The extraction of actinides by diphenyl(dibutylcarbamoylmethyl)phosphine oxide in the absence of a solvent

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The possibility of changing the aggregate state of several solid mono- and bidentate neutral organophosphorus compounds in contact with various mineral acids was demonstrated. The formation of the liquid phase immiscible with water was observed. The phase exhibits a high extraction capability with respect to actinides in various oxidation states. The diphenyl(dibutylcarbamoylmethyl)phosphine oxide—HNO_3 extraction system was studied in detail. The composition of the liquid phase was shown to be $Ph_2P(O)CH_2C(O)NBu_2 \cdot HNO_3 \cdot nH_2O$ (n = 2 or 3). The kinetics of formation of this phase and extraction of Am^{III} from solutions of nitric acid and its salts was studied. The organic phase exhibits redox properties, due to which some actinides change the oxidation state during extraction. The "aryl strengthening effect" was suggested to exist for extraction of Am^{III} in the absence of a solvent.

Key words: actinides, americium(III), extraction in the absence of a solvent; bidentate neutral organophosphorus compounds, diphenyl(dibutylcarbamoylmethyl)phosphine oxide.

The structure and reactivity of bidentate neutral organophosphorus compounds (BNOPC) have long ago attracted the attention of researchers.[1] Interest in these compounds as extractants for actinides from acidic media was caused by the necessity to remove the latter from highly-active waste of the radiochemical industry. At the first stages, researchers from USA gave principal attention to BNOPC alkyl-substituted at the phosphorus atom, whereas in our country, works were oriented toward the synthesis and use of aryl-substituted BNOPC. This was due to the discovery of M. I. Kabachnik and co-workers of the special properties of organophosphorus compounds containing aryl groups at the P atom. For example, it has been shown[2] that alkaline metal halides with *P*-phenyl-substituted BNOPC of lower basicity form more stable complexes than with more basic alkyl-substituted BNOPC ("aryl strengthening effect," ASE). An increase in the extraction capability of BNOPC toward actinides on going from alkyl- to aryl-substituted reagents has been observed later.[3] Correlations between the structure of BNOPC (methylenediphosphine dioxides, polyoxides of polyphosphines with the complex structure of the bridge, and carbamoylmethylphosphine oxides) and their extraction capability and selectivity toward actinides as well as their physicochemical properties have been established.[4–8] It is shown, in particular, that BNOPC aryl-substituted at the phosphorus atom in all cases possess the highest extraction capability toward actinides due to ASE. The requirements to the structure, at which this effect is manifested to a maximum extent, were formulated.[6] The results obtained formed the basis for the directed synthesis of efficient extractants for extraction of actinides from highly-active waste.

Based on the σ,ρ-correlation analysis[9] and extraction data mentioned above, concepts explaining the reasons for ASE were formulated,[10] which are connected with the biphilic nature of phenyl (aryl) groups, substituents at the P atoms that are capable of manifesting acceptor properties in weak complexes with metals and donor properties in stable complexes, such as complexes of BNOPC with actinides. Authors[10] have drawn the basically important conclusion about the dependence of the electronic effect of the phenyl group on the nature of the solvent and have shown that the influence of a diluent on ASE is the solvation of complexes of a metal with BNOPC rather than the solvation of the reagents (*cf.* Ref. 11). Strongly solvating solvents, for example, *m*-nitrobenzotrifluoride ("fluoropol"),[12] stabilize a complex; the charge on the P atom increases, the phenyl groups become donors, and an additional strengthening of the complex appears.

The choice of the solvent is one of the key problems for the practical use of BNOPC because of stringent requirements to diluents in the technology. Therefore, the use of BNOPC in the absence of a solvent seems to be most promising. This idea appeared in connection with the ability of solid diphenyl(dibutylcarbamoylmethyl)phosphine oxide to form in contact with nitric

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acid the liquid phase, which is immiscible with water and possesses a high extraction capability with respect to actinides and lanthanides.[13,14]

The purpose of this work is to establish the composition of the liquid extractant and perform a systematic study of the influence of different factors on a change in the aggregate state of the organophosphorus reagent and its extraction capability with respect to actinides and lanthanides.

Experimental

Reagents. We used ^{237}Np, a mixture of isotopes ^{239}Pu and ^{238}Pu with an average specific activity $2.35 \cdot 10^6$ Bq mg^{-1}, and a mixture of ^{243}Am and ^{241}Am with an average specific activity $9.07 \cdot 10^6$ Bq mg^{-1}. Solutions of actinides were prepared in HNO_3 (ultrapure grade), and solutions of americium were also prepared in H_2SO_4 (ultrapure grade), $HClO_4$, and H_3PO_4 (reagent grade). The procedures described previously[8] were used for the preparation of nitric-acid solutions of Np^{IV}, Np^{V}, Pu^{III}, Pu^{IV}, Pu^{VI}, Am^{V}, and Am^{VI}.

