate, while expression (2) is valid in the range of $1 \times 10^{-1} N$ to $2 \times 10^{-3} N$ of uranyl nitrate.

The coprecipitation of uranium with strontium carbonate exceeded $85^{0/0}$ when the following relations existed between the equivalent concentrations of the precipitating components:

$[SrCl_2]$	\gg	$[Na_2]$	$CO_3]$		(3)
$[Na_2CO_3]$	≫	0.76	$[UO_2(NO_3)_2] +$	0.7	(4)

Expression (4) was found to be valid in the range of $6 \times 10^{-2} N$ to $2 \times 10^{-4} N$ of uranyl nitrate. The region where more than $85^{\circ}/_{0}$ of uranium was coprecipitated was represented by means of a precipitation body in a coordination system of the concentrations of strontium chloride (x — axis), sodium carbonate (y — axis) and uranyl nitrate (z — axis).

INTRODUCTION

Several authors have reported on the chemistry of soluble²⁻¹² and sparingly soluble^{5,12,13} complex uranyl carbonates and the interaction of uranyl nitrate with sodium carbonate in aqueous solution^{10,14}.

Alkaline earth carbonates are well known as sparingly soluble^{15,16}, crystalline compounds. The amount of their precipitation or dissolution in aqueous

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solution is directed by the equilibrium existing between undissolved alkaline earth carbonates and the ions in the solution in contact with the precipitate (Me²⁺, OH⁻, H⁺, HCO₃⁻, CO₃²⁻; Me²⁺ = Ba²⁺, Sr²⁺, Ca²⁺, Mg²⁺). This equilibrium was frequently calculated especially for natural waters, containing undissolved calcium carbonates^{18,19}.

The mutual interaction of uranyl nitrate, sodium carbonate and alkaline earth chlorides in aqueous solutions was investigated, in order to show the influence of changes in concentrations of the reactants on the precipitation and dissolution of uranium. In a previous paper¹ the influence of changes in concentrations of sodium carbonate and barium, strontium, calcium and magnesium chlorides was already shown by means of precipitation bodies and contour diagrams and the particular precipitation systems were compared. For reasons of simplicity uranyl nitrate was kept at constant concentration $(2 \times 10^{-3} N)$. In the system with strontium chloride the characteristic regions of precipitation, coprecipitation and dissolution of uranium were found to be well developed and good reproducibility of the results was obtained. Therefore this system was chosen as a model to evaluate additionally the influence of changes in the concentration of uranyl nitrate.

Using these and previously published results^{1,20,21}, an overall survey of the phenomenology of the precipitation and dissolution of uranium in the three component precipitation system uranyl nitrate — sodium carbonate — strontium chloride could be obtained.

Considering that the knowledge of the systems investigated may be important in uranium technology (carbonate leaching of uranium in the presence of alkaline $earths^{22}$ etc.), an attempt was made to present the results in a simple way, in order to facilitate their use. The method for the presentation of two-component precipitation systems by means of three-dimensional diagrams was therefore adapted for the presentation of three-component systems. Some simple empirical expressions, giving the relations of the concentrations of the precipitating components in the region of precipitation and coprecipitation of uranium, were also derived.

EXPERIMENTAL

Analytical grade chemicals and bidistilled water were used for the preparation of standard solutions and samples. The concentrations of the 2N standard solutions of strontium chloride were determined by the ion exchange (Dowex — 50 X-8, 50/100 mesh) technique²³, while the standard solutions of uranium (2N uranyl nitrate) were analysed gravimetrically with 8-oxyquinoline²⁴. The precipitation systems were prepared, and the precipitate separated as described previously^{1,21}. In an aliquot of the clear mother liquor the uranium content was determined spectrophotometrically²⁵. The pH of the samples was determined with a glass electrode and a calomel electrode using a Pye pH meter.

Turbidimetric measurements were performed by means of a Zeiss tyndallometer in connection with a Pulfrich photometer.

