

The Inorganic Chemist's Guide to Actinide Radiation Chemistry: A Review

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Table of Contents

Aqueous Chemistry.....	3
Solid-State Chemistry	19
References	23

Aqueous Chemistry

Table S1. Table summarizing γ -irradiated actinides Th-Cf and their redox chemistry with the hydrated electron (e_{aq^-}).

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions	pH	Ref.
			Conc. (M)		
Th(IV)	Th(III)	-	-	acidic	1
		HClO ₄	0.01	acidic	2
Th(IV)SO ₄ ²⁺	Th(III)	-	-	acidic	1
U(IV)	U(III)	HClO ₄	0.05-1.0	acidic	3, 4
		H ₂ SO ₄	0.68	acidic	2
U(VI)	U(IV)O ₂ nanoparticles	uranyl nitrate/10% isopropanol	0.01	1.2-4.0	5
	U(V)	-	-	5.3-6.8	6, 7
		artificial sea water	-	basic	8
		HClO ₄	1.0	acidic	1
U(VI)carbonate complex	U(V)	CO ₃ ²⁻	0.01	basic	8
		CO ₃ ²⁻	0.05	basic	9
U(VI)IMDA complex	U(V)	IMDA	0.01-0.25	acidic	8
U(VI)NTA complex	U(V)	NTA	0.01-0.25	acidic	8
U(VI)oxalate complex	U(V)	C ₂ O ₄ ²⁻	0.1	acidic	8
Np(IV)	Np(III)	HClO ₄	0.023-0.91	acidic	2
		H ₂ SO ₄	0.042-0.68	acidic	2
Np(IV) carbonate complex	Np(III)	CO ₃ ²⁻	0.1-4.3	basic	10
		CO ₃ ²⁻	6.0	basic	2
Np(V)	Np(IV)(OH) ₄	-	3.6-5.8	acidic	11
		-	5.3-6.1	acidic	6
Np(V) carbonate complex	Np(IV)	CO ₃ ²⁻	0.1-5.0	basic	2, 12
Np(V)	Np(IV)	OH ⁻	1.0-7.5	basic	13
Np(VI)	Np(V)	-	5.3-5.8	-	6
		-	2.5	-	14
Np(VI)	Np(V)	OH ⁻	0.5-12.5	basic	13, 15
	Np(VII)	sat. N ₂ O/NaOH	1.0	basic	16, 17
Np(VI)O ₂ (OH) ⁺	Np(V)O ₂ OH	-	-	-	14
Np(VI) carbonate complex	Np(V)	HCO ₃ ⁻ /CO ₃ ²⁻	0.05	basic	9
		HCO ₃ ⁻ /CO ₃ ²⁻	0.1-5.0	basic	10
		HCO ₃ ⁻ /CO ₃ ²⁻	0.5 : 0.5	basic	10, 15
Np(VII)	Np(VI)	OH ⁻	0.1-12.5	basic	13
Pu(IV)	Pu(III)	H ₂ SO ₄	0.018-0.078	1	2
Pu(IV) carbonate complex	Pu(III)	CO ₃ ²⁻	6.0	basic	18

	Pu(V)	CO_3^{2-}	-	basic	19
Pu(VI)	Pu(V)	-	-	5.6-6.1	6
		OH^-	2.0-7.5	basic	20
		CO_3^{2-}	0.05	basic	9
Pu(VI) carbonate complex	Pu(V)	CO_3^{2-}	0.1-1.0	basic	10
Pu(VII)	Pu(VI)	OH^-	1.0-7.5	14	20
Am(III)	Am(II)	LiClO_4	0.11	-	6, 21, 22
Am(IV)	Am(III)	HNO_3	1.0-9.0	acidic	23
Am(V)	Am(IV)	LiClO_4	0.11	-	21
Am(VI)	Am(V)	LiClO_4	0.11	-	21
Cm(III)	Cm(II)	$\text{N}_2\text{O}/\text{HClO}_4/(\text{CH}_3)_3\text{O}$ H	0.18-0.72 : 0.4-0.5	acidic	24
Bk(III)	Bk(II)	$(\text{CH}_3)_3\text{OH}$	0.0025	5	25
Cf(III)	Cf(II)	$\text{HClO}_4/(\text{CH}_3)_3\text{OH}$	dilute : 0.1	5.1-5.3	26
	Cf(II)	$\text{HClO}_4/(\text{CH}_3)_3\text{OH}$	0.001 : 0.5	acidic	27

Table S2. Table summarizing γ -irradiated actinides U-Cm and Cf and their redox chemistry with the H \cdot radical.

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions		Ref.
			Conc. (M)	pH	
U(IV)	U(III)	H ₂ SO ₄	0.5	acidic	2
U(V)	U(VI)	HClO ₄	0.1	acidic	2, 28
		H ₂ SO ₄	0.4	acidic	2, 28
U(VI)	U(V)	HClO ₄	1.0	acidic	29
		H ₂ SO ₄	0.5	acidic	2
	U(VI)/U(IV)	H ₂ SO ₄	0.4	acidic	30
Np(III)	Np(IV)	HClO ₄	1.0	acidic	1, 28
		H ₂ SO ₄	1.0	acidic	1, 28
Np(IV)	Np(III)	HClO ₄	0.9	acidic	1
Np(V)	Np(IV)	HClO ₄	1.0	acidic	31
Np(VI)	Np(V)	HClO ₄	1.0	acidic	1
		HClO ₄	1.0	acidic	14
Pu(III)	Pu(IV)	HClO ₄	3.0	acidic	1, 28
		HClO ₄ /H ₂ SO ₄	0.2 : 1.8	acidic	1, 28
Pu(IV)	Pu(III)	HClO ₄	3.0	acidic	1
		H ₂ SO ₄	0.6-1.6	acidic	1
Pu(VI)	Pu(V)	HClO ₄	1.0	acidic	1
Am(V)	Am(IV)	HClO ₄	1.0	acidic	1
	Am(IV)/Am(VI)	HNO ₃	1.0-9.0	acidic	23
Am(VI)	Am(V)	HClO ₄	-	acidic	1
Cm(III)	Cm(II)	sat. N ₂ O/HClO ₄ /(CH ₃) ₃ OH	0.07-1.0 : 0.010-0.020	acidic	32
Cf(III)	Cf(II)	HClO ₄	1.0	acidic	27

Table S3. Table summarizing γ -irradiated actinides U-Cm and Cf and their redox chemistry with $\cdot\text{OH}$.