BNOPC reagents were synthesized in the Laboratory of Organophosphorus Complex-Forming Agents in the A. N. Nesmeyanov Institute of Organoelement Compounds of the Russian Academy of Sciences, according to the known procedure.[15] Trioctylphosphine oxide (TOPO) and triphenylphosphine oxide (TPPO) (Chemapol) were used. 1,2-Dichloroethane was dried over $CaCl_2$ and distilled.

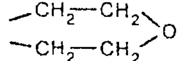
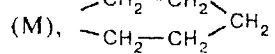
Extraction procedure. A weighed sample of the reagent and a solution of the element in the corresponding acid were added to a tube with the ground-glass stopper. The mixture was stirred during the time necessary for the transition of the reagent to the liquid state and establishment of extraction equilibrium (1—25 min depending on the experimental conditions).

Measurements. In the work with trace amounts of radionuclides, after extraction, separation of phases, and centrifugation an aliquot was taken from an aqueous phase for measuring radioactivity. The content of the element in the organic phase (extraction %) was calculated from the difference between the initial radioactivity and radioactivity of the aqueous phase after extraction. For measurement of the overall α-activity, a flow-type Protoka methane counter was used. α-Spectra were measured on a multi-channel LP-4900 pulse analyzer. When weighed amounts of elements were used, the oxidation state of actinides and their concentrations in different oxidation states were determined by spectrophotometry on a Shimadzu instrument.

The concentration of the acid in the solution was measured by pycnometry and titration with an alkali. The density of the organic phase for large volumes was determined by pycnometry. The volumes of small quantities of the organic phase (0.01—0.10 mL) were calculated from its density and determination of the weight.

Results and Discussion

Study of the possibility to change the aggregate state of different mono- and bidentate phosphine oxides on contact with mineral acids

We studied a series of diphenyl(dialkylcarbamoylmethyl)phosphine oxides with various substituents at nitrogen atoms with the general formula $R_2'P(O)CH_2C(O)NR_2$, where R = Ph and R′ = Bu, octyl (Oct),  (M),  (Pp), as well as R = Bu and R′ = Et. For simplicity, these reagents will be designated hereinafter as Ph_2Bu_2, Ph_2Oct_2, Ph_2M, Ph_2Pp, and Bu_2Et_2, respectively. Octylphenyl(diisobutylcarbamoylmethyl)phosphine oxide $OctPhP(O)CH_2(CO)NBu^i_2$ (designated hereinafter as $OctPhBu^i_2$), tetraphenylmethylenediphosphine dioxide $Ph_2P(O)CH_2P(O)Ph_2$ (DTPMDP) and monodentate reagents trioctylphosphine oxide (TOPO) and triphenylphosphine oxide (TPPO) were also used. The quantities of the solid reagents used were $1 \cdot 10^{-4}$ moles. On contact of Ph_2Bu_2, Ph_2Oct_2, $OctPhBu^i_2$, and DTPMDP with a 3 *M* solution (1 mL) of HNO_3, a liquid phase is formed within 3 min, which extracts quantitatively americium. TOPO and TPPO also form a liquid phase, which either extracts Am^{III} by ~16% (TOPO) or does not extract it (TPPO). Ph_2M and Ph_2Pp do not change the phase state even within 22 h.

A change in the aggregate state of the reagents occurs on contact not only with HNO_3, but also with other mineral acids; for example, Ph_2Bu_2 with 3 *M* H_2SO_4, H_3PO_4, or $HClO_4$ forms a liquid phase within 3 min, which, in the case of $HClO_4$, extracts Am^{III} rapidly and quantitatively, and in the case of H_2SO_4 and H_3PO_4, the extraction is very slow.

For the further detailed study of the factors affecting a change in the phase state of the reagent, the composition of the liquid phase formed, and its extraction properties, we used the Ph_2Bu_2—HNO_3 system.

