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Recent achievements of actinide radiation chemistry

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Abstract

Data are given on the mathematical modelling of the radiation-chemical behaviour of U, Np, Pu, Am in $HClO_4$ and HNO_3 solutions. Based on the comparison between the calculated and experimental data the optimized reaction schemes were developed for each system; the previously unknown rate constants were found for a lot of radiation-chemical reactions; mechanisms and kinetic regularities were established in the radiolytic behaviour of individual valent forms of a given actinide in a solution under study. © 1998 Elsevier Science S.A.

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1. Introduction

By the late 1980s a lot of experimental data had been accumulated on the radiation-chemical behaviour of actinides in various aqueous solutions, basically in perchloric and nitric acid solutions. To some extent those data have been generalized in a book by Vladimirova [1]. This material demonstrates the most complicated, sometimes hard to explain, relationships for the valent transitions of actinide ions. Now the time has come to establish the detailed mechanisms and kinetic regularities in the radiation-chemical behaviour of actinides and to create an instrument for predicting their behaviour under specified conditions. To cope with this problem the author made use of the mathematical modelling method. To date radiolysis models have been designed for the systems: HNO_3 [2], $HClO_4$ [3,4], U+HNO_3 [5], U+$HClO_4$ [6], Np+HNO_3 [7–11], Np+$HClO_4$ [12,13], Pu+HNO_3 [14–22], Am+$HClO_4$ [23].

2. Uranium

It is experimentally shown that under irradiation U(IV) is oxidized to U(VI). With an increase of [$HClO_4$] and [HNO_3] from 1 to 12 mol/l at [U(IV)]=0.006 mol/l under α-radiolysis in $HClO_4$ *G*[-U(IV)] is increased from 2 to 5.5, in HNO_3 it is decreased from 1.4 to 0.1 ion/100 eV [24]. The radiation-chemical oxidation of U(IV) in 0.1–12 mol/l $HClO_4$ and HNO_3 solutions under α- and γ-radiolysis was modelled.

The U+$HClO_4$ system schema comprises the following reactions:U(VI)+HO_2→U(V), U(V)+OH→U(VI), U(IV)+OH→U(V), U(IV)+HO_2→U(V), U(IV)+H_2O_2→U(V), U(IV)+ClO_3^-→U(V), U(IV)+ClO_2→U(V), U(IV)+ClO_2^-→U(V), U(IV)+ClO^-→U(V), U(IV)+O_2→U(V), U(V)+U(V)→U(IV)+U(VI). As to the U+HNO_3 system, aside from the above mentioned reactions with water radiolysis products, the U(IV)+NO_3→U(V), U(IV)+NO_2→U(V) reactions are included. To make the text shorter here and below the stoichiometry of the reactions is not given fully. For the calculations, the rate constants of the reactions of U(IV) and U(V) with OH were assumed similar and equal to 1×10^9 l/(mole s). The rate constants of the U(IV)+H_2O_2, U(IV)+ClO_3^-, U(IV)+O_2, U(V)+U(V) reactions at [$HClO_4$]<1 mol/l were taken from the literature [25]; at higher concentrations of the acid they were found by extrapolation. The calculated results were compared to the experimental data which allowed the optimization of the reaction schemes and the unknown rate constants to be found (Table 1).

Using the derived rate constants the rates of individual reactions were calculated and the following equations were derived for the initial rates of the U(IV) oxidation in $HClO_4$ and HNO_3:

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Table 1
Rate constants found for U reactions, l/(mole s)

Reaction	Acid concentrations, mol/l					
	1	2	4	6	8	12
	$HClO_4$					
$U(VI)+HO_2$	1×10^3	1×10^2	1	0.01	—	—
$U(IV)+HO_2$	10	1×10^2	5×10^2	1.5×10^3	5×10^3	1×10^5
$U(IV)+ClO_2$	5	4	2	0.04	0.04	0.04
$U(IV)+ClO_2^-$	40	35	16	0.4	0.4	0.4
$U(IV)+ClO^-$	3.5	2.5	1.5	0.04	0.04	0.04
	HNO_3					
$U(IV)+NO_3$	1×10^5	2×10^4	7×10^3	3×10^3	2×10^3	1×10^3
$U(IV)+NO_2$	1×10^2	80	50	35	15	2.0

$$\begin{aligned}-\mathrm{d}[U(IV)]/\mathrm{d}t &= \mathrm{d}[U(VI)]/\mathrm{d}t\\ &= [U(IV)](k[HO_2]+k[OH]+k[H_2O_2]\\ &\quad +k[ClO_3^-]+k[ClO_2]+k[ClO_2^-]\\ &\quad +k[ClO^-]+k[O_2])-k[U(V)]^2\\ &= k[U(V)]^2 \qquad (1)\end{aligned}$$

$$\begin{aligned}-\mathrm{d}[U(IV)]/\mathrm{d}t &= \mathrm{d}[U(VI)]/\mathrm{d}t\\ &= [U(IV)](k[OH]+k[H_2O_2]+k[NO_3]\\ &\quad +k[NO_2])-k[U(V)]^2=k[U(V)]^2 \qquad (2)\end{aligned}$$

In expressions (1), (2) and below, k is the rate constant of a particular reaction.