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions		Ref.
			Conc. (M)	pH	
U(IV)	U(III)	H ₂ SO ₄	0.5	acidic	2
U(V)	U(VI)	HClO ₄	0.1	acidic	2, 28
		H ₂ SO ₄	0.4	acidic	2, 28
U(VI)	U(V)	HClO ₄	1.0	acidic	29
		H ₂ SO ₄	0.5	acidic	2
	U(VI)/U(IV)	H ₂ SO ₄	0.4	acidic	30
Np(III)	Np(IV)	-	-	acidic	33
Np(IV)	Np(V)	HClO ₄	1	acidic	34
Np(V)	Np(VI)	HClO ₄	0.1-2.95	acidic	34
		HClO ₄	1.0	acidic	35
		-	< 4.0	acidic	14
		-	3.64-4.75	acidic	35
		HNO ₃	0.09-4.0	acidic	36
Pu(III)	Pu(IV)	HClO ₄	3.0	acidic	1, 28
		HClO ₄ /H ₂ SO ₄	0.2 : 1.8	acidic	1, 28
Pu(IV)	Pu(III)	HClO ₄	3.0	acidic	1
		H ₂ SO ₄	0.6-1.6	acidic	1
Pu(VI)	Pu(V)	HClO ₄	1.0	acidic	1
Am(III)	Am(IV)	-	1.0-4.0	acidic	37
		HClO ₄	1.0	acidic	37
		LiClO ₄	0.11	acidic	18
Am(V)	Am(VI)	HNO ₃	1.0-9.0	acidic	23
Cm(III)	Cm(IV)	sat. N ₂ O/HClO ₄ /KSCN	0.010 : 73-100 x 10 ⁻⁶	acidic	32
Cf(III)	Cf(III)	sat. N ₂ O	-	5.1	26
Cf(IV)/Cf(IV)(SCN) ²⁺ complex		sat. N ₂ O/HClO ₄ /KSCN	0.01 : 0.1	acidic	27

Table S4. Table summarizing γ -irradiated actinides Np and its redox chemistry with O^\bullet radical species.

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions		Ref.
			Conc. (M)	pH	
Np(IV)	Np(V)	LiOH	1.03-2.0	basic	38
		NaOH	0.5-1.7	basic	38
Np(V)	Np(VI)	LiOH	1.0	basic	38
Np(VI)	Np(VI) peroxy/hydroxy- complexes	LiOH	0.033-0.24	basic	38

Table S5. Table summarizing γ -irradiated actinides Th-Np and Am and their redox chemistry with HO_2^\cdot .

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions	Ref.	
			Conc. (M)	pH	
Th(IV)	Th(IV) peroxy-complexes	H_2O_2	0.1	acidic	39
Th(IV)- HO_2	Th(IV) peroxy-complexes	H_2O_2	0.1	acidic	39
U(VI)	U(VI)peroxy-complexes	$\text{H}_2\text{O}_2/\text{HClO}_4$	0.1 : 0.1	acidic	40
U(VI)- HO_2	U(VI)peroxy-complexes	$\text{H}_2\text{O}_2/\text{HClO}_4$	0.1 : 0.1	acidic	40
Np(V)	Np(IV) or Np(V/VI) peroxy-complex	-	-	acidic	41
Am(IV)	Am(V)/Am(IV) peroxy-complexes	-	-	-	37
	Am(V)	HNO_3	1.0-9.0	acidic	23

Table S6. Table summarizing γ -irradiated actinides Np and its redox chemistry with O_2^\cdot .

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions	Ref.	
			Conc. (M)	pH	
Np(VI)	Np(V) or Np(V/VI) peroxy-complex	-	-	acidic	42
		LiOH	0.1-1.0	basic	42

Table S7. Table summarizing γ -irradiated actinides Np and its redox chemistry with O_3^\cdot .

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions	Ref.	
			Conc. (M)	pH	
Np(IV)	unknown	LiOH	0.2-2.0	basic	43
Np(V)	unknown	LiOH	0.2-2.0	basic	43

Table S8. Table summarizing γ -irradiated actinides U-Am and their redox chemistry with H_2O_2 .