Effect of the concentration of HNO_3 and duration of contact of the phases on the change in the aggregate state of the Ph_2Bu_2 reagent

We found that the concentration of HNO_3 and duration of contact of the phases are the determining and interrelating factors of the transition of Ph_2Bu_2 from the solid state to the liquid (gel-like) state. At low concentrations of the acid (0.1—0.2 mol L^{-1}), the reagent ($1 \cdot 10^{-4}$ moles in 2 mL of the solution) remains as a porous powder-like mass independently of the duration of stirring up to 20 h. In a 1 *M* solution of HNO_3 within 3 min, the reagent is transformed into an irregular lump as if covered with an oily film, and its appearance remains unchanged when the duration of stirring increases. For the formation of the liquid phase in the form of a transparent gel-like droplet, 3—5 min are enough if the concentration of HNO_3 exceeds 2.5—3.0 mol L^{-1}. The higher the concentration of the acid, the faster the change in the aggregate state, and no change is observed if the acid is replaced by a solution of $NaNO_3$ with the same concentration.

Reaction of Ph_2Bu_2 with nitric acid

The dependence of the weight of dry Ph_2Bu_2 (m_1) on the weight of the liquid phase (m_{liq}) formed due to the contact between the reagent and acid is presented below.

m_l/g	m_{liq}/g	[HNO_3]/mol L^{-1}	V_{H_2O}/mL
0.9396	1.2056	4.70	8.0
0.6936	0.8796	4.18	2.0
0.5839	0.7398	4.70	4.0
0.4159	0.5340	4.18	2.0
0.2254	0.3263	4.18	2.0
0.1050	0.1290	4.70	2.0

(V_{H_2O} is the volume of a solution of the acid added to the reagent.)

The transition of Ph_2Bu_2 to the liquid state is accompanied by an increase in the weight of the reagent. The coefficient that takes into account this increase is equal to 1.266±0.015; the density of the liquid phase is 1.163±0.012 g cm^{-1}.

An increase in the weight of Ph_2Bu_2 on going to the liquid state indicates that a compound of this reagent with HNO_3 is formed. The measurements of the volumes of the aqueous phase and concentration of the acid in the aqueous phase before and after extraction show that the complex formed has the composition $Ph_2Bu_2 \cdot HNO_3 \cdot nH_2O$ (n = 2 or 3).

Study of the kinetics of extraction of americium(III) with the Ph_2Bu_2 reagent

The equilibrium upon extraction of Am^{III} with the liquid phase ("droplet") preliminarily prepared by contact of a weighed sample of the reagent with 3 M HNO_3 is achieved more rapidly than that upon the simultaneous addition of the reagent and Am^{III} to a solution of the acid, as described in Experimental (Fig. 1, *a*), and, hence, the kinetics of extraction of Am^{III} is described by the kinetics of formation of a compound of the reagent with HNO_3. In the experiments described hereinafter, extraction was performed by the second method (except especially emphasized cases).

The dependence of the kinetics of extraction of Am^{III} on the amount of the reagent (Fig. 1, *b*) indicates that the greater the weight of the reagent, the faster the transition of americium to the organic phase.

The greater the volume of the initial aqueous phase, the slower the establishment of the equilibrium (Fig. 2, *a*). Most likely, the kinetics of extraction of americium plays the determining role in the given case rather than the kinetics of formation of a complex of the reagent with HNO_3. The close resemblance of the dependences of the kinetics of extraction of Am^{III} (Fig. 2, *b*) and kinetics of formation of the liquid phase on the acid concentration indicates that the kinetics of extraction of Am^{III} is determined by the rate of formation of the liquid phase of the reagent.

Study of extraction of actinides with the Ph_2Bu_2 reagent from solutions of nitric acid

The dependence of extraction of trace amounts of Am^{III} with the Ph_2Bu_2 reagent in the absence of a

