FINAL REPORT

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"Actinide-aluminate speciation in alkaline radioactive waste"

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3. Executive Summary

Investigation of behavior of actinides in alkaline media containing Al(III) showed that no aluminate complexes of actinides in oxidation states (III-VII) were formed in alkaline solutions. At alkaline precipitation (pH 10-14) of actinides in presence of Al(III) formation of aluminate compounds is not observed. However, in precipitates contained actinides(IV),(VI), and to a lesser degree actinides(III), some interference of components takes place that is reflected in change of solid phase properties in comparison with pure components or their mechanical mixture. The interference decreases with rise of precipitation pH and at pH 14 is exhibited very feebly. In the case of Np(VII) the individual compound with Al(III) is obtained, however it is not aluminate of neptunium(VII), but neptunate of aluminium(III) similar to neptunates of other metals obtained earlier.

The obtained results prove absence of interaction of aluminate-ions with actinides ions in alkaline solution and there are not evident possibility of formation of solid aluminates of actinides in various oxidation states under investigated conditions. However, the presence of interaction of components in precipitates has caused the second part of the work - clearing up of the reasons and nature of the indicated interaction by examination of simultaneous hydrolysis An(III), (IV), (V), (VI) with Al(III).

The new An(VI),(IV) species have been found - the mixed hydroxocomplexes with Al(III). It is shown, that the yield of such complexes in solutions at the first stages of An(VI) hydrolysis (An=U, Np, Pu) can reach tens percents at [Al(III)] $\sim 10^{-2}$ M and pH about 4. The composition of stoichiometric complexes formed at first stages of hydrolysis has been determined and the stability constants of hydroxocomplexes have been measured. Analogous complexes have been found in the case of An(IV) (ðÍ~2). The tend of actinides in different oxidation states to formation of mixed hydroxocomplexes increases in a series VI > IV > III >> V, and in case of An(V) (namely, Np (V)), formation of mixed hydroxocomplexes has not been revealed. Heptavalent actinides were not explored in view of their instability in feebly acidic solutions.

The co-polymerization of An(VI) or An(IV) with Al(III) takes place at pH rising, terminating by formation of a mixed precipitate. The tend to co-polymerization during hydrolysis of solutions containing ions An and Al(III) determines degree of interference of components (actinide - aluminium) in a precipitate.

Third part of the work concerns an opportunity of formation of compounds of actinides with some other Al-containing ions. Radioactive wastes contain various elements, that gives an opportunity of formation not only aluminate-ions, but also of other complex ions containing aluminium. Among them, for example, AlF_6^{3-} and heteropolyanion of composition $[Al(OH)_6Mo_6O_{18}]^{3-}$.

Complex formation of trivalent actinides and lanthanides with $[Al(OH)_6Mo_6O_{18}]^{3-}$ is extremely weak and it can not influence the actinide species in the solution under conditions of storage of the wastes. However, the conditions have been found of separation of the f-elements solid compounds with such heteropolyanion.

The general conclusion can be drown that the Al(III) influence on the actinides species in waste tanks can be noticeable only at pH 10-11. At pH=4-5 it is stipulated by an opportunity of formation of mixed hydroxocomplexes An(VI), (IV). At higher pH the precipitates containing both actinide and Al forms which have some properties different from the properties of mechanical intermixture of aluminium and actinide hydroxides. The formation of mixed hydroxocomplexes and interference of components in precipitates fall down in the series An(VI) > An(IV) > An(III) >> An(V). At pH 13-14 interference of Al with actinides practically disappears. The formation of aluminate compounds both in a solution, and in a solid phase is represented to be extremely improbable at any conditions. Influence of heteropolyanions containing Al, on the actinides species in tanks is negligible, though the solid complexes of trivalent f-elements have been separated.