RESULTS

According to previous results¹, obtained at constant concentration $(2 \times 10^{-3}N)$ of uranyl nitrate, the extent of uranium precipitation in the system uranyl nitrate — sodium carbonate — strontium chloride was almost independent of changes in concentration of strontium chloride when the latter was present in excess of sodium carbonate. In the following the percentage

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of uranium precipitated and the pH values will be presented as a function of variable concentrations of sodium carbonate (1N to $1 \times 10^{-4}N$) and uranyl nitrate ($1 \times 10^{-1}N$ to $4 \times 10^{-5}N$) at constant concentration ($1 \times 10^{-1}N$) of strontium chloride by means of a contour diagram (Fig. 1) and the parallel



Fig. 1. The contour diagram of the precipitation system uranyl nitrate — sodium carbonate — strontium chloride $(1 \times 10^{-1}N)$. The 10% differences in uranium precipitated are marked by figures 0 to 10. The isoprecipitating regions with more than 85% of uranium precipitated are shaded,

projection of a three dimensional pH diagram (Fig. 2). Both diagrams were obtained from their cross sections at various constant concentrations of uranyl nitrate and variable concentrations of sodium carbonate. The corresponding cross section at $2 \times 10^{-3}N$ of uranyl nitrate was previously¹ shown. In order to facilitate orientation, the equivalent concentrations of sodium carbonate and uranyl nitrate as well as of sodium carbonate and strontium chloride are indicated by dotted lines.

In Fig. 1 the precipitation diagram is shown. The results expressed as the percentage of uranium precipitated are plotted in a plane of concentrations of uranyl nitrate (x - axis) and sodium carbonate (y - axis). The 10% differences of uranium precipitated are marked by figures 0 to 10 and

the regions of equal percentage of uranium precipitated are indicated by contours. The regions with more than $85^{0/0}$ of uranium precipitated are shaded. The boundary regions where the percentage of uranium precipitated rapidly decreased, and therefore poor reproducibility of the results was obtained, are accordingly numbered.

The regions of uranium precipitation (»concentration maximum«) and coprecipitation as well as the regions of complex solubility of uranium, observed earlier¹ at constant concentration $(2 \times 10^{-3}N)$ of uranyl nitrate, are evidently existing through a wide range of concentrations of uranyl nitrate.

The contour lines, limiting the »concentration maximum« are almost parallel to the dotted line, indicating equivalent concentrations of sodium carbonate and uranyl nitrate. This means that when a certain concentration of uranyl nitrate was exceeded at $1 \times 10^{-1}N$ of strontium chloride the occurrence of the »concentration maximum« depended on the ratio of sodium carbonate and uranyl nitrate concentrations rather than on the actual concentration of uranyl nitrate.

The coprecipitation region is limited by contour lines parallel to both equivalence lines of the precipitation diagram. Accordingly, in the concentration range examined the extent of the coprecipitation of uranium with strontium carbonate depended on the ratio of sodium carbonate and uranyl nitrate concentrations as well as on the ratio of sodium carbonate and strontium chloride concentrations.



Fig. 2. Parallel projection of the three — dimensional pH diagram of the system uranyl nitrate — sodium carbonate — strontium chloride $(1 \times 10^{-1}N)$. The diagram is cut with a plane at pH = 7. The region of concentrations where pH is about 7 is shaded.

In Fig. 2 a paralel projection of the corresponding pH diagram is shown, where the pH values (z - axis) are plotted against the concentration of sodium carbonate (x — axis) and uranyl nitrate (y — axis). In order to get an impression of the space form of the diagram the pH curves are cut by aplane with pH = 7. In the region where the ratio of sodium carbonate and uranyl nitrate concentrations was higher than about 2.5:1 and strontium chloride was present in excess of sodium carbonate (shaded region in Fig. 2), the final pH values of the systems were about 7, as a result of the buffer equilibrium existing between strontium carbonate precipitated and the ions in the mother liquor (U(VI), Sr²⁺, H⁺, OH⁻, HCO₃⁻, CO₃²⁻, etc.). On both sides of the mentioned region each pH curve has an inflection, one of them resulting from the interaction of sodium carbonate and uranyl nitrate, the other from the interaction of strontium chloride and sodium carbonate (precipitation titration). When the ratio of sodium carbonate and uranyl nitrate concentrations was lower than 0.5, dissolution of the solid phase due to the increased acidity of the systems was observed.