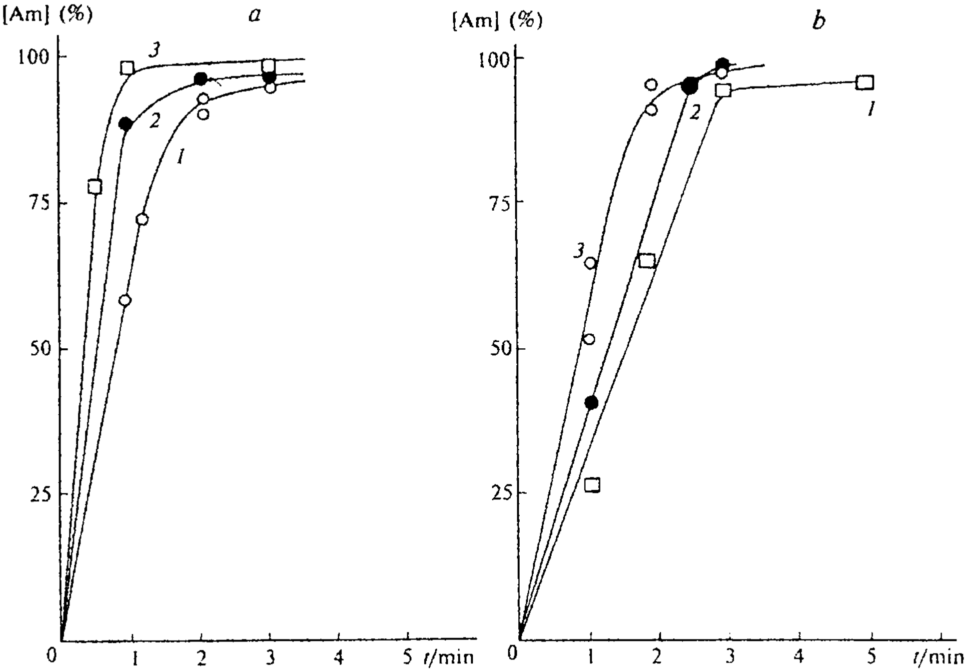


Fig. 1. *a*. Dependences of extraction of Am^{III} from a 1 M solution of HNO_3 with Ph_2Bu_2 in the absence of a solvent on the duration of contact between the phases with the simultaneous addition of a weighed sample of Ph_2Bu_2 ($1 \cdot 10^{-4}$ mol) to a solution of Am^{III} (*1*) and for extraction with a prepared "droplet" (*2*, *3*) formed within 3 (*2*) and 10 min (*3*). *b*. Dependence of extraction of Am^{III} from a 3 M solution (1 mL) of HNO_3 with Ph_2Bu_2 in the amount: $2.5 \cdot 10^{-5}$ (*1*), $5 \cdot 10^{-5}$ (*2*), and $1 \cdot 10^{-4}$ mol (*3*) on the duration of contact between the phases.