4. Research Objectives.

Aluminate compounds of actinides are known but all of these compounds were prepared by solid state chemistry method at high temperature [1]. There are not evident proofs

that interaction between aluminate-ion and actinides in different oxidation states takes place in the solutions. It was observed [2] that solubility of Pu(IV) in alkaline solutions slightly increases in the presence of aluminate. Complexation or ionic strength influence could explain this observation. The behavior of actinides in the presence of aluminate-ion is of interest due to the problems of high level radioactive wastes (HLW) treatment which contain considerable amounts of aluminum. The composition of different tanks with stored HLW [3] gives the possibility to suppose the formation not only the hydroxo-aluminate like $Al(OH)_4^+$ in wastes, but also some complex aluminates such as AlF_6^{3-}, $[Al(OH)_6Mo_6O_{18}]^{3-}$, etc. Main part of transuranium elements (TRU) in wastes is incorporated in solid phase in the form of hydroxides especially as mixed hydroxides with different metal hydroxides. In mixed hydroxides the formation of "ol"-bridges: An-O-Me (An – actinide, Me – metal ion) is postulated [4]. In the case of Me=Al in some approach the mixed hydroxide could be considered as a model of interaction between An and aluminate-ion because the oxygen surrounding of aluminum atoms in hydroxide might be close to coordination sphere of aluminum in aluminate.

Thus we have tried to clarify some features of behavior of actinide with $Al(OH)_4^+$, AlF_6^{3+}, $[Al(OH)_6Mo_6O_{18}]^{3-}$ - ions and mixed actinide-aluminum hydroxides.

5. Method and Results

Method: Experimental methods, preparation and characterization of stock solutions of actinides in different oxidation states, equipment and apparatus used in this work are described in detail recently [4,5].

Results:

1. Search for interaction between aluminate and actinide ions.

The spectra of actinide ions in solutions as a rule are very sensitive towards change of near coordination sphere. This permits to investigate many chemical processes with actinide and especially their complexation. The aluminate-ion and actinide ions, which exhibit noticeable solubility in alkaline media, represent the hydroxo-complexes. It is doubtful to assume the possibility of the complex formation between such particles in

alkaline solutions. Nevertheless we checked-out the influence of aluminate on the spectra of Np(VII), Pu(VI) and Np(V) in strong alkali.

The successive addition of ~0.3 M aluminate in 2 M NaOH prepared by dissolution of $Al(OH)_3$ in NaOH solution to $(7-9) \cdot 10^{-3}$ M Np(VII) or Pu(VI) in 2 M NaOH leads only to decreasing of intensities due to dilution. $5 \cdot 10^{-3}$ M Np(V) was prepared by addition of 0.13 M NpO_2NO_3 to 2.5 M $(C_2H_5)_4NOH$, and 0.15 M aluminate was prepared by addition 2.3 M $Al(NO_3)_3$ to 2.5 M $(C_2H_5)_4NOH$. Mixing of these solutions does not change the character of Np(V) spectrum.

Some information about the interaction of different ionic species could be concluded from coprecipitation experiments. We investigated the coprecipitation of Pu(VI) with moderate soluble lithium aluminate. The solutions for coprecipitation experiments were prepared by mixing of $(5-8) \cdot 10^{-3}$ M Pu(VI) in 2 M NaOH and aluminate solution obtained by addition of 2.3 M $Al(NO_3)_3$ to concentrated NaOH solution. Final composition of solution was $(3-4) \cdot 10^{-3}$ M Pu(VI) and 0.125-0.3 aluminate in 2 M NaOH. The 2 M Li_2SO_4 solution were added up to molar ratio Li:Al = 1 or 2. The precipitation of lithium aluminate takes place in some minutes. The IR-spectra and X-ray diffraction patterns of obtained precipitates have shown that Li_2CO_3 did not contaminate the solids. The precipitates were washed by 2 M NaOH solution then dissolved in nitric acid and quantity of Pu was determined radiometrically. The content of Pu coprecipitated was about 1-10%. Obtained results we could explain by sorption and occlusion because there was not observed relation between Pu content in precipitate and of micro- to macro-component ratio.

