

THE CHEMICAL EFFECTS OF ALPHA DECAY IN U^{235} COMPLEX IONS

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(Received April 30, 1979)

The chemical effects of nuclear transformation associated with alpha decay of U^{235} have been studied in the U^{235} (IV) polyaminopoly carboxylic acid complexes of EDTA, DCTA, and DTPA. The observed break up values lie between a minimum of zero and a maximum of around 40%. These break-up values are dependent on certain ranges of pH and a different pH range is observed for each of the three complex systems studied. The addition of KI, a reducing agent, and an increase of U(IV) concentration (about 7 times its concentration in the other experiments) to investigate any reducing effect of U(IV) ion itself, does not alter the break-up value. The freezing of the reaction mixture, however, enhanced the break-up value to 56%. Considering the high recoil energies available following the alpha decay of U^{235} the break up values are thought to be low. It has been suggested that the possible localised oxidation of the U(IV) complex caused by the radiolysis products of water and the reducing effect of U(IV) ions, is responsible for the release of free ligand ions and the ultimate recombination with the daughter atoms, resulting in low break-up values.

INTRODUCTION

A number of studies have been published in which chemical effect studies of beta and gamma type nuclear transformations in the metal polyaminocarboxylate complexes have been described [1–11]. The nuclear transformations by alpha decay are accompanied by a different mode of excitation and energisation than that of beta and gamma transitions and they have not been extensively studied. Whereas, in the case of beta and gamma decays recoil energies being low, bond rupture has been attributed to the production of multiple charged states of the daughter atoms [1–11] (formed by processes like Auger effect, internal conversion and shake-off), the alpha decay effects may be considered to be result of primarily the recoil energy imparted to the daughter atom following alpha emission. In this work studies have been carried out to determine the role of these bond rupturing origins in the alpha decay of U^{235} which is complexed with the multi-dentate ligands; EDTA, DCTA and DTPA. Both uranium and its daughter thorium, being in the actinide family, have analogous chemical properties and they differ only by two in atomic number. Also uranium in its (IV) oxidation state and its daughter Th(IV) form very stable complexes [12, 13] with EDTA, DCTA and DTPA. Therefore it was possible to set up favourable conditions for the investiga-

tion of the chemical effects of alpha emission.

Alpha decay experiments were performed in the presence of free Cd^{2+} ions. Separate experiment were carried out to establish that in the experimental conditions of this study Cd^{2+} ions have slow exchange with either of the U(IV) and Th(IV) complexes and they are also able to stop the reported fast exchange [14] of free Th(IV), produced after alpha decay of U^{235} (IV) complex with the parent complexed ion.

EXPERIMENTAL

Reagents. Cd^{2+} stock solutions were prepared by the dissolution of accurately weighed amount of $CdCl_2$ (Koch Lab) in approximately $10^{-2}M$ HCl. The A. R. disodium salt of EDTA was dissolved in HCl and then neutralized by ammonia to precipitate the pure anhydrous H_4EDTA solid, whereas pure H_4DCTA and H_5DTPA were obtained by twice recrystallizing the commercial samples of the acids from hot water. U^{238} stock solutions were prepared by dissolving A.R. uranyl nitrate (B.D.H.) in $0.2M$ HCl, these solutions were then subjected to catalytic hydrogenation [15] in order to prepare U^{238} (IV) stock solutions. U^{235} tracer solutions in $3M$ HNO_3 were obtained from Radiochemical Centre, Amersham, England. HNO_3 was removed

Table 1. Radiochemical composition of the U^{235} solution.

Isotope	U^{235}	U^{234}	U^{236}	U^{238}
Percentage	93	0.75	0.25	6
Total alpha activity in nano curies in 200 mg sample	428	9130	29.78	3.97

by evaporation and the residue was redissolved in 0.2M hydrochloric acid. The solution was found to have traces of copper which interfered in catalytic hydrogenation of U(VI) to U(IV) and, therefore, these traces of copper were removed by an anion exchange separation [16] procedure prior to reduction. Another complication was experienced due to the radiochemical composition of the U^{235} sample. The sample contained high specific activity alpha emitting isotopes like U^{234} and U^{236} . Table 1 shows the distribution of the various isotopes of uranium in the Amersham U^{235} samples found by mass spectrometric analysis. The presence of these other alpha emitters complicated the radiochemical assay of the U^{235} atom, and hence that of the total daughter atoms, produced with in the decay period.