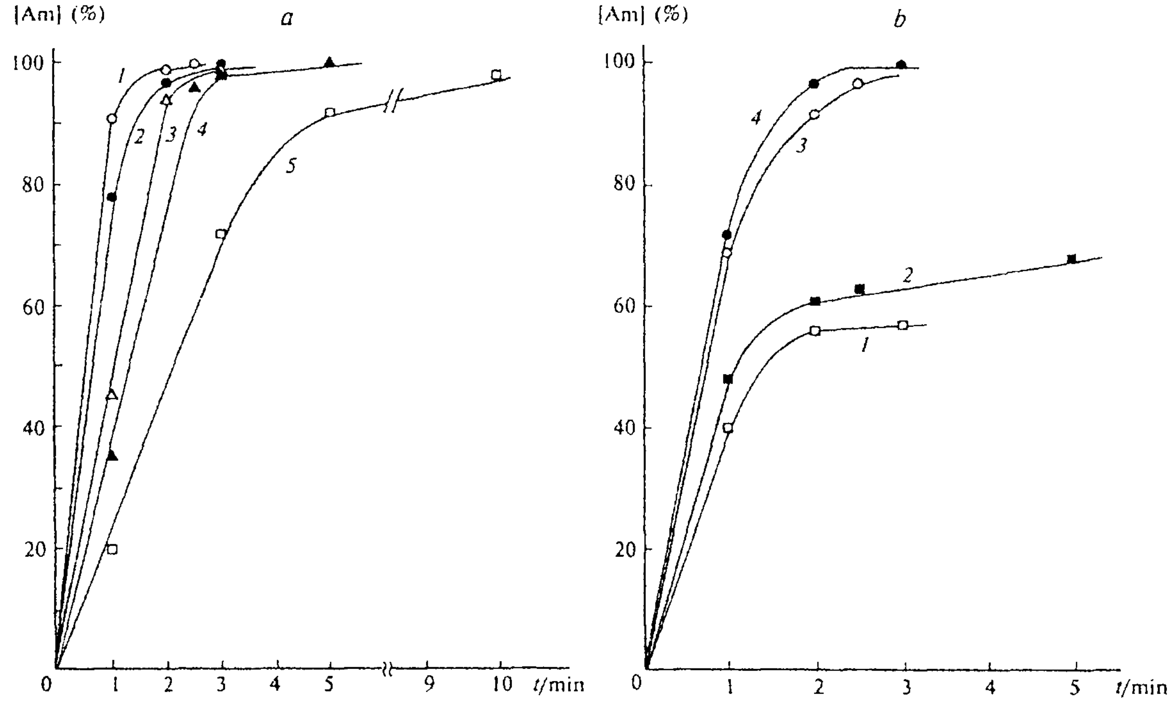


Fig. 2. *a*. Dependence of extraction of Am^{III} from 0.25 (*1*), 0.5 (*2*), 1 (*3*), 3 (*4*), and 5 mL (*5*) of a 3 *M* solution of HNO_3 with Ph_2Bu_2 ($5 \cdot 10^{-5}$ mol) on the duration of contact between the phases. *b*. Dependence of extraction of Am^{III} from 0.1 *M* (*1*), 0.2 *M* (*2*), 3 *M* (*3*), and 5 *M* (*4*) solutions (1 mL) of HNO_3 with Ph_2Bu_2 ($5 \cdot 10^{-5}$ mol) on the duration of contact between the phases.

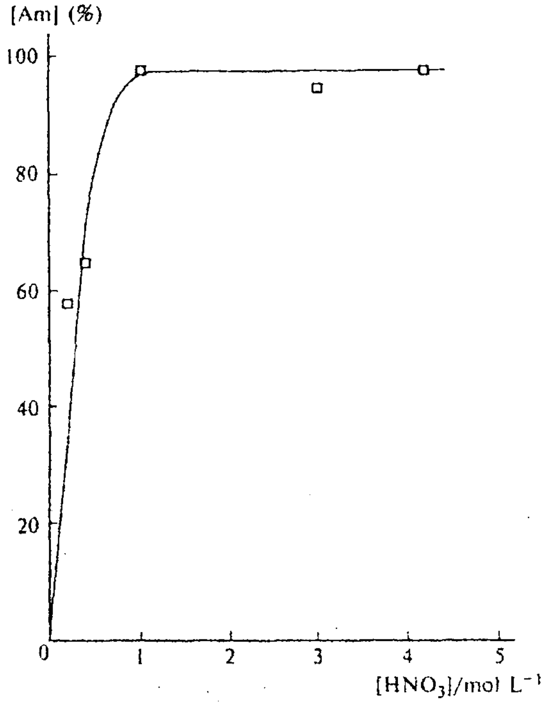


Fig. 3. Dependence of extraction of Am^{III} with Ph_2Bu_2 ($1 \cdot 10^{-4}$ mol) on the concentration of HNO_3; duration of contact between the phases is 10 min.

diluent on the concentration of HNO_3 under equilibrium conditions (duration of stirring the phases was 10 min) is presented in Fig. 3. In the systems containing a diluent, weak extraction from solutions of HNO_3 with the concentration <0.5 mol L^{-1} is explained[4] by the low concentration of neutral complexes of Am^{III} being extracted, while in the given case, an insufficient amount of the liquid extractant also plays a certain role. As shown above, the reagent virtually does not change its initial form on contact with 0.1 *M* HNO_3, *i.e.*, it is only partially transformed into the liquid state due to a deficiency of the acid.

Americium(III) can be quantitatively extracted from a 0.1 *M* solution of HNO_3 with the same amount of the reagent prepared beforehand by contact of the weighed sample with a 3—7 *M* solution of HNO_3. The "droplet" thus prepared extracts quantitatively americium from a 0.1 *M* solution (1 mL) of HNO_3 within 3 min.

To establish the existence of ASE for extraction in the absence of a diluent, we compared the extraction properties of carbamoylphosphine oxides (Ph_2Bu_2 and Bu_2Et_2) aryl- and alkyl-substituted at the phosphorus atom (Fig. 4). The presence of different substituents at N atoms plays no role, since they have almost no effect on the change in the extraction properties of carbamoylmethylphosphine oxides.[8] It follows from Fig. 4 that Ph_2Bu_2 is much better than the Bu_2Et_2 reagent in extraction capability toward americium. Thus, we have