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions	Ref.	
			Conc. (M)	pH	
U(III)	U(IV)	HClO_4	0.5	acidic	33
U(IV)	U(V)	HClO_4	1.0	acidic	44
		H_2SO_4	0.98	acidic	44
		H_2SO_4	0.4	acidic	44
		HCl/NaCl	0.05 : 1.95	acidic	44
		HCl/NaCl	0.1 : 1.9	acidic	44
		HCl/NaCl	0.25 : 1.75	acidic	44
		HCl/NaCl	0.316 : 1.684	acidic	44
		HCl/NaCl	0.5 : 1.5	acidic	44
		HCl/NaCl	1.0 : 1.0	acidic	44
		HCl/NaCl	1.5 : 0.5	acidic	44
		HCl/NaCl	0.5 : 1.5	acidic	44
U(V)	U(VI)	-	-	acidic	44
U(VI)	U(VI) peroxyo-complexes	-	-	acidic	6
Np(III)	Np(IV)	HClO_4	-	acidic	45
Np(IV)	Np(V)	HNO_3	-	acidic	46
Np(V)	Np(IV)	HNO_3	6.0-8.4	acidic	47
Np(VI)	Np(V)	-	-	-	42
		LiOH	0.1-13.7	basic	42
		HClO_4	-	acidic	45
		HNO_3	7.5	acidic	6
Np(VII)	Np(VI)	-	-	-	42
		LiOH	0.1-13.7	basic	42
Pu(III)	Pu(IV)	H_2SO_4	-	acidic	48
Pu(IV)	Pu(III)	HClO_4	1.0	acidic	49
		HClO_4	1.5	acidic	50
		$\text{HClO}_4/\text{Na}_2\text{SO}_4$	0.3 : 0.4	acidic	51
		HNO_3	0.98-1.96	acidic	52
Pu(VI)	Pu(V) or Pu(V)/(VI)peroxyo-complexes	HClO_4	1.0	acidic	45
		HNO_3	1.0	acidic	45
		H_2SO_4	0.18	acidic	45
		NaHCO_3	0.05	basic	53
		NaHC_3	0.05	basic	53
Am(IV)	Am(III)	-	4.0	acidic	37
Am(V)	Am(IV)	HClO_4	0.1-0.2	acidic	45
Am(VI)	Am(V)	HClO_4	0.98	acidic	54
		HNO_3	1.0-9.0	acidic	23

Table S9. Table summarizing α -irradiated and multi-source irradiation of U systems and its redox chemistry under various conditions.

Starting Oxidation State	Resulting Oxidation State	Conditions			Radiation Source	Ref.	
		Medium	Conc. (M)	pH			
U(IV)	U(IV)	HClO ₄	0.2-11.5	acidic	α	55	
		HNO ₃	0.5-8.0	acidic	α	55	
	U(VI)	dearaerated H ₂ SO ₄	0.4-1.5	acidic	α	41	
			0.4	acidic	α	30	
		H ₂ SO ₄ / sat. O ₂	0.4	acidic	α	30	
	[(U(VI)O ₂) $(\mu_2,\eta^2-$ O ₂)(H ₂ O) ₂] \cdot 2H ₂ O	-	3.8	α	56-58		
		dearated	-	-	α	59	
		Ar/H ₂ atm	-	-	α	59	
		aerated	-	-	α	59, 60	
			H ₂ O ₂	conc.	-	61	
	U(VI)O _{2.33}	-	-	-	$\alpha/\beta/\gamma/n$	62-65	
commercial SNF	U(VI) ₃ O ₇	-	-	-	internal $\alpha >$ 250 μ Ci	62	
	[(UO ₂) ₈ O ₂ (OH) ₁₂] \cdot 10H ₂ O	-	-	-	$\alpha/\beta/\gamma/n$	66	
		[(U(VI)O ₂) $(\mu_2,\eta^2-$ O ₂)(H ₂ O) ₂] \cdot 2H ₂ O	2 years of water exposure	-	$\alpha/\beta/\gamma/n$	65	
			2 years of water exposure	-	$\alpha/\beta/\gamma/n$	65	
	U(IV)(SO ₄) ₂	U(IV)	dearaerated H ₂ SO ₄	0.1-18	0.1- 1.95	$\alpha/\beta/\gamma/n$	67
		U(VI)	aerated H ₂ SO ₄	0.1-18	0.1- 1.95	$\alpha/\beta/\gamma/n$	67
U(IV)/U(VI)	U(VI)	H ₂ SO ₄	-	-	α	41	
U(VI)	U(VI)	H ₂ SO ₄	-	acidic	α	41	
U(VI)O ₂ SO ₄	U(VI)O ₂ SO ₄	U(VI)O ₂ SO ₄ solution	conc.	0.1- 3.26	$\alpha/\beta/\gamma/n$	67	
U(VI)O ₂ F ₂	U(VI)O ₂ F ₂	U(VI)O ₂ F ₂ solution	conc.	1.35- 4.25	$\alpha/\beta/\gamma/n$	67	
enriched U(VI)O ₂ (NO ₃) ₂	enriched U(VI)O ₂ (NO ₃) ₂	enriched U(VI)O ₂ (NO ₃) ₂ solution	conc.	0.60- 2.05	$\alpha/\beta/\gamma/n$	67	

Table S10. Table summarizing γ -irradiated actinides U-Am and their redox chemistry with non-specified reactive species.

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions	Ref.	
			Conc. (M)	pH	
Th(IV)(NO ₃) ₄	Th(IV)	HNO ₃	5.0	1.21-5.9	68
		NaNO ₃	5.0	0.5-11	68
U(IV)	U(IV)	HNO ₃	0.5-3.0	acidic	69
	U(VI)	H ₃ PO ₄	14.8	acidic	70
U(VI)	U(IV)O ₂ nanoparticles	deaerated uranyl nitrate/10% propanol	0.01	3.4	71
	U(VI)	aerated uranyl nitrate/10% propanol	0.01	3.4	71
Np(IV)	Np(IV)	HNO ₃	0.4-1.0	acidic	72-74
	Np(V)	HClO ₄	1.0	≥ 4	72-74
		XeO ₃	-	≤ 1	72-74
		N ₂ O	-	≥ 4	72-74
	Np(V)/Np(VI)	NaOH	-	basic	75
Np(VI)	Np(IV)/Np(V)	NaOH	0.9-8.5	basic	74
	Np(IV) precipitate	NaOH/N ₂ O	1.0-3.0	basic	72-74
	Np(V)	NaOH	0.9-8.5	basic	74
	Np(VI)	NaOH	0.9-7.0	basic	72-74
		NaOH	-	basic	75
		NaOH/BrO ₄ ⁻	1.0-3.0 : 0.02	basic	72-74
		NaOH/BrO ₄ ⁻	0.2-3.8 : 0.02	basic	72-74
		NaOH/Na ₂ CO ₃ /N ₂ O	1.0 : 0.5	basic	72-74
		NaOH/Na ₂ CO ₃ /N ₂ O	0.1 : 0.5	basic	72-74
		N ₂ O/CO ₃ ²⁻	-	basic	72-74
		HClO ₄	-	acidic	72-74
	Np(VII)	NaOH/Na ₂ S ₂ O ₈	0.05-2.0 : 0.02	basic	72-74
Pu(III)	Pu(III)	deaerated HClO ₄	0.2-0.5	acidic	41, 76
		HCl	0.8	acidic	72-74
		HCl	6.00	acidic	72-74
		H ₂ SO ₄	0.4	acidic	41, 76
Pu(III)/Pu(IV)	Pu(III)/Pu(IV)	HCl	0.5-0.8	acidic	76
	Pu(III)	HClO ₄	0.2-0.5	acidic	76
Pu(IV)	Pu(III)/Pu(VI)peroxo-complexes	HClO ₄	0.2	acidic	76
Pu(IV)O ₂	Pu(III)	ground water	-	acidic	77
Pu(IV)/Pu(VI)	Pu(IV)	HNO ₃	4.0	acidic	78, 79
Pu(V)/Pu(VI)	Pu(IV)sulfate complexes	H ₂ SO ₄	-	acidic	76
Pu(VI)	Pu(IV)/Pu(V)	alkaline	1.3-6.9	basic	72-74
	Pu(IV)/Pu(VI)	HCl	0.2	acidic	41