Fig. 3. The percentage of uranium precipitated and the pH values plotted against the ratio of the concentrations of sodium carbonate and uranyl nitrate. The concentration of strontium chloride is constant $(1 \times 10^{-1}N)$ and in excess of sodium carbonate concentration.

In Fig. 3 the percentage of uranium precipitated was plotted against the molar ratio of sodium carbonate and uranyl nitrate at constant concentration $(1 \times 10^{-1}N)$ of strontium chloride. Sodium carbonate was present in all systems in concentrations lower than $1 \times 10^{-1}N$. Curves 1—6 represent the

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function at various constant concentrations (2 imes 10⁻²N to 4 imes 10⁻⁵N) of uranyl nitrate.

Curves 1 - 4 ($2 \times 10^{-2}N$ to $5 \times 10^{-4}N$ of uranyl nitrate) have a common maximum where $90^{0}/_{0}$ to $100^{0}/_{0}$ of the uranium present was precipitated. It occurs at a molar ratio 1.5 of sodium carbonate and uranyl nitrate, at pH = 6, and thus corresponds to the previously defined »concentration maximum«. At $1 \times 10^{-4}N$ of uranyl nitrate (curve 5) this maximum is shifted to a molar ratio 2.5 of the mentioned precipitated components, and only $50^{0}/_{0}$ of uranium was precipitated. Finally at $4 \times 10^{-5}N$ of uranyl nitrate (curve 6) this maximum completely disappeared.

The minimum following the »concentration maximum« on Fig. 3 corresponds to the region of complex solubility of uranium. A second increase of the percentage of uranium precipitated is due to the coprecipitation of uranyl nitrate the limits between the coprecipitation region and the region of complex solubility are shifted to higher molar ratios of sodium carbonate and uranyl nitrate (curves 2-6, Fig. 3). The acidity of the systems was influenced by the actual concentration of uranyl nitrate only at a molar ratio of the mentioned reactants lower than 1.5. At this molar ratio and at pH about 6 all of the pH curves have an inflection reaching the value of pH about 7 at a molar ratio 2.5-3 of sodium carbonate and uranyl nitrate. When the molar ratio was further increased, the acidity of the systems remained almost constant (pH about 7); the common pH curve corresponds to the shaded region of sodium carbonate and uranyl nitrate concentrations in Fig. 2.

An attempt will be made in the following to define precisely the mutual relations of the concentrations of all of the precipitating components in the main regions of precipitation and coprecipitation of uranium. For reasons of simplicity the »concentration maximum« and the coprecipitation region will be discussed separately.

A. The »Concentration Maximum«

This maximum was shown to occur (Figs. 1, 3, previous paper¹) at constant ratio of sodium carbonate and uranyl nitrate concentrations:

$$[Na_2CO_3] = 1.5 [UO_2(NO_3)_2]$$
(1)

the final pH value of the systems being about 6 if the concentration of strontium chloride was higher than its »precipitation value« and if the concentration of uranyl nitrate was higher than approximately $1 \times 10^{-4}N$.

The »precipitation values« of strontium chloride at various constant concentrations of uranyl nitrate ($1 \times 10^{-1}N$ to $5 \times 10^{-4}N$) were determined tyndal-lometrically.

In Fig. 4 the tyndallometric values taken 10 minutes after mixing the systems are shown together with the final pH values of the systems (measured 24 hours after preparation of the samples). All the experiments were carried out at a constant relation of concentrations of sodium carbonate and uranyl nitrate, according to equation (1). The concentration of strontium chloride where the tangent on the steepest part of the tyndallometric curve cuts the x — axis, was regarded as the »precipitation value« of strontium chloride

in the same manner as the coagulation values of neutral electrolytes $^{26-28}$ were evaluated.



Fig. 4. Tyndallometric values measured 10 min after mixing, and pH values measured 24 hours after mixing plotted against the concentration of strontium chloride at various constant concentrations of uranyl nitrate. The concentration of sodium carbonate is 1.5 times the concentration of uranyl nitrate. The "precipitation values" are given by the intercepts of the tangents on the steepest part of the tyndallometric curves and the x-axis.