Cation Exchange Resin. Zeo-carb 225 cation exchange resin less than 200 mesh was used in the sodium form.

Analytical Techniques. U(IV) stock solutions were analysed by three methods to give identical results. They were: (1) potentiometric titrations [17] with standard solutions of $K_2Cr_2O_7$, (2) volumetric [17] titrations with standard $K_2Cr_2O_7$, and (3) the complexometric titrations [18] with standard EDTA solutions, whereas the solutions of uranous complexes of EDTA, DCTA and DTPA were analysed by volumetric titrations against standard ceric sulphate solutions [19]. The analysis of the carbon, hydrogen and nitrogen content of the complexes was carried out by the departmental microanalyst. The results were accurate within a limit of $\pm 0.3\%$.

Measurement of Radioactivity. All the samples were counted on the Packard 3310 Tricarb liquid scintillation counter. The two ml aqueous solution, to be counted, was mixed with 10 ml organic scintillation solution, prepared according to the formula of Bruno and Christian [20], without the occurrence of any phase separation. The chemical composition of each of the samples was kept constant in order to ensure the same degree of quenching. The discriminator levels of the instrument were set in such a way that the 4–5 meV alpha particle pulses of uranium isotopes were excluded and only 0.3 meV Th^{231} beta energy pulses were recorded; oxalic acid which was used to elute the Th^{231} free ions from the cation exchange column was

found to quench the liquid scintillation counting of Th^{231} . The extent of this quenching was experimentally determined and a proper allowance was made for it when oxalic acid containing activity was compared with the activity of the samples, not containing oxalate ions.

Procedure

Both the active and inactive U(IV) Y complexes were prepared by normal procedure [21]. The U(IV) content of both complexes was determined by analytical methods described earlier. C, H and N analysis of the active U^{235} (IV) complexes was not practicable, however U^{238} (IV) complexes were precipitated in identical condition and they were microanalysed. These precipitated complexes were dissolved in appropriate HCl solutions to give the required stock solutions at known pH.

The reaction mixtures were prepared in 100-ml volumetric flasks, sealed with serum caps to avoid oxidation. All the solutions were deoxygenated by bubbling with white spot nitrogen. The reaction mixtures contained the following:

Complex solutions of U^{235} (IV), 25–30 ml, at an appropriate pH, are maintained by varying amounts of acid concentration of the solution. It also contained one ml of the standard $CdCl_2$ solution which was introduced in the reaction mixture to initiate the reaction. Before the addition of Cd^{++} free ions all of the U^{235} (IV) and Th^{231} (IV) ions will be in the complex form. A decay period of seven days was given to ensure that almost all of the Th^{231} ($t_{1/2}$ 25.6 hr) already present had decayed out and fresh Th^{231} with measurable activity was grown in.

Total Activity Measurement. Two ml of the reaction mixture was counted on the Packard 3310 liquid scintillation counter within a limited discriminator level setting of A/B = 50/350 at an optimum gain of 11%. In a separate experiment it was ensured that this range of window opening of the instrument excludes essentially all of the alpha particles pulses from uranium isotopes. The activity recorded by the sample was taken to be the total activity, i.e. the total Th^{231} produced in the reaction mixture in any form, either complexed or uncomplexed.