Pu(VI)*/Pu(VI)	Pu(IV)	alkaline	1.0-6.0	basic	75, 80
	Pu(V)	HNO ₃	0.3-4.0	acidic	81
	Pu(VII)	HCl	0.2	acidic	72-74
		alkaline/N ₂ O	-	basic	16, 82
		NaOH/K ₂ S ₂ O ₈	1.0	basic	83, 84
Pu(VII)	Pu(VI)	alkaline	0.87-3.5	basic	76
Am(III)	Am(V)	sat. N ₂ O/HClO ₄	4.0	acidic	41
Am(VI)	Am(III)	H ₃ PO ₄	3.0	acidic	41
	Am(VII)	NaOH with N ₂ O or Na ₂ S ₂ O ₈	1.0-3.0	basic	41, 85
Cm(III)	Cm(II)	HClO ₄ /(CH ₃) ₃ OH	0.1 : 0.1	5.3-6.0	24
Cm(IV)	Cm(III)	sat. N ₂ O/HClO ₄ /(CH ₃) ₃ OH	0.1 : 0.1	5.3-6.0	24

Table S11. Table summarizing α -irradiated Pu systems and its redox chemistry under various conditions.

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions	Ref.
			Conc. (M)	pH
Pu(III)	Pu(III)	HNO ₃ /HNO ₂ /NO ₂	-	acidic
	Pu(IV)	HNO ₃	> 1.0	acidic
		HNO ₂	1.0-5.0	acidic
		H ₂ SO ₄	-	acidic
	Pu(IV) peroxy-complexes/Pu(VI)	HNO ₃	0.12	acidic
		HNO ₃ /HNO ₂ /NO ₂	-	acidic
	Pu(IV)	HCl	-	acidic
		HCl/HClO ₄	-	acidic
		HClO ₄	1.0	acidic
Pu(IV)	Pu(IV)	HNO ₃	4.0-6.0	acidic
	Pu(VI)	HNO ₃	6.0	acidic
	Pu(VI)/Pu(IV)	HNO ₃	4.0-6.0	acidic
	Pu(IV) nitrate complexes	HNO ₃	≥ 6.0	acidic
		NaOH	< 3.0	basic
	Pu(V)	NaOH	> 3.0	basic
	Pu(VI)	HClO ₄	1.0	acidic
	Pu(IV)/Pu(VI)	LiClO ₄	0.1	acidic
	Pu(IV) colloids/Pu(VI)	HNO ₃ /HNO ₂ /NO ₂	-	acidic
	Pu(VI)	HNO ₃	-	acidic
Pu(VI)	Pu(III)	HClO ₄	0.2	acidic
		HCl	0.5	acidic
	Pu(III)/(IV)	HNO ₃	< 1.0	acidic
	Pu(IV)	-	-	3.8
		H ₂ SO ₄	0.05-2.0	acidic

		HClO ₄	0.2	acidic	41
		HCl	0.5	acidic	89
Pu(IV) colloids		NaNO ₃	-	3.0	76
		LiClO ₄	0.1	acidic	96
	Pu(V)	HClO ₄	0.48-1.9	acidic	76
		HClO ₄	0.2	acidic	41
Pu(VI)		HClO ₄ /NaClO ₄	0.516 : 0.5	1.01-7.32	100
		HClO ₄ /NaClO ₄	0.101 : 0.9	1.01-7.32	100
		HNO ₃	1.0-4.0	acidic	76, 98
		HNO ₃ /HNO ₂ /NO ₂	-	acidic	86
		HCl	0.5	acidic	89
	Pu(VI)	HCl	0.062-0.5	acidic	101
Pu(VI)*/Pu(VI)	Pu(VI) dimers/Pu(V)	-	-	-	102

Table S12. Table summarizing α -irradiated Am systems and its redox chemistry under various conditions.