The calculations show that an increase of G[-U(IV)] with [$HClO_4$] is caused by the growth of both the rate constant of the $U(IV)+HO_2$ reaction and chlorine-containing radiolysis products. A decrease of G[-U(IV)] with an increase of [HNO_3] is due to lower rate constants of the $U(IV)+NO_3$ and $U(IV)+NO_2$ reactions.

3. Neptunium

It is shown experimentally that under irradiation at any concentration of HNO_3 Np(VI) is reduced and Np(IV) is oxidized to Np(V). The latter in its turn is also capable of being reduced or oxidized. As a result, HNO_3 solutions always contain three valent forms of Np in equilibrium [1].

The radiation-chemical behaviour of neptunium under α- and γ-radiolysis in 1–6 mol/l HNO_3 was modelled.

The $Np+HNO_3$ system schema comprises the following reactions: $Np(VI)+H\rightarrow Np(V)$, $Np(VI)+H_2O_2\rightarrow Np(V)$, $Np(VI)+HNO_2\rightarrow Np(V)$, $Np(V)+NO_2\rightarrow Np(VI)$, $Np(V)+OH\rightarrow Np(VI)$, $Np(V)+NO_3\rightarrow Np(VI)$, $Np(V)+H\rightarrow Np(IV)$, $Np(IV)+OH\rightarrow Np(V)$, $Np(IV)+NO_3\rightarrow Np(V)$, $Np(IV)+H_2O_2\rightarrow Np(V)$, $Np(IV)+Np(VI)\rightarrow Np(V)+Np(V)$.

The calculation made use of the rate constants of the $Np(VI)+H$ and $Np(V)+OH$ reactions that according to [26] are equal to 4×10^9 and 6×10^8 l/(mole s). From the comparison between the calculated and numerous experimental data an optimized model was designed and the constants of some reaction rates of neptunium in HNO_3 were found (Table 2).

Based on the calculated results the following equations were derived for the initial rates of Np(VI) reduction and Np(IV) oxidation in HNO_3:

$$\begin{aligned}-\mathrm{d}[Np(VI)]/\mathrm{d}t &= \mathrm{d}[Np(V)]/\mathrm{d}t\\ &= [Np(VI)](k[H]+k[HNO_2]+k[H_2O_2])\\ &\quad -[Np(V)](k[NO_2]+k[OH]+k[NO_3])\end{aligned} \qquad (3)$$

$$\begin{aligned}-\mathrm{d}[Np(IV)]/\mathrm{d}t &= \mathrm{d}[Np(V)]/\mathrm{d}t\\ &= [Np(IV)](k[OH]+k[NO_3]\\ &\quad +k[Np(VI)]) \qquad (4)\end{aligned}$$

It is shown experimentally that under α-irradiation in $HClO_4$ at a concentration ≤ 2 mol/l only Np(V) is in equilibrium while at [$HClO_4$]≥ 4 mol/l only Np(VI) is in equilibrium [27]. The stabilization of Np(VI) upon radiolysis is an unconventional result.

The radiation-chemical behaviour of neptunium under α-radiolysis in 1–6 mol/l $HClO_4$ was modelled. As to the $Np+HClO_4$ system, aside from the above mentioned reactions with water radiolysis products, the schema comprises the following reactions: $Np(VI)+HO_2\rightarrow Np(V)$, $Np(V)+HO_2\rightarrow Np(IV)$, $Np(IV)+HO_2\rightarrow Np(V)$, $Np(V)+ClO_3^-\rightarrow Np(VI)+ClO_2$, $Np(V)+ClO_2\rightarrow Np(VI)$, $Np(IV)+ClO_3^-\rightarrow Np(V)$, $Np(IV)+ClO_2\rightarrow Np(V)$.

From the comparison between the calculated and experimental data the unknown rate constants for $HClO_4$ were found (Table 2). The calculations showed the rate constants of the Np(VI) and Np(IV) reactions with HO_2 to be independent of the acidity and equal to 1×10^3 and 1×10^2 l/(mole s), respectively.