It was found that phosphate- and silicate-ions are strongly sorbed by $PuO_2 \cdot xH_2O$ in alkaline solutions [6]. We have examined the possibility of aluminate sorption by $PuO_2 \cdot xH_2O$. Aluminate solution was prepared by dissolving of $Al(OH)_3$ in NaOH concentrated solution (~10 M) and added to $8 \cdot 10^{-3}$ M Pu(VI) in 2 M NaOH. Final concentration of Al was about 0.3 M, [Pu(VI)]=$5 \cdot 10^{-3}$ M in 2 M NaOH. To this solution 1 M $N_2H_4 \cdot HNO_3$ solution was introduced up to hydrazine content ~10^{-2} M. Reaction mixture was maintained at 80°C for 30 minutes. $PuO_2 \cdot xH_2O$ was precipitated completely.

The IR-spectra and X-ray diffraction patterns of obtained precipitates have shown that $PuO_2 \cdot xH_2O$ did not contain the aluminate.

Thus we have not any sign confirming even the possibility of interaction of aluminate-ion with actinide ions in alkaline media.

2. Interaction between aluminum and actinides during the hydrolysis of their hydroxo-anions.

During the acidification of alkaline solutions the aluminium hydroxo-complexes as well as actinide hydroxo-complexes such as $AnO_2(OH)_4^{2-}$; $AnO_4(OH)_2^{3-}$ etc., transform into hydroxides. There are no precise data in available literature on the hydrolytic behavior of actinides in different oxidation states in alkaline solutions enough to estimate the conditions where anionic forms of actinides would be stable and could react with cations giving the salts.

Among An(VII) compounds with triply charged cations, well-known $[Co(NH_3)_6]NpO_5$, and $[Pt(NH_3)_6]NpO_5$ complexes and unstable compounds with $[Co(en)_6]^{3+}$ and $[Cr(NH_3)_6]^{3+}$ cations were described [7]. All these complex cations are bulky and do not undergo the hydrolysis. On the contrary, triply charged aluminium cation is rather small and considerably undergoes hydrolysis. Nevertheless, we found that a green precipitate (product **1**) was formed at the slow addition of a 0.1-0.5 M aluminium nitrate solution to a Np(VII) ($\sim 10^{-2}$ M) solution in 0.4-0.5 M NaOH. The precipitation was complete at pH 10-11. The residual Np concentration in the solution was found to be lower than 10^{-5} M. The Al:Np molar ratio in the precipitate was close to 1:1. X-rays diffraction data showed the substance is amorphous. The IR spectrum is similar to the one for $Ba_3(NpO_5)_2$ given in [8]. The wide and intense band with a maximum at 730 cm^{-1} is related to the stretching vibrations of -Np-O-Np- chains. The intense bands at 900-1100 and 1250-1450 cm^{-1} result from the deformation vibrations of water molecules and OH groups. The band at 1550-1700 cm^{-1} is characteristic of the deformation vibrations of coordinated water. The intense band with a maximum at 540 cm^{-1} is due to the stretching vibrations of -Al-O-Np-bridges. The IR spectra of all compounds were measured in liquid paraffin because the grinding of Np(VII) compounds with KBr promotes the reduction of Np(VII) to Np(VII).

Therefore published earlier spectral data for Np(VII) compounds in KBr matrices may be incorrect. The electron absorption spectrum (EAS) of the freshly prepared precipitate was measured also in liquid paraffin deposited on quartz disk. The spectrum was identical to the one of Np(VII) anions in alkaline solutions (Figure 1) which exhibits characteristic maximums at 412 and 620 nm [9]. Therefore it is believed that in the obtained compound Np(VII) occurs as the NpO_5^{-3} anion as well as in alkaline solutions [5]. The thermal analysis showed the presence of a large amount of water (number of water molecules is 12) in the test samples. At the same time these samples did not contain detectable amounts of CO_2 . Thus we can conclude that the synthesized substance has a polymeric structure which contains large amounts of coordinated and interlayer water and OH groups. The chemical composition of the newly obtained compound can be described by the empirical formula $AlNpO_5 \cdot nH_2O$.