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions	Ref.	
			Conc. (M)	pH	
Am(III)	Am(IV)/Am(V)	HNO ₃ /Na ₂ S ₂ O ₈	-	-	103
		HNO ₃ or NaNO ₃	0.5	-	104
Am(III)	Am(V)	NaCl	5.0	basic	41, 105
		K ₂ CO ₃	3.0-5.5	basic	41
		(NH ₄) ₂ S ₂ O ₈ /HNO ₃	0.2	acidic	106, 107
Am(IV)	Am(III)	HClO ₄	0.2	acidic	106, 107
		H ₂ SO ₄	0.2	acidic	106, 107
		H ₃ PO ₄	10.0	acidic	41
		HClO ₄	2.0-4.0	acidic	41
Am(V)	Am(III)	HNO ₃	0.4	acidic	41
		NH ₄ F	13	basic	41
		HClO ₄	0.1-2.0	acidic	41
		HClO ₄	0.2	acidic	108
		HClO ₄	1.0	acidic	106
		HClO ₄	2.0	acidic	109
		HClO ₄ /H ₃ PO ₄	0.1 : 0.01	acidic	41
		H ₂ SO ₄	1.0	acidic	41
		H ₂ SO ₄	1.0	acidic	109
		H ₂ SO ₄	0.1	acidic	106
Am(IV)	Am(V)	HNO ₃	0.5-9.0	acidic	41
		HCl	0.5	acidic	41
		HClO ₄	0.1	acidic	110
Am(VI)	Am(IV)	HClO ₄ or LiClO ₄	2.0	-	111
		HNO ₃ /Na ₂ S ₂ O ₈	-	-	103
Am(VI)	Am(III)	H ₃ PO ₄	3.0, 10.0-11.0	acidic	41

Am(IV) $P_2W_{17}O_{61}$ complex	$K_{10}P_2W_{17}O_{61}$	0.003-0.008	-	41
Am(V)	HClO ₄	0-12.0	acidic	41, 112
	HClO ₄	0.1-11.0	acidic	113
	HClO ₄ /NaClO ₄	0.2-1.0 : 7.0	-	113
	H ₂ SO ₄	0.1-10.0	acidic	41
	HNO ₃	0.1-4.7	acidic	41
	NaHCO ₃	-	basic	41
Na[Am(V)(CO ₃) ₃] precipitate	NaHCO ₃	0.1	basic	114
	Na ₂ CO ₃	0.1-2.0	basic	114
	K ₂ CO ₃	-	basic	41
	HClO ₄ or LiClO ₄	1.0	-	54

Table S13. Table summarizing α -irradiated multi-element systems and their redox chemistry under various conditions.

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions	Ref.
			Conc. (M)	pH
Am(III)/Cm(III)	Am(V)/An(III) hydroxides/Cm(III) hydroxides	sat. NaClO ₄	-	acidic 115
Pu(IV)O ₂ / Am(IV)(OH) ₄	Pu(VI)/Am(IV)	NaCl (Ar atm)/HClO ₄	1.0-5.0	< 7.0 94
	Pu(VI)/Am(V)	NaCl (Ar atm)/HClO ₄	5	> 7.0 94
		NaCl (ambient atm)/HClO ₄	1.0-5.0	2.0-12.0 94
Pu(IV)/U(IV) (MOX47 fuel)	Pu(V)precipitate/U(VI)	-	0	- 116
	Pu(V)/U(VI)O ₄ ·4H ₂ O	H ₂ O ₂	6.0x10 ⁻⁵	acidic 116

Table S14. Table summarizing α -irradiated Cm systems and its redox chemistry under various conditions.

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions	Ref.
			Conc. (M)	pH
Cm(IV)	Cm(IV)	CsF	15.0	basic 41
Cm(IV) or Cm(IV) $P_2W_{17}O_{61}$ complex	Cm(III)	$K_{10}P_2W_{17}O_{61}$	-	1.5-6.1 41, 117

Table S15. Table summarizing self-irradiated (β^-) Bk systems and its redox chemistry under various conditions.

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions		Ref.
			Conc. (M)	pH	
Bk(III)	Bk(IV)	K ₂ CO ₃	2.0	basic	41
Bk(IV)	Bk(III)	H ₂ SO ₄	-	-	41, 118-120
		H ₂ SO ₄	0.05-4.5	acidic	121
		H ₂ SO ₄	1.0	acidic	122
		H ₂ SO ₄ /Na ₂ SO ₄	0.1 : 1.0	acidic	121
		HClO ₄	0.1-5.0	acidic	121
		HClO ₄	1.0-5.0	acidic	118
		HClO ₄	1.0	acidic	122
		HClO ₄	1.0-8.0	acidic	123, 124
		HClO ₄ /HNO ₃	-	1.0	124
		HNO ₃	0.5-6.0	acidic	118
Bk(III)pyrophosphate	Bk(IV)	HNO ₃	-	acidic	125
		HNO ₃	9.0	acidic	119
		K ₂ CO ₃	2.0	basic	122
		HNO ₃ /AgNO ₃ /(NH ₄) ₂ S ₂ O ₈	4.0 : 0.01 : 0.1	acidic	120
		Na ₄ P ₂ O ₇ /H ₂ SO ₄	0.12	1.5	121

Table S16. Table summarizing α -irradiated Cf systems and its redox chemistry under various conditions.

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions		Ref.
			Conc. (M)	pH	
Cf(IV)	Cf(III)	K ₂ P ₂ W ₁₇ O ₆₁ /K ₂ S ₂ O ₈	-	-	41
Cf(IV)P ₂ W ₁₇ O ₆₁ complex	Cf(III)	K ₂ P ₂ W ₁₇ O ₆₁		2.5-6.1	117

Table S17. Table summarizing γ -irradiated actinides Np-Cm and their redox chemistry with NO_3^- .