The main objective of modelling was to reveal why Np(VI) is stabilized in concentrated perchloric acid solutions. This proved feasible by including the reaction between Np(V) and chlorate ion, ClO_3^- into the schema. The calculations showed that in the course of this reaction

Table 2
Rate constants found for Np reactions, l/(mole s)

Reaction	Acid concentrations, mol/l				
	1	2	3	4	6
	HNO_3				
Np(V) + H	5×10^{6}	1×10^{7}	2.5×10^{7}	5×10^{7}	2×10^{8}
Np(VI) + HNO_2	2.7	1.3	0.9	0.8	0.7
Np(V) + NO_2	90	120	175	250	700
Np(VI) + H_2O_2	10	6.0	4.5	4.0	3.8
Np(V) + NO_3	1×10^{5}	1×10^{5}	1×10^{5}	1×10^{5}	1×10^{5}
Np(IV) + OH	3×10^{8}	1.5×10^{8}	1×10^{8}	5×10^{7}	2×10^{7}
Np(IV) + NO_3	1.5×10^{6}	6×10^{5}	3×10^{5}	2×10^{5}	1×10^{5}
Np(IV) + H_2O_2	0.1	0.07	0.05	0.03	0.02
Np(IV) + Np(VI)	0.1	0.02	4×10^{-3}	1×10^{-3}	2×10^{-4}
	$HClO_4$				
Np(V) + HO_2	1.5	2	5	10	5×10^{2}
Np(V) + ClO_3^-	1×10^{-4}	2×10^{-4}	5×10^{-4}	1.5×10^{-3}	0.5
Np(V) + ClO_2	7×10^{-4}	1.5×10^{-3}	5×10^{-3}	0.01	0.5
Np(IV) + ClO_3^-	1.5×10^{-4}	2×10^{-4}	5×10^{-4}	1.5×10^{-3}	0.1
Np(IV) + ClO_2	5	4	3	2	0.1

there takes place an increase in the concentration of ClO_2. This leads to higher rates of some reactions, specifically, the $H_2O_2 + ClO$ one, a lower concentration of H_2O_2 that is the main reductant of Np(VI) and to the stabilization of the latter.

The following equations were derived for the rates of the Np(VI) reduction and the Np(IV) oxidation in $HClO_4$:

$$\begin{aligned} -\mathrm{d}[\mathrm{Np(VI)}]/\mathrm{d}t &= \mathrm{d}[\mathrm{Np(V)}]/\mathrm{d}t \\ &= [\mathrm{Np(VI)}](k[\mathrm{H}] + k[\mathrm{H_2O_2}] + k[\mathrm{HO_2}]) \\ &\quad - [\mathrm{Np(V)}](k[\mathrm{OH}] + k[\mathrm{ClO_3^-}] + k[\mathrm{ClO_2}]) \end{aligned} \tag{5}$$

$$\begin{aligned} -\mathrm{d}[\mathrm{Np(IV)}]/\mathrm{d}t &= \mathrm{d}[\mathrm{Np(V)}]/\mathrm{d}t \\ &= [\mathrm{Np(IV)}](k[\mathrm{OH}] + k[\mathrm{H_2O_2}] + k[\mathrm{HO_2}] \\ &\quad + k[\mathrm{ClO_3^-}] + k[\mathrm{ClO_2}]) \end{aligned} \tag{6}$$

The calculations show that at $[HClO_4] \leq 2$ mol/l the first term in expression (5) is larger than the second one, at $[HClO_4] > 4$ mol/l the opposite is true.

4. Plutonium

The radiation-chemical behaviour of Pu(VI), Pu(IV) was mainly studied in HNO_3 solutions. Pu(VI) is shown to be reduced to Pu(IV), the latter is oxidized to Pu(VI). At a specific dose rate above some threshold for the given experimental conditions no Pu(VI) reduction takes place. At [Pu(IV)] above some limit under the given experimental conditions, no oxidation takes place [1].

The radiation-chemical behaviour of plutonium under α- and γ-radiolysis in 1–6 mol/l HNO_3 was modelled.