In Fig. 5 the »precipitation values« of strontium chloride, observed 10 min. after mixing (curve 1), as well as the concentrations of strontium chloride of the last clear systems, observed 24 hours after the preparation of the samples (curve 2) are shown as a function of the concentration of uranyl nitrate. Linear relations were obtained within the range of $1 \times 10^{-1}N$ to $2 \times 10^{-3}N$ of uranyl nitrate. At concentrations of uranyl nitrate lower than $2 \times 10^{-3}N$ the function changes significantly. The examination of systems with a concentration of uranyl nitrate below $5 \times 10^{-4}N$ was beyond the capability of the technique used, as no distinct differences in the turbidity of the systems could be observed.

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The equation of the straight line 2 can be written as follows: $[SrCl_2] = 0.7 [UO_2(NO_3)_2] - 1.36$ (2)

for equivalent concentrations of strontium chloride and uranyl nitrate.

By means of expressions (1) and (2) the conditions for the occurrence of the »concentration maximum« in the system uranyl nitrate — sodium carbonate — strontium chloride are thus given. Expression (1) relates the critical ratio of sodium carbonate and uranyl nitrate concentrations, the corresponding pH value of the systems being about 6. This equation is valid in the range of



Fig. 5. Precipitation values of strontium chloride obtained 10 min. after mixing (curve 1) and the last clear systems observed 24 hours after mixing (curve 2) are plotted as a function of the concentration of uranyl nitrate. The concentration of sodium carbonate is 1.5 times the concentration of uranyl nitrate in all systems.

 $1 \times 10^{-1}N$ to $1 \times 10^{-4}N$ of uranyl nitrate. The concentration of strontium chloride has to be equal or higher than given by equation (2) which is valid in the range of $1 \times 10^{-1}N$ to $2 \times 10^{-3}N$ of uranyl nitrate.

B. The Coprecipitation Region

The relations of the concentrations of the precipitating components at the boundaries of the region, where more than 85^{0} of uranium was coprecipitated with strontium carbonate will be defined graphically and analytically.

According to the experiments carried out at constant concentration $(2 \times 10^{-3}N)$ of uranyl nitrate and with variable concentrations of sodium carbonate and strontium chloride¹, as well as to the results obtained at constant concentration $(1 \times 10^{-1}N)$ of strontium chloride and variable concentrations of sodium carbonate and uranyl nitrate (Fig. 1) at the boundary of the coprecipitation region, the concentrations of strontium chloride and sodium carbonate are equal.

If the concentration of strontium chloride was higher than the concentration of sodium carbonate, and the ratio of the concentrations of sodium carbonate and uranyl nitrate exceeded a limiting value, more than $85^{0/0}$ of uranium was coprecipitated with strontium carbonate. For instance, at $2 \times 10^{-3}N$ of uranyl nitrate a concentration of sodium carbonate higher than

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about 20 times the concentration of uranyl nitrate, and a concentration of strontium chloride higher than the corresponding concentration of sodium carbonate was required for the coprecipitation of this amount of uranium. The corresponding ratios of the concentrations of sodium carbonate and uranyl nitrate can be obtained from Fig. 3 for various other concentrations of uranyl nitrate within the range of $2 \times 10^{-2}N$ to $4 \times 10^{-5}N$. Considering all these facts and assuming that the actual concentration of strontium chloride does not influence the critical ratio of sodium carbonate and uranyl nitrate concentrations, as was already shown at $2 \times 10^{-3}N$ of uranyl nitrate¹, the boundaries of the regions where uranium was coprecipitated, more than $85^{0}/_{0}$ were constructed and experimentally verified for various constant concentrations of uranyl nitrate.



Fig. 6. The regions where more than 85% of uranium was coprecipitated with strontium carbonate observed at various constant concentrations of uranyl nitrate are shown in a plane of concentrations of strontium chloride (x = axis) and sodium carbonate (y - axis).

In Fig. 6 the coprecipitation regions at various constant concentrations of uranyl nitrate are shown in a plane of concentrations of strontium chloride (x - axis) and sodium carbonate (y - axis). The region limited by curve 4 corresponds to $2 \times 10^{-3}N$ of uranyl nitrate (regions 9, 10 in Fig. 4 of the preceeding paper¹). At concentrations of uranyl nitrate lower than $1 \times 10^{-4}N$ poor reproducibility of the results was obtained probably due to the adsorption of uranyl ions at the walls of the centrifuge cones.