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions	Ref.	
			Conc. (M)	pH	
Np(V)	Np(V)	HNO ₃	0.09-4.0	acidic	126
		HNO ₃ /LiNO ₃	1.0 : 3.0	acidic	127
		HNO ₃ /LiNO ₃	1.0 : 6.0	acidic	127
	Np(VI)	HNO ₃	1.0-5.0	acidic	127
		HNO ₃	0.09-4.0	acidic	36, 128, 129
		HNO ₃	4.0	acidic	36
Np(VI)	Np(V)	HNO ₃	0.09-4.0	acidic	126, 127
		HNO ₃	0.5-6.0	acidic	130
		HNO ₃	0.001-2.0	acidic	72
		HNO ₂	0.09-4.0	acidic	36, 128, 129
		HNO ₃	2.0	acidic	131
Pu(III)	Pu(IV)	HNO ₃	-	acidic	132
		dearated HNO ₃	-	acidic	132, 133
		HNO ₂	-	acidic	132
		NO ₂ /N ₂ O ₄ from HNO ₃ /NO ₃ ⁻	-	acidic	132
		HNO ₃	0.3-1.0	acidic	134
Pu(IV)	Pu(III)/Pu(VI)	HNO ₃	1.0-15.0	basic	135
	Pu(IV)OH ₄	NaOH/NO ₃ ⁻	-	acidic	97
	Pu(VI)	HNO ₂	0.001-0.5	acidic	76, 97
Pu(VI)*/Pu(VI)	Pu(VI)	HNO ₃	> 8.0	acidic	81
Am(V)	Am(IV)	HNO ₃	2.5	acidic	136
Am(VI)	Am(V)	HNO ₃	1.0-9.0	acidic	23
Cm(III)	Cm(IV)	sat. N ₂ O/HNO ₃	4.5-6.0	acidic	32
Cf(III)	Cf(III)	HNO ₃	6.0	acidic	27

Table S18. Table summarizing α -irradiated Np systems and its redox chemistry under various conditions.

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions	Ref.	
			Conc. (M)	pH	
Np(IV)	Np(IV)/Np(V)/Np(VI)	HNO ₃	> 1.0	acidic	137
		HClO ₄	< 1.0	acidic	138
		HNO ₃	0.11	acidic	137, 139
	Np(VI)	HNO ₃	0.33-0.62	acidic	137, 139
		HClO ₄	> 1.0	acidic	138
		HClO ₄	0.48-1.70	acidic	140
Np(VI)	Np(V)				

Table S19. Table summarizing γ -irradiated actinides U-Am and their redox chemistry with CO_3^{2-} radical species.

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions	Ref.	
			Conc. (M)	pH	
U(IV)	U(IV)carbonate complexes	sat. KHCO_3	-	basic	18
		K_2CO_3	0.05	basic	9
Np(III)	Np(III) carbonate complexes	K_2CO_3	1.0-3.0	basic	2
Np(IV)	Np(IV) carbonate complexes	CO_3^{2-}	-	basic	141
Np(V)	Np(V) carbonate complexes	Na_2CO_3	0.5 : 1.0	basic	9
		$\text{Na}_2\text{CO}_3/\text{NaOH}$	0.5 : 0.2	basic	38
			0.5 : 0.6	basic	38
			0.5 : 1.0	basic	38
		CO_3^{2-}	-	basic	73
Np(VI)	Np(VI) carbonate complexes	Na_2CO_3	-	basic	73
		NaHCO_3	1.0	basic	72-74
		$\text{Na}_2\text{CO}_3/\text{NaHCO}_3$	0.25 : 0.75	basic	72-74
		$\text{Na}_2\text{CO}_3/\text{NaOH}$	0.5 : 1.0	basic	38
		sat. $\text{N}_2\text{O}/\text{CO}_3^{2-}$	-	basic	38
Np(VII)	Np(VII)	$\text{Na}_2\text{CO}_3/\text{NaOH}$	0.5 : 0.2	basic	142
			0.5 : 0.6	basic	152
			0.5 : 1.0	basic	152
Pu(III)	Pu(IV)	K_2CO_3	1.0-4.3	basic	2
Pu(IV)	Pu(V)	CO_3^{2-}	-	basic	141
Pu(V)	Pu(V)	Na_2CO_3	0.05	basic	9
Pu(VI)	Pu(VI)peroxy-carbonate complexes	Na_2CO_3	0.01-0.005	basic	143
Am(III)	Am(III) carbonate complexes	K_2CO_3	3.0-6.0	basic	2

Table S20. Table summarizing γ -irradiated actinides U-Am and Cf and its redox chemistry with SO_4^{2-} .

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions		Ref.
			Conc. (M)	pH	
U(IV)	U(V)	deaeerated	-	acidic	144
		$\text{H}_2\text{SO}_4/\text{HClO}_4$			
U(V)	U(VI)	H_2SO_4	2.0-5.0	acidic	145
		H_2SO_4	1.5	acidic	146
Np(III)	Np(IV)	H_2SO_4	3.0	acidic	147
Np(IV)	Np(V)	H_2SO_4	-	acidic	141
		H_2SO_4			
Np(V)	Np(VI)	H_2SO_4	0.01-0.4	acidic	72-74
Pu(III)	Pu(IV)	H_2SO_4	2.0	acidic	127
Pu(IV)	Pu(V)	H_2SO_4	5.0	acidic	131
		H_2SO_4	-	acidic	141
Am(III)	Am(IV)	$\text{H}_2\text{SO}_4/\text{Na}_2\text{S}_2\text{O}_8$	1.0 : 1.0	acidic	144
Cf(III)	Cf(IV)	$\text{K}_2\text{S}_2\text{O}_8/\text{HClO}_4$	0.1 : 0.003	-	148

Table S21. Table summarizing γ -irradiated actinides U-Am and Cf and their redox chemistry with the molecular halogen anions.