The Pu + HNO_3 system comprises the following reactions: Pu(VI) + H → Pu(V), Pu(VI) + H_2O_2 → Pu(V), Pu(VI) + HNO_2 → Pu(V), Pu(V) + NO_2 → Pu(VI), Pu(V) + OH → Pu(VI), Pu(V) + NO_3 → Pu(VI), Pu(IV) + OH → Pu(V), Pu(IV) + NO_3 → Pu(V), Pu(IV) + H_2O_2 → Pu(III), Pu(IV) + HNO_2 → Pu(III), Pu(III) + NO_2 → Pu(IV), Pu(III) + OH → Pu(IV), Pu(III) + NO_3 → Pu(IV), Pu(V) + Pu(V) → Pu(IV) + Pu(VI), Pu(IV) + Pu(IV) → Pu(III) + Pu(V), Pu(III) + Pu(V) → Pu(IV) + Pu(IV), Pu(III) + Pu(VI) → Pu(IV) + Pu(V), Pu(IV) + Pu(V) → Pu(III) + Pu(VI).

For the calculations at all concentrations of HNO_3 the rate constants of the Pu(VI) + H, Pu(V) + OH, Pu(V) + NO_3 reactions were assumed to be identical and equal to 1×10^{8} l/(mole s), those of Pu(VI) reactions with H_2O_2 and HNO_2 were taken equal to 0.1 and 0.03 l/(mole s) [1] and the ones of Pu(III) reactions with OH and NO_3 were taken equal to 5×10^{8} and 2×10^{8} l/(mole s) [26], respectively.

From the comparison between the calculated and numerous experimental data an optimized model was designed and the constants of some reaction rates of plutonium in HNO_3 were found (Table 3).

The calculations show the rate constants of the Pu(V) +

Table 3
Rate constants found for Pu reactions, l/(mole s)

Reaction	$[HNO_3]$, mol/l			
	1	2	3	6
Pu(V) + NO_2	9×10^{3}	1.5×10^{4}	2×10^{4}	2.5×10^{4}
Pu(V) + Pu(V)	2×10^{-3}	3×10^{-3}	5×10^{-3}	2×10^{-2}
Pu(IV) + Pu(IV)	1×10^{-5}	2×10^{-6}	5×10^{-7}	5×10^{-8}
Pu(V) + Pu(III)	0.5	1.0	1.6	3.0
Pu(IV) + OH	6×10^{6}	4×10^{6}	2×10^{6}	3×10^{5}
Pu(IV) + NO_3	7.5×10^{5}	5×10^{4}	3×10^{4}	1×10^{4}
Pu(IV) + HNO_2	0.025	0.015	0.007	0.0025
Pu(III) + NO_2	1.9×10^{3}	2×10^{3}	2.5×10^{3}	3×10^{3}

Pu(IV) and Pu(III) + Pu(VI) reactions to affect little the kinetics of the Pu valence changes. In 2, 3 and 6 mol/l HNO_3 the minimal values of those constants are 0.5, 2 and 3; 0.4, 4 and 15 l/(mole s). It is found that the adequate agreement between the calculated and experimental data is reached at any $[HNO_3]$ at $k/Pu(IV)+H_2O_2/=2\times10^3$ l/(mole s).

The calculations resulted in the following equations of the rates of individual valence changes:

$$\begin{aligned}-\,d[Pu(VI)]/dt &= d[Pu(V)]/dt\\ &= [Pu(VI)](k[H]+k[HNO_2]+k[H_2O_2])\\ &\quad -[Pu(V)](k[NO_2]+k[OH]+k[NO_3])\end{aligned} \tag{7}$$

$$\begin{aligned}-\,d[Pu(VI)]/dt &= d[Pu(IV)]/dt\\ &= k[Pu(V)]^2+k[Pu(V)][Pu(III)]\\ &\quad -[Pu(IV)](k[Pu(IV)]+k[OH]+k[NO_3])\end{aligned} \tag{8}$$

The equation for the rate of the Pu(IV)→Pu(VI) oxidation takes the form of Eq. (8) with the opposite signs of the terms.

The significance of the individual terms in the equations depends on $[HNO_3]$, [Pu], the radiation dose rate (D). It is shown that in Eq. (7) at $[HNO_3]\geq3$ mol/l the main part is played by the rates of reactions $Pu(VI)+HNO_2$ and $Pu(V)+NO_3$. Since as is shown by the calculations $[HNO_2]$ is $fD^{1/4}$, $[NO_3]$ is fD with a growth of the dose rate, the rate of reaction between Pu(VI) and HNO_2 does not almost increase but the rate of reaction between Pu(V) and NO_3 becomes higher which results in the stabilization of Pu(VI) at high dose rates. The negative dependence of the Pu(IV) oxidation rate upon the term $k[Pu(V)][Pu(III)]$ leads to the fact that at a specific concentration of Pu(IV) the latter is not oxidized.