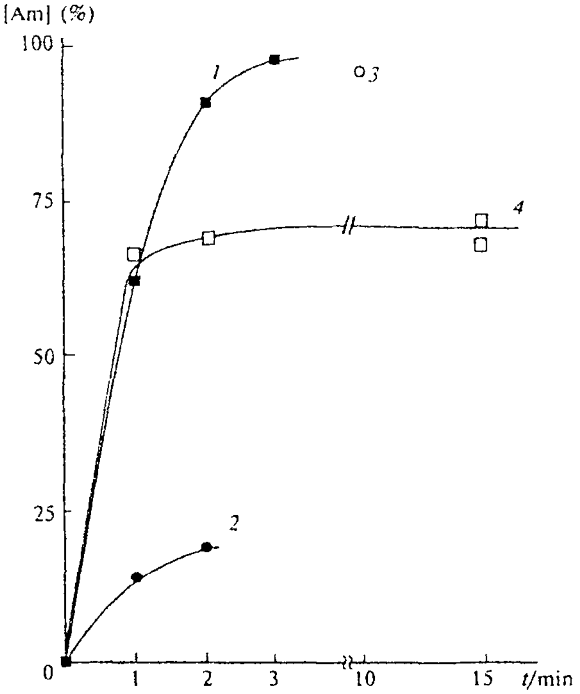


Fig. 4. Dependence of extraction of Am^{III} from a 3 *M* solution (1 mL) of HNO_3 with Ph_2Bu_2 (*1*, *3*) and Bu_2Et_2 (*2*, *4*) reagents at the concentrations of the reagents: $1 \cdot 10^{-4}$ (*1*, *2*) and $3 \cdot 10^{-4}$ mol (*3*, *4*) on the duration of contact between the phases.

established for the first time that the observed ASE also exists in systems without a diluent, although its has previously been assumed[11] that the nature of the diluent is the main reason for the "aryl effect." The existence of ASE can be verified only after the determination of the extraction constants of Am^{III} with these reagents.

The results of studying extraction of actinides in different, mainly unstable, oxidation states with the Ph_2Bu_2 reagent in the absence of a diluent from 4 *M* solutions of HNO_3 (duration of contact between the phases was 3 min) are presented in Table 1.

As follows from the data in Table 1, extraction of actinides depending on their oxidation state changes in the following series: IV > VI > III > V. Concentrating by 60—80 times is achieved by extraction. When the reagent is transferred to the phase, Np^{IV} and Np^{VI}, like Pu^{IV} and Pu^{VI}, do not change oxidation states. When the extract is dissolved in 1,2-dichloroethane, the absorption spectra exhibit a slight shift of maxima of the bands and an increase in their intensity as compared to similar spectra in nitric-acid solutions, which indicates the formation of stable bonds between the actinide and reagent.

Table 1. Extraction of actinides in different oxidation states from a 4 *M* solution of HNO_3 with Ph_2Bu_2

Element	$C \cdot 10^3$ /mol L^{-1}	V_{H_2O} /mL	$m_1 \cdot 10^5$ /mol	V_{org} /mL	Extraction (%)
Np^{IV}	1.50	1.2	4.4	0.018	99.6
Np^{V}	1.47	1.2	3.6	0.015	43.0
Np^{VI}	1.72	1.2	4.4	0.018	98.8
Pu^{III}	3.01	1.5	8.1	0.033	98.5
Pu^{IV}	3.30	1.5	8.1	0.033	99.7
Pu^{VI}	3.59	1.5	8.1	0.033	97.1
Am^{III}	0.81	1.2	8.1	0.033	98.2
Am^{V}	1.55	1.2	8.1	0.033	54.0
U^{VI}	1.50	10.0	10.0	0.050	98.8

Note. *C* is the concentration of the element in the solution; V_{H_2O} is the volume of the aqueous phase; m_1 is the amount of the reagent; and V_{org} is the volume of the organic phase.

Only Am^{III} and Am^{V} were found in the aqueous and organic phases when Am^{VI} was extracted. Extraction of Pu^{III} is accompanied by a change in its oxidation state, which is indicated by the change in color of the organic phase in time from blue-violet to yellow-brown. Only bands typical of Pu^{IV} were observed in the absorption spectrum of the organic phase dissolved in 1,2-dichloroethane.

Extraction of Np^{V} is also accompanied by a change in its valence state on going to the reagent phase. The distribution coefficient of Np is low, which indicates that it goes to the organic phase in the pentavalent state followed by the oxidation in the reagent phase to the hexavalent state (Np^{VI}).

Changes in the valence states of metals during their extraction suggest that the reagent possesses redox properties. Based on the values of formal redox potentials of the Pu^{IV}/Pu^{III}, Np^{VI}/Np^{V}, and Am^{VI}/Am^{V} pairs equal to +0.982, +1.37, and +1.60 V, respectively,[16] we can approximately estimate the redox potential of the reagent as 1.5 V.

Extraction of Am^{III} from solutions of salts

In radiochemical practice, americium is often extracted from solutions with a complex salt composition. Therefore, it was necessary to elucidate the effect of salts on the extraction degree of Am with the Ph_2Bu_2 reagent in the absence of a diluent. For this purpose, we studied extraction of Am^{III} with the Ph_2Bu_2 reagent ($5 \cdot 10^{-5}$ mol) from solutions (1 mL) with different concentrations of HNO_3 and its salts. The content of salts (with respect to cation) was the following (g L^{-1}): Al, 5—15; Na, 5—20; Ca, 0.5—2.0; Cr, 0.5—2.5; and Fe, 0.5—5.0.