The initially green compound $AlNpO_5 \cdot nH_2O$ became black (product **2**) after storage for several hours either in a dry atmosphere free of CO_2, or in air at room temperature. The EAS and IR spectra and X-ray diffraction data confirmed the identity of both aged substances. This fact is also indicative of the absence of active carbonization of the products which is known for other Np(VII) anionic species. X-ray diffraction data also indicate that differently from initial $AlNpO_5 \cdot nH_2O$ the aged substance is crystalline, however structural parameters cannot be calculated from these data. Our attempts to obtain better-crystallized samples were unsuccessful. The EAS of product **2** is much smoother than the spectrum of $AlNpO_5 \cdot nH_2O$. The maximum at 412 nm (Figure 1) is shifted to a long-wave region of 420- 460 nm. This shift corresponds to the behavior of Np(VII) in solutions as the pH is decreased. If black product **2** was treated with a dilute alkaline solution the substance became green again and the EAS became similar to the spectrum of $AlNpO_5 \cdot nH_2O$ which is shown in Figure 1. Note that after storage for half a day the product **2** became irreversibly aged and the alkaline treatment of the substance did not result in any changes. This can be explained by the formation of a rigid structure as a result of the removal of interlayer water and a part of OH groups during the ageing, which prevents the easy interaction of hydroxyl ions with the aged compound. The X-ray

diffraction data for aged product **2** remained almost unchanged but the signals become more pronounced. Product **2** can also be obtained by the direct interaction of Al and Np(VII) in solution. For this purpose a Np(VII) solution (10^{-2} M) in 0.4-0.5 M NaOH was slowly added to a 0.5 M Al(NO)$_3$ solution to pH 5-6. The X-ray diffraction data confirm the identity of both of the products obtained by ageing of AlNpO$_5 \cdot$nH$_2$O in a dry atmosphere or in air and by the direct interaction of the components. The Al:Np molar ratio in product 2 was found to be close to 1:1. The NpO$_5{}^{3-}$ anion can be transformed into NpO$_4{}^-$ by decreasing pH of the solution [10]. Therefore the composition of product **2** can be described by the empirical formula AlO(NpO$_4$)$\cdot$nH$_2$O. The IR spectra of the compound retained the bands due to the vibrations of -Np-O-Np- chains. The bands at 900-1100 and 1250-1450 cm^{-1} became less pronounced in comparison with those in the spectrum of AlNpO$_5 \cdot$nH$_2$O. This can be explained by the dehydration of AlNpO$_5 \cdot$nH$_2$O in the course of ageing or by the synthesis at lower pH.
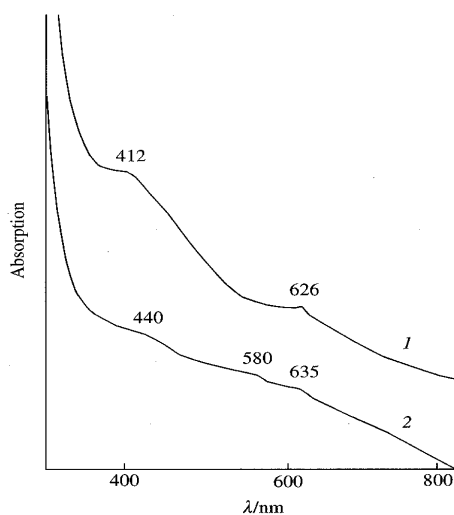

Figure1. EAS of AlNpO$_5 \cdot$nH$_2$O (**1**) and of AlO(NpO$_4$)$\cdot$nH$_2$O (**2**)

Coordinated water is clearly seen in the spectrum of AlO(NpO$_4$)$\cdot$nH$_2$O (a band at 1550-1700 cm^{-1}). It is interesting to note that an intense band at 540 cm^{-1} occurs in the spectrum of AlNpO$_5 \cdot$nH$_2$O, whereas the IR spectrum of LiAlO$_2$ exhibits a very intense band at 525 cm^{-1}. At the same time, such a band is absent in the IR spectrum of aluminum hydroxide. Therefore,

it is possible to assume that the oxygen environments of aluminum atoms in $AlNpO_5 \cdot nH_2O$ and $AlO(NpO_4) \cdot nH_2O$ are similar to the aluminate and hydroxide structures, respectively.