In Fig. 7 a parallel projection of the coprecipitation body is presented, the volume of which represents the region of concentrations of strontium



Fig. 7. Parallel projection of the coprecipitation body representing the region where more than 85% of uranium was coprecipitated with strontium carbonate in a coordination system of the concentrations of strontium chloride (x — axis), sodium carbonate (y — axis) and uranyl nitrate (z — axis).



Fig. 8. The critical ratio of the concentrations of sodium carbonate and uranyl nitrate, existing at the boundary of the region where more than 85% of uranium was coprecipitated, plotted against the concentration of uranyl nitrate. The concentration of strontium chloride was in all systems higher than the concentration of sodium carbonate. chloride (x — axis), sodium carbonate (y — axis) and uranyl nitrate (z — axis), where more than $85^{0/6}$ of uranium is coprecipitated with strontium carbonate. The boundaries of the region are represented by surfaces as functions of the changes in concentration of the precipitating components. In order to obtain an analytical expression defining the relationship between the concentrations of sodium carbonate and uranyl nitrate at the boundary of the coprecipitation region, in Fig. 8 the critical ratio of their concentrations was plotted against the concentration of uranyl nitrate. The corresponding values for the region with more than $85^{0/6}$ of uranium coprecipitated were obtained form Fig. 6.

A straight line was obtained in the range of $1 \times 10^{-1}N$ to $2 \times 10^{-4}N$ of uranyl nitrate. The equation of the straight line can be written in general form as follows:

$$\log [Na_2CO_3] / [UO_2(NO_3)_2] = b \log [UO_2(NO_3)_2] + a$$
(3)

where a and b are empirical constants, a being the intercept at the y — axis, and b being the slope of the straight line. Rearranging equation (3), equation (4) was obtained:

$$\log [Na_2CO_3] = (b + 1) \log [UO_2(NO_3)_2] + a$$
(4)

Inserting the constants a and b, as obtained form Fig. 8, the first condition for the coprecipitation of more than $85^{0/0}$ of uranium with strontium carbonate can be written as follows:

$$\log [Na_2CO_3] \ge 0.76 \log [UO_2(NO_3)_2] + 0.7$$
 (5)

Expression (5) is valid in the range of $1 \times 10^{-1}N$ to $2 \times 10^{-4}N$ of uranyl nitrate for equivalent concentrations of the precipitating components.

The second condition is given by expression (6):

$$[SrCl_2] \ge [Na_2CO_3] \tag{6}$$

DISCUSSION

Considering the results obtained at constant concentration $(2 \times 10^{-3}N)$ of uranyl nitrate¹ and the results outlined in this paper, it seems probable that in the system uranyl nitrate — sodium carbonate — strontium chloride the following processes prevail:

a) Precipitation of uranium in a narrow range of the ratio of sodium carbonate and uranyl nitrate concentrations (Fig. 1, 3) at a nearly constant pH value (pH about 6) of the systems in equilibrium (Fig. 2, 3). The exact conditions for the occurrence of this precipitation maximum (»concentration maximum«) are given by expressions (1) and (2).

b) Coprecipitation of uranium with strontium carbonate was found to prevail in a wide range of concentrations of the precipitating components. The boundaries given as the ratios of the concentrations of the precipitating components (Fig. 8 and equality of expressions (5) and (6)) are dividing the region, where more than $85^{0/0}$ of uranium was coprecipitated from the region where the percentage of uranium coprecipitated was lowered by the formation of soluble complex uranyl di- and tri-carbonates.

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c) The formation of soluble complex uranyl di- and tri-carbonates was found to prevail in the region where sodium carbonate was present in excess of strontium chloride. In the region where strontium chloride was in excess this process was limited to a narrow range of concentrations of the precipitating components, dividing the regions of precipitation and coprecipitation of uranium (Figs. 1, 3).