5. Americium

The radiation chemical behaviour of Am is studied in detail in $HClO_4$ solutions. At $[HClO_4]\leq2$ mol/l Am(VI) is shown to be self-reduced to Am(V); Am(III) is formed when [Am(VI)] becomes low. At $[HClO_4]=6$ mol/l Am(V) and Am(III) are formed simultaneously. In 9 mol/l $HClO_4$ Am(VI) is reduced to Am(III). In 12 mol/l $HClO_4$ no reduction of Am(VI) takes place [28]. The radiation-chemical behaviour of americium under α-radiolysis in 1–12 mol/l $HClO_4$ was modelled.

The $Am+HClO_4$ system comprises the following reactions: $Am(VI)+HO_2\rightarrow Am(V)$, $Am(VI)+H_2O_2\rightarrow Am(V)$, $2Am(VI)+H_2O\rightarrow 2Am(V)$, $Am(VI)+Cl^-\rightarrow Am(V)$, $Am(VI)+ClO_2\rightarrow Am(V)$, $Am(V)+OH\rightarrow Am(VI)$, $Am(V)+HO_2\rightarrow Am(IV)$, $Am(V)+ClO_2\rightarrow Am(IV)$, $Am(IV)+HO_2\rightarrow Am(III)$, $Am(IV)+H_2O_2\rightarrow Am(III)$, $Am(III)+OH\rightarrow Am(IV)$, $Am(III)+ClO_3^-\rightarrow Am(IV)$, $Am(V)+Am(V)\rightarrow Am(IV)+Am(VI)$, $Am(IV)+Am(IV)\rightarrow Am(III)+Am(V)$, $Am(IV)+Am(V)\rightarrow Am(III)+Am(VI)$, $Am(III)+Am(VI)\rightarrow Am(IV)+Am(V)$.

The rate constants of the $Am(V)+Am(V)$, $Am(IV)+Am(IV)$, $Am(IV)+HO_2$, $Am(IV)+H_2O_2$, $Am(III)+OH$ reactions were taken from the literature [26,29,30].

From the comparison between the calculated and numerous experimental data an optimized model was designed and the constants of some reaction rates of americium in $HClO_4$ were found (Table 4).

The calculation showed the rate constants of the $Am(VI)+HO_2$, $Am(VI)+Cl^-$, $Am(V)+OH$, $Am(V)+ClO_2$ reactions to be acidity independent and equal to 1×10^6, 10, 1×10^7 and 1.0 l/(mole s).

The calculations resulted in the following equations for the rates of Am(VI) and Am(V) reduction:

$$\begin{aligned}-\,d[Am(VI)]/dt &= d[Am(V)]/dt\\ &= [Am(VI)](k[Am(III)+k[HO_2]\\ &\quad +k[H_2O_2]+k[Am(VI)][H_2O]+k[Cl^-]\\ &\quad +k[ClO_2])-[Am(V)](k[Am(V)]\\ &\quad +k[Am(IV)+k[OH]+k[ClO_3^-])\end{aligned} \tag{9}$$

$$\begin{aligned}-\,d[Am(V)]/dt &= d[Am(III)]/dt\\ &= [Am(V)]([HO_2]+k[ClO_2]\\ &\quad +k[Am(V)])-k[Am(IV)]^2\end{aligned} \tag{10}$$

Table 4
Rate constants found for Am reactions, l/(mole s)

Reaction	$[HClO_4]$, mol/l					
	0.1	1	2	4	6	9
$Am(V)+HO_2$	5×10^2	1×10^4	1×10^5	1×10^5	1×10^5	1×10^5
$Am(III)+ClO_3^-$	$\leq10^{-4}$	1×10^{-4}	5×10^{-4}	0.02	0.05	20
$Am(VI)+H_2O_2$	20	10	5	1	0.5	0.15
$2Am(VI)+H_2O$	1.5×10^{-4}	1×10^{-4}	5×10^{-5}	1×10^{-5}	$\leq1\times10^{-5}$	$\leq1\times10^{-5}$
$Am(VI)+ClO_2$	100	70	50	10	2	1×10^{-4}
$Am(V)+ClO_3^-$	$\leq1\times10^{-4}$	1×10^{-4}	5×10^{-4}	0.01	0.05	20

The calculations show that a decrease in the Am(VI) reduction rate with an increased [$HClO_4$] is caused by the rate acceleration of the Am(V)-ClO_3^- reaction.

6. Conclusion

Almost all the experimental data on the radiation-chemical behaviour of U, Np, Pu, Am in HNO_3 and $HClO_4$ solutions are described by the developed models and rate constants found.

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