A compound of Np(VII) with Eu(III) was synthesized under the same conditions as $AlNpO_5 \cdot nH_2O$ was obtained. The IR spectrum of the compound exhibits a broad band at 500-900 cm^{-1}, which is a result of the superposition of the vibrations of Eu-O-Np bridges and Np-O-Np chains. The composition of this compound corresponds to the empirical formula $EuNpO_5 \cdot nH_2O$. It is interesting to note that the obtained compound was not transformed into a species similar to product **2**. Well-known compounds of the $M_3(NpO_5)_2 \cdot nH_2O$ type, where M is an alkaline earth metal, exhibit a similar behavior. It is likely that this fact can be explained by the much more rigid Eu-O and M-O bonds in the lattices in comparison with Al-O because of the large size of rare earth and alkaline earth metal atoms in comparison with the aluminum atom.

The compounds of Pu(VII) with Al(III) and Eu(III) were obtained under the same conditions as for $AlNpO_5 \cdot nH_2O$. However, the properties of these substances are difficult to study because of the instability of Pu(VII) resulting from a high redox potential of the Pu^{VII}/Pu^{VI} pair in comparison with the potential of the Np^{VII}/Np^{VI} pair.

3. Interaction of actinides with aluminum in mixed hydroxides.

In the literature the particular attention is given to the problems of uranium and rare earths elements (REE) coprecipitation with aluminum hydroxide. On the one hand it concerns with the problems of concentrating and partitioning of microamounts of radioelements [11], on the other hand - method coprecipitation of aluminium and REE hydroxides with their subsequent incineration gives the possibility to obtain REE monoaluminates [12]. The latest serve, for example, as luminophores. At combined precipitation of Y, La or lanthanides with aluminium by ammonia (ðÍ 9-10) some interaction of components takes place in precipitates [3]. Such interaction becomes visible at thermal analysis. A crystalline phase of REE monoaluminates forms as a result of incineration of such precipitates at temperatures higher than 650-900°Ñ [12].

In the present work the results of examination of precipitates obtained at coprecipitation of lanthanum, neodymium (model elements), americium(III),

uranium(VI), thorium and neptunium(V) with aluminium at ðÍ 10 (ammonia solution) to 14 (1 M alkali excess) are given. No aluminates of actinides were isolated although some interaction of components has been found in precipitates not only of trivalent, but of tetra- and hexavalent actinides.

The precipitation with ammonia was used to determine the extent of components interaction in obtained hydroxides. For this purpose thermogravimetric, X-ray, IR-spectroscopic methods, and as well as EAS and measuring of dissolution rate of precipitates were utilized. The work is also devoted to the study of precipitates obtained at conditions of higher alkalinity – 0.1-1 M NaOH. At that the pure aluminum hydroxide should transfer into the soluble species. The problem was put to determine the effect of the components interaction on the aluminum solubility and other properties of precipitates.

Solid phases obtained in systems Al-Là, Al-Nd by ammonia addition at ambient temperature are poorly crystallized. The powder X-ray diffraction shows relatively narrow lines of bayerit $Al(OH)_3$ and wide lines of poorly crystallized lanthanum or neodymium hydroxide. No lines, which could be attributed to mixed crystalline hydroxide compound of aluminium with f-element, were observed.

The thermograms of precipitates Al-Là and Al-Nd do not differ essentially from ones described in the literature [12], i.e. they point out on some interaction of aluminium with the second component. This deduction is confirmed by the data of spectroscopic examination in visible and IR ranges of the indicated solid phases, and also of the precipitate containing Al and Am. At comparison of the spectra of solid pure and mixed hydroxides it is possible to see a number of differences. It is necessary to mark that IR spectra of crystalline hydroxides of lanthanides are well described in the literature [13,14]. The small number of absorption bands in these spectra corresponds to rather simple crystal lattice of compounds. As a rule, the narrow intensive absorption bands of "free" hydroxyl groups about 3600 cm^{-1}, the bands of deformation vibrations $\delta(\tilde{I}\tilde{I})$ in the range of 750-615 cm^{-1}, and the bands with composite structure about 400 cm^{-1} are observed. The stretching vibrations of metal-oxygen bonds are exhibited in the latest [15].