Free Ion Activity Measurements. Within a period of seven days a measureable amount of Th^{231} activity would be expected to be accumulated in the free ion form, owing to the molecular disruption of the Th^{231} complexes of EDTA, DCTA and DTPA which are produced as a consequence of alpha decay of the corresponding U^{235} complexes. Therefore, a 5-ml aliquot of the reaction mixture

was taken out and forced through cation exchange column by nitrogen pressure, to absorb any Th^{231} free ions present in the reaction mixture. The cation exchange column was washed with deionized water and the effluent along with the washings was collected in a 25-ml volumetric flask (will be called effluent hereafter). The Th^{231} ions, absorbed on the column were then eluted with 2 ml 1M oxalic acid solution. Then the column was washed three times with a 1-ml portion of deionized water. The eluent and the washings were collected in a 5-ml volumetric flask (called eluent hereafter). Separate experiments had shown that 99.9% of the thorium was eluted by this method. Two ml of the eluent was then counted in the Packard scintillation counter in exactly the same way as the total activity sample. Free ion Th^{231} containing samples (eluent samples) were counted about every 400 min for the first three to four days and then at least twice a day for a total period of 10 days in order to obtain a precise decay curve of Th^{231} atoms. ($t_{1/2}$ 25.6 hr).

Effluent Activity Measurements. In the reaction mixture Th^{231} is always in radiochemical equilibrium with the parent U^{235} activity. When the mixture is passed through the ion exchange column it loses its Th^{231} free ions and hence the radioactive equilibrium is disturbed in the complex ion fraction of the reaction mixture which we get in the form of effluent. In order to reestablish the equilibrium, Th^{231} activity will be expected to grow in the effluent. Therefore, the growth of Th^{231} activity in the effluent was also followed by counting an aliquot of the complex ion fraction of the reaction mixture under the same conditions in which other two samples were counted. Growth of Th^{231} atoms in the effluent was followed until a constant value was obtained.

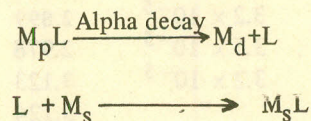
Check for the Extent of Oxidation in the Radioactive Mixture. Oxidation of the U(IV) complex in the reaction mixture was expected to yield free uranyl ions [21]. Therefore, a 2-ml aliquot of reaction mixture was passed through a cation exchange column in order to ensure the removal of the free uranyl cations from the reaction mixture. Total activity of the reaction mixture, the activity recorded by counting the sample in the Packard counter with a fully open window of 50 to infinity, at 11% gain, so that all the alpha particles of uranium could be counted, was compared with the total activity of the complex ion fraction, i.e. effluent, counted in the identical conditions. In those cases where there was not any difference in the alpha count rates of the reaction mixture and the effluent, it was assumed that oxidation has not taken place.

Checks for Precipitation and Adsorption. Two equal aliquots from the reaction mixture, at the end of each run

were counted with window settings of 50 to infinity and it was found that identical counts were registered for each sample and hence precipitation and adsorption were shown to be absent.

RESULTS

Free Cd^{2+} ions were introduced to the reactions mixtures, prepared for the study of alpha decay in U^{235} chelate complexes. These were added to scavenge the free ligands, generated by the expected bond rupture of the daughter Th^{231} complex ions produced as a consequence of alpha decay of corresponding U^{235} chelate complexes. In a separate set of experiments it was established that under these experimental conditions: (1) free Cd^{2+} ions have a slow rate of exchange with the U(IV) and Th(IV) complexes of EDTA, DCTA and DTPA and, therefore, undesirable redistribution of activities in the free ion and complexed ion form is not possible; (2) these free ions are able to stop the possible exchange [14] of free Th(IV) ion (tracer amount), produced after alpha decay of the U^{235} (IV) complexes, with the parent complex, as free Th(IV) ions were found to stick on cation exchange column when solutions of identical composition were passed through them. The reactions which proceed in the reaction mixture are considered to be the following:

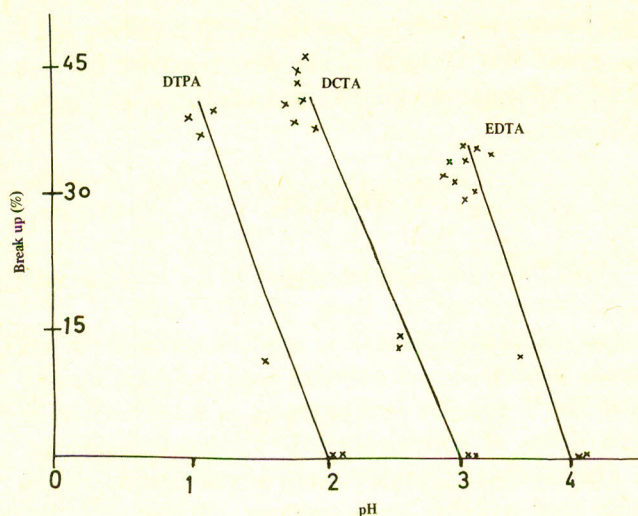


Where M_p , M_d and M_s stand for the parent, daughter and the scavenger metal ions respectively, and L, the free ligand. Therefore, the percentage bond break up was determined by the equation:

$$X = \frac{N_d^f}{N_d^t} \times 100$$

Where N_d^f is the number of daughter atoms observed in the free ions form as a consequence of molecular disruption because of alpha decay and N_d^t is the total number of such atoms generated (either free or bound, within the decay period).

The percentage break-up observed under the various conditions such as, variation of pH, ligand and freezing of the reaction mixture are shown in Tables 2 - 4 and Fig. 1 depicts the pH dependence of the 'X' values. In order to examine the likely effects, of the presence of a reducing agent on the percentage break up results, two separate experiments were carried out. Two reducing agents em-

Fig. 1. Results of $U^{235} \alpha Th^{231}$ experiments.Table 2. Percentage break-up results for the $U^{235}(IV)$ – EDTA system at 21°

$U^{235}(IV)$ EDTA (M)	Cd^{2+} (M)	pH ± 0.001	Break-up percentage $\pm 5\%$
6.60×10^{-4}	1.6×10^{-5}	3.234	33.1
8.81×10^{-4}	"	3.212	33.0
9.42×10^{-4}	"	2.988	33.9
1.93×10^{-3}	3.2×10^{-5}	2.899	32.1
2.08×10^{-3}	3.6×10^{-5}	2.978	32.5
2.18×10^{-3}	3.2×10^{-5}	3.123	34.9
1.30×10^{-3}	"	3.124	33.1
1.84×10^{-3}	"	4.236	0
2.28×10^{-3}	"	2.125	0
1.18×10^{-3}	1.6×10^{-5}	3.189	29.8
1.18×10^{-3}	"	3.189	30.2
8.80×10^{-3}	"	2.989	33.0
1.30×10^{-3}	"	3.725	8.0

ployed for this study were KI and U(IV) itself, thought to have more powerful reducing effect as compared to $Na_2S_2O_3$ used by Asano [11]. The results of the per cent break-up observed in the presence of $10^{-2}M$ KI and in the presence of 7 times excess U(IV) in the form of $U^{238}(IV)$ DTPA are shown in Tables 6 and 7 which are not different from the earlier results. However, the freezing of the reaction mixture of U(IV) DTPA system at pH 1 enhanced the percentage break-up value as shown in the Table 5.

DISCUSSION

From these results the following points would appear

Table 3. Percentage break-up results for the $U^{235}(IV)$ DCTA system at 21°

$U^{235}(IV)$ DCTA (M)	Cd^{2+} (M)	pH ± 0.001	Break-up percentage $\pm 5\%$
1.26×10^{-3}	3.2×10^{-5}	3.188	0
6.05×10^{-4}	1.6×10^{-5}	1.889	41.3
6.40×10^{-4}	"	1.983	42.7
6.40×10^{-4}	"	1.994	41.7
2.00×10^{-3}	3.2×10^{-5}	2.123	41.9
4.18×10^{-4}	1.60×10^{-5}	1.925	44.1
1.15×10^{-3}	6.4×10^{-5}	2.246	40.0
5.73×10^{-4}	"	2.112	41.7
1.78×10^{-3}	"	2.598	13.7
1.72×10^{-3}	"	1.598	10.9
1.56×10^{-3}	"	1.562	44.5
1.32×10^{-3}	"	3.135	0

Table 4. Percentage break-up results for the $U^{235}(IV)$ – DTPA system at 21° .