As shown above, at a fixed volume of the solution and weighed sample of the reagent, the completeness of extraction of americium is determined by the amount of the compound of Ph_2Bu_2 with HNO_3 which constitutes the organic phase and depends on the concentration of the acid and duration of contact of the phases. In extraction systems containing a diluent, salting out of the element and acid to the organic phase are competing processes, whereas in the absence of a diluent, both processes favor an increase in extraction of americium. It is most likely that salting out of the acid results in a more complete formation of a compound of the reagent with acid (liquid organic phase) and, hence, in an increase in extraction of americium.

It is seen from the extraction of Am^{III} from solutions of aluminum salts that when the concentrations of the acid and salt increase, the degree of extraction of Am^{III} increases, and the time of establishment of extraction equilibrium decreases (Fig. 5). The effect of the aluminum concentration on the completeness of formation of the organic phase can be observed visually for extraction from a 0.1 *M* solution of HNO_3. As the aluminum salt is added and its concentration increases, the powder-like reagent, which does not change its form upon prolonged contact with a 0.1 *M* solution of HNO_3 in the absence of the aluminum salt, is transformed into a porous oily irregular lump, *i.e.*, the transition (although not complete) from the solid state to the liquid state occurs, which is observed during a gradual increase in the acid concentration.

A similar picture is observed for extraction of Am^{III} from solutions of Na (0.1—3 *M* HNO_3), Ca, Co, and Ni (1—3 *M* HNO_3) salts. These elements behave as typical salting out agents, although last two are noticeably transferred to the organic phase, which gains a more intense blue or green color, respectively, as the salt

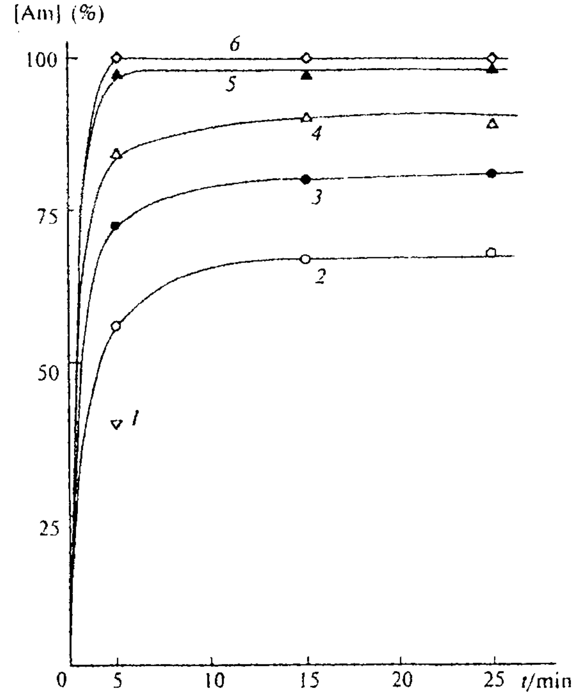


Fig. 5. Dependence of extraction of Am^{III} with Ph_2Bu_2 ($5 \cdot 10^{-5}$ mol) from 0.1 *M* (*1—4*); 1 *M* (*5*), and 3 *M* (*6*) solutions (1 mL) of HNO_3 containing aluminum in the concentration: 0 (*1*), 5 (*2*), 10 (*3*), and 15 g L^{-1} (*4—6*) on the duration of contact between the phases.