Hydroxyl groups in La(OH)$_3$ do not participate in hydrogen bridges, that was confirmed by neutron-diffraction [16]. The IR spectra of pure lanthanum and neodymium hydroxides obtained at indicated above conditions have little distinction from given in the literature. Apparently, owing to feeble crystallinity of the samples, alongside with narrow bands which are characteristic for oscillations of "free" OH-groups in the range of 3640-3620 cm^{-1}, the intensive wide bands with maximums at 3450-3420 cm^{-1}, referred to stretching vibrations $\nu(\tilde{I}\tilde{I}\tilde{I})$ are present in the spectra. The absorption bands at 1650-1620 cm^{-1}, which are characteristic for deformation oscillations of molecular water, specify that the hydroxides of lanthanum and neodymium have been obtained as hydrated substances such as Ln(OH)$_3$•nH$_2$O. There are two intensive bands in the range 1550-1320 cm^{-1}, at that the low-frequency one can be basically stipulated by some carbonization of precipitates. However in this case one can expect occurrence of bands of deformation oscillations of carbonates-ions about 860 or 700 cm^{-1} . It seems more exact to refer absorption in this region to nonplanar deformation oscillations of OH-groups. At adding of aluminum to REE the character of IR spectra of obtained solids varies by a different fashion in case of lanthanum and neodymium. The band in the region of 570-550 cm^{-1} which is characteristic for deformation oscillations of the Al-O bonds is present only in the spectrum of the precipitate Al-Nd. Besides, in the spectra of both mixed phases there is a band of small intensity in the region of 850-800 cm^{-1}, which is splitted on two components in the system Al-Nd.

Despite of practically equal radii of Am^{3+} and Nd^{3+} ions, the spectrum of pure Am hydroxide is distinct from the spectrum Nd(OH)$_3$. At first, the intensity of the band $\nu(\tilde{I}\tilde{I}\tilde{I})$ in the range of 3500-3300 cm^{-1} in the case of Am(OH)$_3$ is noticeably lower. Secondly, the absorption about 1500 cm^{-1} is practically absent. In the spectrum of the precipitate obtained at combined precipitation of americium(III) with aluminum(III), intensity of bands of valent and deformation vibrations of water molecules considerably increases, that it is possible to explain by formation of a mixed phase. At more low-frequency region the bands with composite structure having maxima at 850, 650 and 550 cm^{-1} are present, similar to the ones in spectrum of Al-Nd precipitate. Undoubtedly, in

the absence of any data about structure of Am(OH)₃ and of Al-Nd precipitate, the occurrence of new vibrational bands in the spectrum of mixed phases can testify both to formation of double compounds, and to formation of mixtures of starting components. However additional evidence that the precipitates, formed in double systems, are not mechanical mixtures, can be the results of measuring of visible spectra of precipitates obtained in systems Al-Am and Al-Nd (ions Am^{3+} and Nd^{3+} have characteristic absorption bands in visible range). For americium contained solids the absorption band with maximum about 503-510 nm is selected as analytical. In case of double phases the changes in the shape, intensity and position of characteristic absorption bands both of neodymium and americium are observed and in both cases the shift of maximum of bands appears in short-wave region of the spectrum in comparison with individual Nd or Am hydroxides. Such changes can be the result of some structural differences between mixed phases and individual hydroxides.