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions	pH	Ref.
			Conc. (M)		
U(III)	U(IV)	Cl_2^- from HClO_4/HCl	0.5 : 0.1	acidic	33
		Br_2^- from $\text{HClO}_4/\text{NaBr}$	0.5 : 0.1	acidic	33, 136, 149
		I_2^- from HClO_4/KI	0.9 : 0.1	acidic	
		aerated HCl	-	acidic	41
U(IV)	U(V)	deareated HCl	-	acidic	41
		HCl	0.8	acidic	150
U(V)	U(VI)	Cl_2^- from $\text{HClO}_4/\text{HCl}/\text{UO}_2(\text{ClO}_4)_2$	0.1 : 0.7 : 0.1	acidic	136
		Cl_2^- from NaCl	1.0	acidic	151
Np(III)	Np(IV)	Cl_2^- from HCl/HClO_4	0.1 : 0.9	acidic	152
		Br_2^- from HBr	1.0	acidic	152
		I_2^- from NaI/HClO_4	0.1 : 0.9	acidic	152
Np(V)	Np(VI)	Cl_2^- from HCl	1.0-3.0	acidic	152
		Cl_2^- from HCl/LiCl	0.5 : 5.0	acidic	152
		Cl_2^- from HCl/LiCl	0.9 : 4.0	acidic	152
		Cl_2^- from $\text{HCl}/\text{LiClO}_4$	0.7 : 1.5	acidic	152
		Cl_2^- from HCl/MgCl_2	0.8 : 3.0	acidic	152
		Cl_2^- from HCl/MgCl_3	1.0	basic	151
		Cl_2^- from HCl/MgCl_4	1.0	basic	152
Pu(III)	Pu(IV)	Cl_2^- from HCl	1.0-5.0	acidic	153
		Cl_2^- from HCl/HClO_4	0.01 : 5.0	acidic	153
		Cl_2^- from $\text{HCl}/\text{H}_2\text{SO}_4$	0.01 : 2.5	acidic	153
		Cl_2^- from NaCl	1.0	basic	151
Am(III)	Am(IV)	Cl_2^- from NaCl	1.0	basic	151
Cf(III)	Cf(IV)	Cl_2^- from $\text{NaCl}/\text{HClO}_4/\text{K}_2\text{S}_2\text{O}_8$	1.0 : 0.003 : 0.1	-	148

Table S22. Table summarizing γ -irradiated actinides in multi-element systems and their redox chemistry with non-specified reactive species.

Starting Oxidation State	Resulting Oxidation State	Medium	Conditions	Ref.	
			Conc. (M)	pH	
Gd-doped U(IV)O ₂	UO ₂ /U(VI)	H ₂ O ₂	-	acidic	154
Pu(VI)/U(VI)/Pd(II)	Pu(V)/U(V)/Pd(II)	HNO ₃	4.0	acidic	78
Pu(VI)/Pd(II)	Pu(VI)/Pu(IV)/Pd(II)	HNO ₃	4.0	acidic	78
Pu(IV)/Np(VI)	Pu(V)	HNO ₃	0.5	acidic	155
Pu(VI)/Np(VI)	Pu(VI)/Np(V)	HNO ₃	0.01-0.1	acidic	156
	Pu(V)/Np(V)	HNO ₃	0.01-0.1	acidic	156
Pu(IV)/U(IV) (MOX47 fuel)	Pu(V)/U(VI) colloids	-	0	-	116

Solid-State Chemistry

Table S23. Summary of radiation studies on Pu solid-state materials.

Starting Complex	Resulting Complex/Species	Radiation Source (energy/dose)	Atmosphere	Conditions	Temperature (°C)	Ref.
δ -Pu	Frenkel defects	self-radiation	ambient	< 25	157	
PuAl ₂	Frenkel defects	self-radiation	ambient	< 25	157	
PuGa ₃	Frenkel defects	self-radiation	ambient	< 25	157	
PuCoGa ₅	Frenkel defects	self-radiation	ambient	< 25	157	
	> 0.1 Å distortions + atom displacements	self-radiation	ambient	-255	158, 159	
PuCoIn ₅	Frenkel defects	self-radiation	ambient	< 25	157	
PuPt ₂ In ₇	Frenkel defects	self-radiation	ambient	< 25	157	
Pu ₂ PtGa ₈	Frenkel defects	self-radiation	ambient	< 25	157	
Pu-doped CaZrTi ₂ O ₇	Pu-O bond contraction	self-radiation	ambient	20-25	160	
CaF ₂ + 0.7 wt% Pu(III)	CaF ₂ + Pu(III)/Pu(IV)	γ	ambient	0-25	161	
	CaF ₂ + Pu(III)/Pu(IV)	self-radiation	ambient	25	162	
PuF ₆	PuF ₄ + F ₂	γ (1 MGy)	ambient	60-70	163	
	PuF ₄ + F ₂ / PuF ₆	γ (1 MGy)	He	60-70	163	
	PuF ₄ + F ₂ / PuF ₄ + KrF ₂	γ (1 MGy)	Kr	60-70	163	

Table S24. Summary of radiation studies on Th solid-state materials.

Starting Complex	Resulting Complex/Species	Radiation Source (energy/dose)	Conditions		Ref.
			Atmosphere	Temperature (°C)	
ThO ₂	ThO ₂ + O vacancies	γ (0.5-2 MGy)	ambient	25	164-166
		H ⁺ (18 MeV)	ambient	50	167
		¹⁶ O (84 MeV)	ambient	50	167
		He ⁺ (100 keV)	ambient	25	168
		Ag ⁺ (100 MeV)	ambient	25	168
	ThO _{2-x} + ThO _{2+x} + Th ₄ O ₉	H ⁺ (2 MeV)	ambient	600	169
	ThO ₂ + dislocation loops anion + cation vacancies/interstitials	H ⁺ (2 MeV)	ambient	600	170
Th ₄ (PO ₄) ₄ P ₂ O ₇	ThO ₂ + Pb impurities	e ⁻ (3 MeV)	ambient	< 100	172
	phosphate-based radicals	γ (1 MGy)	ambient	20-22	173
		Kr (840 MeV)	-	25	174
	ThO ₂ + (ThO) ₃ (PO ₄) ₂ + P ₄ O ₁₀	He ⁺ (1.6 MeV)	ambient	20-23	173
		Pb ²⁺ (200 keV)	ambient	20-24	173
		Au ³⁺ (5 MeV)	ambient	20-25	173

Table S25. Summary of radiation studies on U solid-state materials.