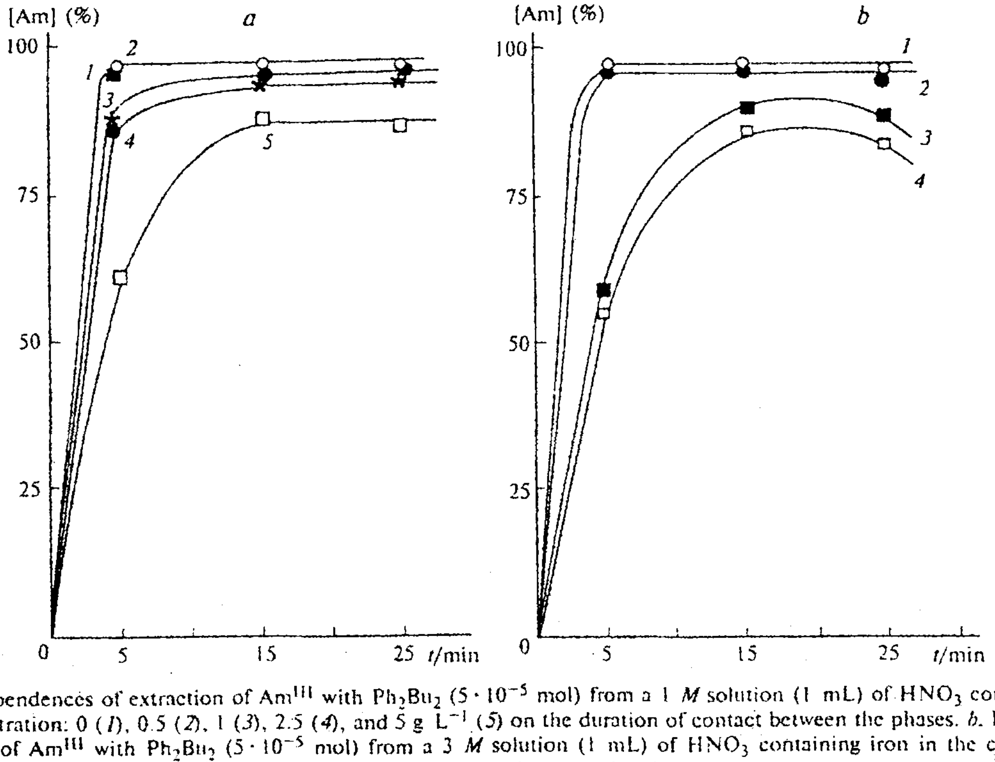


Fig. 6. *a*. Dependences of extraction of Am^{III} with Ph_2Bu_2 ($5 \cdot 10^{-5}$ mol) from a 1 *M* solution (1 mL) of HNO_3 containing iron in the concentration: 0 (*1*), 0.5 (*2*), 1 (*3*), 2.5 (*4*), and 5 g L^{-1} (*5*) on the duration of contact between the phases. *b*. Dependences of extraction of Am^{III} with Ph_2Bu_2 ($5 \cdot 10^{-5}$ mol) from a 3 *M* solution (1 mL) of HNO_3 containing iron in the concentration 0.5 (*1*), 1 (*2*), 2.5 (*3*), and 5 g L^{-1} (*4*) on the duration of contact between the phases.

concentration increases. However, this does not prevent extraction of americium.

Thus, the presence of salts of weakly extracting elements (Al, Ca, Na, Co, and Ni) in sufficiently high concentrations favors an increase in extraction of Am^{III} from a 0.1 *M* solution of HNO_3 and does not prevent extraction from 1—3 *M* solutions of HNO_3.

A somewhat different situation is observed for extraction of Am^{III} from solutions of Cr^{III} and Fe^{III} salts. According to the data on extraction of Fe^{III} with solutions of carbamoylmethylphosphine oxides in 1,2-dichloroethane from nitric-acid solutions,[17,18] Fe^{III}, unlike the elements mentioned above, is extracted with distribution coefficients (D_{Fe}) comparable to D_{Am}. The D_{Fe} value increases sharply as the acid concentration increases, whereas D_{Am} decreases as the concentrations of the acid and iron increase (Fig. 6). A slight tendency of the D_{Am} value to decrease is also observed upon prolonged contact of the phases in the system with a 3 *M* solution of HNO_3 and low concentrations of iron. The results obtained agree with the concepts[17,18] on the behavior of Fe^{III} in the extraction systems with carbamoylmethylphosphine oxides: an increase in the concentrations of the acid and iron and an increase in the duration of contact between the phases favor the accumulation of iron in the organic phase and, hence, the suppression of extraction of americium. A similar situation is observed for extraction of Cr^{III}. It is extracted into the organic phase, which is indicated by the appearance of the blue color.

Thus, in the presence of salts of noticeably extracted elements (Fe^{III}, Cr^{III}), extraction of Am^{III} decreases as their content increases. Under the conditions described above, iron in concentrations to 1 g L^{-1} in 1 *M* and 3 *M* solutions of HNO_3 does not prevent the quantitative extraction of Am^{III}.

Thus, the liquid complex of the Ph_2Bu_2 reagent with HNO_3 is an efficient extractant with respect to actinides in different oxidation states, which allows actinides to be extracted from solutions of acids and salts in a wide concentration range. This extractant does not require the use of toxic diluents and possesses a very high efficiency, which allows actinides to be concentrated. The method of extraction in the absence of a diluent is especially promising for fast and efficient concentration of actinides from dilute solutions before their determination.

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