The IR spectra of mixed hydroxides of aluminum with uranyl and thorium exhibit some features of intermolecular interaction as in the case of REE and Am(III). A quite different spectral pattern is observed with precipitates formed in the system Np(V)-Al(III). In wide range of Al:Np ratios the measures spectra coincide with the ones of NpO_2OH, and the absorption bands typical for aluminum hydroxide are lacking. That is, no apparent interaction occurs between Al(III) and Np(V) under examined conditions, and the precipitates consist exclusively of NpO_2OH, differing only in the amount of interlayer water. This result agrees with our study [17] of coprecipitation of Np(V) with Fe(III) from alkiline solutions; in that work an Mossbauer spectroscopy examination revealed no interaction between the components.

Comparative examination of dissolution rates of hydroxides mixture and of pure components [18] is one of research methods to study interaction between hydroxides of metals in solid phase. This approach we utilized for estimation of possible interaction in hydroxide systems Al-Nd and Al-Am, obtained by ammonia precipitation. Dissolution of precipitates was carried out at ambient temperature. 1 M HCl was used at comparison of dissolution rates of hydroxides mixture relatively pure aluminium hydroxide. In experiments with pure neodymium and americium hydroxides stoichiometric quantity of ÍÑl was used, that is stipulated by rather high rate of

dissolution of pure components and, therefore, by necessity of retardation of the reaction. Dissolution processes were checked spectrophotometrically. The obtained mixed hydroxides are dissolved faster than pure components. Especially essential acceleration is observed relatively Al hydroxide. At heat treatment of pure aluminium hydroxide not readily soluble AlOOH forms, while the Nd or Am presence, apparently, hinders formation of pure hydroxide Al already at the stage of precipitation from a homogeneous solution. This fact can indirectly specify the presence of mixed «chains» such as -Nd (Am)-O-Al-O-.

We have studied the kinetics of dissolution of mixed hydroxides of some f-elements with aluminum in 0.01-5 M NaOH at 20 and 60°C. Nitrate of aluminum or mixture of aluminum and f-element nitrate solutions were placed in plastic tubes and hydroxides were precipitated by action of concentrated ammonia solution. The tubes were capped and stored at 90-95°C for 2-3 hours. After centrifugation precipitate was dispersed in 0.01-5 M NaOH and stored at 20 or 60°C in capped tube. The aliquots of supernatant were taken at selected time intervals and their pH were adjusted up to 3 by adding of HCl solution. After dilution the Al content was determined by ICP method at wavelength 396.152 nm using the standard solutions. In the case of Al(III)-Np(V) mixed hydroxides Al content was determined spectrophotometrically with xylenol orange in glicocol buffer at wavelength 555 nm.

The obtained results show that recovery of aluminum in system with Nd takes place at the same rate as for pure aluminum hydroxide. Dissolution of Al in Al-U(VI) system is slower. The experiments at 60°C have shown that recovery of aluminum is retarded in mixed hydroxides. We suppose that these results could be interpreted as additional corroboration of interaction of aluminum with f-elements ions in mixed hydroxides.

Once more method for revealing of components interaction in precipitates is measuring of specific volume. At pH rising of mother solution up to 13-14 (0.1-1 M NaOH solutions) the part of aluminum remains captured by REE precipitate. The specific volumes of REE and uranyl hydroxide precipitate with aluminum (measured after 7 hour of coagulation, and after centrifuging) are usually considerably higher, than for precipitate of pure REE hydroxide and sodium diuranate. The results obtained for lanthanum and neodymium do not essentially differ. Such effect was not brightly expressed in the case of americium.

With increase of molar ratio Al:La in starting solution the molar ratio of these elements in the precipitate also increases (0.1 M NaOH excess), though at the same conditions it can noticeably vary. IR spectra and powder X-ray diffraction data of the precipitates obtained at NaOH excess 0.1 M as at ambient temperatures, and at 150oÑ (at hydrothermal conditions), do not reveal any lines, which can unambiguously be referred to mixed compound. Thermograms of Al-La precipitates show the effects belonging to pure La hydroxide. Exo-effect in the region 650oÑ [12], which is characteristic for crystallization of lanthanum monoaluminate was not clearly observed. Probably, it is stipulated by small amounts of aluminium remaining in precipitate, obtained from 0.1 M alkali.