Starting Complex	Resulting Complex/Species	Radiation Source (energy/dose)	Atmosphere	Conditions Temperature (°C)	Ref.
U(III)					
CaF ₂ + doped with U	CaF ₂ + U(IV)	γ	ambient	0-25	161
U(IV)					
UO ₂	dislocation loops + lines	I (0.9 MeV) Fe (27 MeV) I (0.9 MeV) + Fe (27 MeV) 10 ¹³ Kr (740 MeV/cm ²)	ambient ambient ambient	25 25 26.85	175 175 176
(U,Pu)O ₂	UO ₂ + U(V) + O vacancies T _{2g} Raman blue-shift + UO _{2+x} + U ₄ O ₉	He ²⁺ (25 MeV)	ambient	25	177
(U,Am)O ₂	T _{2g} Raman blue-shift + UO _{2+x} + U ₄ O ₉ U(IV)/(V) + Am(III)/(IV) in (U,Am)O ₂	Pu (<i>in-situ</i> α)	inert	25	178
(U _{0.92} Ce _{0.08})O ₂	(U _{0.92} Ce _{0.08})O ₂ + U(V) + O vacancies	Am (<i>in-situ</i> α) 10 ¹³ Kr (740 MeV/cm ²)	air or Ar/H ₂ ambient	25 26.85	179, 180 176
UN	UN + U(V) + O vacancies	3x10 ¹³ Cd (1 GeV/cm ²)	ambient	26.85	176
U(V)					
CaO doped with U	Ca ²⁺ vacancy + O ⁻	γ (4 kGy)	-	-196.15-266.85	181
U(VI)					
γ-UO ₃	U(IV)	Xe (167 MeV) Au (950 MeV)	ambient ambient	25 25	166 166
(UO ₂)(OH) ₂ + (UO ₂) ₈ O ₂ (OH) ₁₂ ·10H ₂ O	U(IV)	Xe (167 MeV) Au (950 MeV)	ambient ambient	25 25	166 166
K[UO ₂ (NO ₃) ₃]	monodentate nitrate defects	γ (20 kGy)	vacuum	25	182
Rb[UO ₂ (NO ₃) ₃]	monodentate nitrate defects	γ (20 kGy)	vacuum	25	182
Cs[UO ₂ (NO ₃) ₃]	monodentate nitrate defects	γ (20 kGy)	vacuum	25	182
Li ₄ [UO ₂ (O ₂) ₃]·10H ₂ O	Li ₂₄ [UO ₂ O ₂ (OH)] ₂₄	γ (3 MGy)	ambient	25	183
Li ₂₄ [UO ₂ O ₂ (OH)] ₂₄	Li ₂ [(UO ₂) ₃ O ₂ (OH)] ₂ ·7H ₂ O	γ (4 MGy)	ambient	30	184
Ca ₂ [UO ₂ (O ₂) ₃]·9H ₂ O	Ca ₂ [UO ₂ (O ₂) _{3-x} (OH) _{2x}]	γ (3 MGy)	ambient	25	183
U ₆₀	weakened UO ₂ ²⁺ bond	γ (1 MGy)	ambient	25	185
U ₆₀ Ox ₃₀	U ₂ O ₇	He ²⁺ (50 MGy)	ambient	25	185
	weakened UO ₂ ²⁺ bond	γ (1 MGy)	ambient	25	185
	breakdown of cluster	He ²⁺ (50 MGy)	ambient	25	185
U ₂₄ Pp ₁₂	weakened UO ₂ ²⁺ bond	γ (1 MGy)	ambient	25	185

[UO ₂ O ₂ (H ₂ O) ₂]·2H ₂ O	U(IV)/U(V)/U(VI) weakened UO ₂ ²⁺ bond	He ²⁺ (50 MGy) γ (1 MGy)	ambient ambient	25 25	185 185
Na ₄ [UO ₂ (O ₂) ₃]·9H ₂ O	[UO ₂ O ₂ (H ₂ O) ₂] + U ₂ O ₇ Na ₄ [UO ₂ (CO ₃) ₃]	He ²⁺ (50 MGy) He ²⁺ (225 MGy) γ (3 MGy) He ²⁺ (225 MGy)	ambient ambient hydrated Ar hydrated Ar	25 25 25 25	186 186 186 186
[(UO ₂) ₈ O ₂ (OH) ₁₂]·10H ₂ O	γ -UO ₃ + U ₂ O ₇	He ²⁺ (100 MGy)	ambient	25	187
	[UO ₂ O ₂ (H ₂ O) ₂]·2H ₂ O	He ²⁺ (100 MGy)	ambient	> 25	187

Table S26. Summary of radiation studies on Am and Cm solid-state materials.

Starting Complex	Resulting Species or Defects	Conditions		Ref.
		Atmosphere	Temperature (°C)	
Am				
CaF ₂ + doped with Am(III)	CaF ₂ + Am(II)	ambient	0-25	161
CaF ₂ + 0.1-0.2 wt% Am(III)	CaF ₂ + Am(II) + e ⁻	ambient	25	188
Am ₂ Zr ₂ O ₇ (<i>Fd</i> -3 <i>m</i>)	<i>Fm</i> -3 <i>m</i> symmetry + cation antisites + Frenkel pairs	N ₂	25	189-191
AmPO ₄	Am(III)/Am(IV)	ambient	25	192
Am _{0.93} Np _{0.07} PO ₄	local P disorder	ambient	25	193
U _{1-x} Am _x O _{2±δ} (x = 0.15 and 0.20)	increased interatomic distances	ambient	25	179, 194, 195
AmO ₂	Am-O bond increase	ambient	25	196-200
AmAlO ₃ (<i>R</i> -3 <i>c</i>)	<i>Pm</i> -3 <i>m</i> symmetry	ambient	25	201
Cm				
CaF ₂ + doped with Cm(III)	CaF ₂ + Cm(IV)	ambient	0-25	161
cubic-C Cm ₂ O ₃	hexagonal-A Cm ₂ O ₃ + CmO ₂	He	25	202
rhombohedral CmAlO ₃	cubic CmAlO ₃	ambient	25-500	202

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