$U^{235}(IV)$ DTPA ⁻ (M)	Cd^{2+} (M)	pH ± 0.001	Break-up percentage $\pm 5\%$
8.83×10^{-4}	1.6×10^{-5}	2.307	0
9.63×10^{-4}	3.6×10^{-5}	2.100	0
1.29×10^{-3}	6.6×10^{-5}	2.100	0
1.29×10^{-3}	2.6×10^{-5}	1.667	8.3
1.62×10^{-3}	6.5×10^{-5}	1.193	39.5
9.33×10^{-4}	6.5×10^{-5}	1.009	40.1
2.18×10^{-3}	6.5×10^{-5}	1.012	40.1

Table 5. Percentage break-up results for the $U^{235}(IV)$ – frozen system.

$U^{235}(IV)$ DTPA (M)	Cd^{2+} (M)	pH	Break-up percentage $\pm 5\%$
9.32×10^{-4}	1.6×10^{-1}		
9.32×10^{-4}	1.6×10^{-5}	1.011	56.6
9.32×10^{-4}	1.6×10^{-5}	1.009	56.6

to emerge: (1) The maximum break-up percentage has been observed to be 40% only. (2) The percentage bond rupture is pH-dependent. (3) A different pH range has been observed for each of the complex system studied. (4) Reducing agents do not seem to affect the break-up result. (5) Freezing of the reaction mixture, produced the bond rupture of higher order. These points are to be

Table 6. Percentage break-up results for the $U^{235}(IV)$ – DTPA system when $10^{-2}M$ KI was added to the reaction mixture.

$U^{235}(IV) DTPA^{-}$ (M)	Cd^{2+} (M)	pH ± 0.001	Break-up percentage $\pm 5\%$
1.89×10^{-3}	1.6×10^{-5}	0.890	37.4
1.89×10^{-3}	1.6×10^{-5}	0.910	37.8

Table 7. Percentage break-up results for the $U^{235}(IV)$ – DTPA system when $U(IV)$ concentration was increased in the form of $U^{238}(IV) DTPA^{-}$.

$U(IV) DTPA^{-}$ (M)	Cd^{2+} (M)	pH ± 0.001	Break-up percentage $\pm 5\%$
7.88×10^{-3}	3.2×10^{-5}	1.011	42.5
7.88×10^{-3}	3.2×10^{-5}	1.011	42.5

interpreted in view of the following.

The important modes of excitation and ionisation, following alpha decay of U^{235} to Th^{231} are as follows:

(1) The production of positive charge on Th^{231} daughter atoms following: (i) shake off-of the electrons, and (ii) internal conversion of the gamma rays. (2) Recoil energies due to the emission of an alpha particle.

Recoil energies, calculated on the basis of the data available in the Nuclear Data Sheets [22] and Table of Isotopes [23] are shown in Table 9 and Table 8 shows the relevant information about gamma rays from the same Nuclear Data Sheet [22]. From Table 9 it is evident that the recoil energies associated with the various alpha particles emitted by U^{235} , are of high values, varying between 73.00 – 79.54 keV. Recoil energies from gamma rays and conversion electrons will be too low to be considered here. The recoil energies can be used to calculate the ranges of alpha decay daughter atoms, by using Bohr's equations [27]. Thus, for the daughter Th^{231} atom recoiling with an energy of 76.12 keV, the range will be 27.1 nm, this distance would be considered to be sufficiently large enough to allow the daughter Th^{231} atoms to escape from the sphere of influence of the fragmented ligand and the initial solvent cage. Hence, every daughter Th^{231} atom produced following alpha decay of U^{235} is expected to break the molecular bonds and come to rest in solution as a free metal ion.

In addition, attention is to be given to the charge pro-

Table 8. The energies, abundances and total conversion coefficients of the principal gamma-rays found in U^{235} decay.

Gamma-ray energy (meV)	Internal conversion coefficient ($\times 10^2$)	Abundance (%)	Converted (%)	Life time of the excited state
0.201	99.2	3	3.0	
0.180	95.5	2	1.9	
0.204	95.0	5.5	5.2	10^{-15}
0.163	95.0	5.8	5.5	sec
0.109	97.5	2.7	2.1	
0.185	46	60	27.6	0.77×10^{-9}
0.143	89	13	12.6	sec

Table 9. Recoil energies of the Th^{231} daughter atoms.