The ratios of the concentrations of strontium chloride and uranyl nitrate at the boundary of the »concentration maximum« (equation (2), Fig. 5) indicate complicated precipitation processes. It was shown (Fig. 5) that the ratio between the »precipitation value« of strontium chloride and the concentration of uranyl nitrate changed with changes in concentration of uranyl nitrate. In the range of $1 \times 10^{-1}N$ to $2 \times 10^{-3}N$ of uranyl nitrate the concentration of strontium chloride necessary for precipitation increased with increasing concentration of uranyl nitrate while at lower concentrations of uranyl nitrate the trend was opposite. This change in the function may indicate either changes in the composition of the solid phase or, more probably, changes in the composition of the mother liquor in contact. In the latter case at concentrations of uranyl nitrate lower than $2 \times 10^{-3}N$ ionic solubility has to be considered while at higher concentrations of uranyl nitrate complex solubility, probably the formation of polynuclear complexes of uranium(VI) could be assumed.

At the boundary of the coprecipitation region the critical ratio of sodium carbonate and uranyl nitrate concentrations was shown to increase with decreasing concentration of uranyl nitrate (Fig. 8). This trend indicates a corresponding lowering of the capacity of strontium carbonate for the coprecipitation of uranium (expressed as the amount of uranium coprecipitated with a unit amount of strontium carbonate).*

Expressions (1), (2), (5), and (6) may be of some practical importance defining the conditions for maximum precipitation and coprecipitation of uranium in the system investigated. It may be emphasized that they are only arbitrarily chosen exampels to show how the complicated processes in a three-component precipitation system can be simply represented. Similar expressions can be derived for any region of precipitation and dissolution of uranium in the systems uranyl nitrate — sodium carbonate — alkaline earth chlorides. With the aid of such expressions the concentrations of the precipitating components necessary for the precipitation of any given percentage of uranium can be calculated. Such an approach offers many practical advantages.

Further investigations are in progress to clarify the mechanism of precipitation and coprecipitation of uranium(VI) in the systems investigated.

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^{*} The amount of strontium carbonate precipitated is given by the concentration of sodium carbonate added, since in the whole coprecipitation region the concentration of sodium carbonate is lower than the concentration of strontium chloride (condition 2 for the coprecipitation of more than $85^{0}/_{0}$ of uranium), and the precipitation of strontium carbonate can be regarded as quantitative in this range of concentrations.

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IZVOD

Taloženje i hidroliza urana (VI) u vodenim otopinama. III. Uranil nitrat — natrijev karbonat — stroncijev klorid

H. Füredi

Ispitan je utjecaj promjene koncentracije uranil nitrata na taloženje urana u sistemu uranil nitrat — natrijev karbonat — stroncijev klorid. Nađeno je, da »koncentracijski maksimum« nastaje kad odnosi ekvivalentnih koncentracija taložnih komponenata odgovaraju slijedećim izrazima:

$$\begin{bmatrix} Na_2CO_3 \end{bmatrix} = 1.5 \quad \begin{bmatrix} UO_2(NO_3)_2 \end{bmatrix}$$
(1)

$$\begin{bmatrix} SrCl_2 \end{bmatrix} > 0.7 \quad \begin{bmatrix} UO_2(NO_3)_2 \end{bmatrix} - 1.36$$
(2)

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Ravnotežne *p*H vrijednosti odgovarajućih sistema iznosile su oko 6. Jednadžba (1) vrijedi u koncentracijskom području od $1 \times 10^{-1}N$ do $1 \times 10^{-4}N$ uranil nitrata, dok izraz (2) vrijedi u području od $1 \times 10^{-1}N$ do $2 \times 10^{-3}N$ uranil nitrata.

Slijedećim izrazima definirani su odnosi ekvivalentnih koncentracija taložnih komponenata u području, gdje više od 85% urana koprecipitira sa stroncijevim karbonatom:

Izraz (4) vrijedi u području od $6 \times 10^{-2}N$ do $2 \times 10^{-4}N$ uranil nitrata. Funkcionalna ovisnost koprecipitacije urana o koncentraciji taložnih komponenata, koja je definirana jednadžbama (3) i (4) prikazana je i grafički pomoću taložnog tijela u koordinatnom sustavu stroncijeva klorida (os x), natrijeva karbonata (os y) i uranil nitrata (os z).

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