Alpha particle abundance (%)	Alpha particle energy (meV)	Recoil energy (keV)
4.6	4.597	79.54
3.7	4.556	78.86
1.2	4.502	78.79
4	4.415	76.19
57	4.396	76.12
18	4.366	75.58
3	4.323	74.84
5.7	4.216	73.00

duction processes in U^{235} alpha decay. The charge spectra of Th^{231} atoms produced as a result of internal conversion of gamma rays following alpha decay of U^{235} has not been reported. However, due to the probability of internal conversion in about 57% of the alpha decay events (Table 8) it is expected that there will be an outburst of conversion electrons from the recoiling Th^{231} daughter atoms in two stages: (i) in 7% of the decay events, when prompt gamma rays are internally converted, the recoil of the daughter atoms and the emission of the electron will be simultaneous, and (ii) the outburst of the second group of electrons, comprising 40% of the decay events, will be in a situation where the daughter atoms have already come to rest.

The low break-up results observed in this work can only be understood, if it is assumed that recombination of the recoil daughter atoms takes place. The interacting ligand can not be its original partner in the complex because our calculations show that the recoil range, is sufficient enough to allow the daughter atom to escape from its initial solvent cage and the sphere of influence of the fragmented ligand.

Thus, the cage recombination proposition put forward by Cottin and Haissinsky [24] to explain the low break-up results in their investigation of U^{238} alpha decay in uranyl benzoyl acetate solution (recoil energy of daughter $Th^{234} = 71$ keV) must be reconsidered. The recoil ranges lead us to think that the recombination of daughter Th^{231} takes place with a free ligand ion at its place of rest. Let us see how these free ligands are originated. The recoiling Th^{231} atoms will come to rest in a time of the order of 10^{-13} sec and there will be a subsequent outburst of conversion electrons as described earlier. The daughter atoms will be highly ionised and radiolysis of adjacent solvent molecules will occur. The radiolysis of the solvent molecules will also be caused by the dissipation of the last few tens of electron volts of the daughter atoms kinetic energy in the immediate vicinity of its place of rest. A factor of major importance to be considered here is the presence in the reaction mixture of 10^{-3} to 10^{-4} molar concentration of U(IV) complex; U(IV) is a strong reducing agent and may neutralize the ionised daughter atoms and the U(IV) complex ions may additionally react with the radiolysis products of the interaction with the solvent. If either of these processes is carried through, the U(IV) complex ions will be oxidised and form free uranyl and ligand ions. These ligand ions will be then free to react with daughter Th^{231} atoms and thus reduce the amount of break-up which may be observed. In order to explain the pH dependence of the break-up results it can be pointed out that the reducing effect of U(IV) [25-26] increases with the increase of pH, and accordingly pH dominates the break-up values, which decrease with the increase of pH. At the pH where the bond rupture completely disappears, it is likely that the oxidation of U(IV) complex takes place to an extent that it produces enough free ligands for the complete recombination of the free daughter atoms.

The tendency that the daughter complex of higher thermodynamic stability shows break-up in higher acidic concentrations can be explained in terms of the protonation tendency of polyaminopolycarboxylic ligands, which blocks interaction of the ligand with metal ions. Therefore, an increase in the acidity of the solution will decrease the probability of recombination of metal and ligand, and at a particular acid concentration, the probability of the reformation of a complex will be more likely in a complex of higher thermodynamic stability. Thus, we observed complete recombination of Th EDTA ($\log K = 23.2$) at pH 4, of Th DCTA ($\log K = 24.9$) at pH 3 and that of Th DPTA ($\log K = 27$) even at pH 2. The addition of KI, a reducing agent, to the reaction mixture, or an increase of U(IV) concentration, approximately 7 times to its normal

concentration in the other experiments, in the U^{235} (IV) DTPA system, does not produce any decrease in the break-up percentage, perhaps signifying that the reduction to its limiting value has already been achieved (Tables 6 and 7). However, the break-up percentage increased to about 50% when the U^{235} (IV) DTPA decay was followed in the frozen solutions (Table 5). This might be due to the released electrons from the recoiling atom being trapped in the frozen solution and prevented from producing substantial radiolysis which probably is responsible for the recombination.

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