So, it is possible to assert that no individual compounds contained both Al and actinide were obtained. In mixed precipitates La, Nd, Am(III), U(VI) and Th(IV) with Al, obtained by ammonia addition some interaction of components occurs which is reflected in some characteristics of solid phase. Such interaction is strongly enough, and at pH rising up to 13-14 a part of aluminium remains captured by precipitate. However, this part is insignificant, and in precipitates obtained from 0.1-1 M NaOH, interactions of f-element with aluminium already is almost not visible.

So, no formation of actinides aluminates has been found and only individual compound of Al(III) and actinide (Np(VII)) has been isolated from alkaline solution. This compound represents aluminium neptunate, but not neptunium aluminate.

4. Interaction of actinides with aluminum in the course of simultaneous hydrolysis.
This part of the work concern the clarification of the reasons which lead to interaction of components in mixed precipitates of Al(III) with actinides. It is known now that some U(VI)-Al(III) interaction takes place at alkaline precipitation of solids from U-Al-containing solutions. Up to now the species formed at U(VI)-A1(III) interaction under lower pH conditions are not studied. The methods of EAS, luminescent and IR spectroscopy were used for characterization of the processes. It has been shown that absorption spectra of 10^{-4}-10^{-3} M uranium(VI) are changed at addition of Al(III) to the solution at pH 3.5-4.5. At pH-3.7 and [U(VI)]=0.005 M the increasing of aluminum

concentration in the range 0.005-0.05 M leads to greater changes in the spectrum. In contrast to pure uranium(VI) solutions, the spectra of the Al-containing solutions have a well-resolved vibrational structure. Absorption maximum positions in the presence of Al at higher pH values are the same as for $UO_2^{2+} \bullet$ aq, and only the intensity distribution is different. Uranium luminescence spectra are changed also in the presence of Al at appropriate pH. Luminescence spectra, as well as absorption ones, at pH 3.5-4.5 have vibrational structure resolved better than in absence of Al ($[U(VI)]=5 \bullet 10^{-4}$ M). However, the luminescence maximum positions are different from luminescence of $UO_2^{2+} \bullet$ aq. Al(III) presence influences the uranyl luminescence lifetime. For example, lifetime of 0.02 M U(VI) at pH~3 is equal to 4.2-5.4 ìs; at introduction of 0.15 M Al(III) it increases up to 7.0 ìs. Luminescence intensity under laser irradiation (ë =337.1 nm) at pH~4 is higher than for U(VI) solutions in the absence of Al. Luminescence spectrum of Al-contained systems is close to the individual spectrum of dimer species $(UO_2)_2(OH)_2^{2+}\cdot$aq obtained by mathematical treatment of the observed spectra [19]. On the other hand, luminescence spectrum in the presence of Al resembles the spectrum of $UO_2OSi(OH)_3^+$ obtained by the time-resolved luminescent spectroscopy method [20]. Our data on absorption and luminescence spectroscopy allow to suppose that at appropriate pH values and U(VI)-A1(III) concentrations aluminum(III) suppresses uranium(VI) polymerization, evidently due to formation of species containing both Al and U(VI). Al(III) hydrolysis takes place at the same pH values as $UO_2^{2+}\cdot$aq hydrolysis. Al(III) forms such species as $AlOH^{2+}$ and $Al(OH)_2^+$. So, two hydrolyzed ions, probably UO_2OH^+ and $Al(OH)_2^+$, interact with the formation of $UO_2OAl(OH)_2^+$ where one of O atoms is bridge. Another possible species is UO_2OAlOH^{2+}. Taking into account the coordination number of U(VI) and Al(III), the full composition of complexes can be presented as $(H_2O)_4UO_2OAl(OH)_2(H_2O)_3^+$ or $(H_2O)_4UO_2OAl(OH)(H_2O)_4^{2+}$. The formation of bridge OH-bonds is also possible: $(H_2O)_3UO_2(î\text{-}OH)_2OAl(OH)(H_2O)_3^{2+}$. This type of U-Al interaction may be described as some kind of cation-cation interaction. At pH increasing higher than 4 the solution grows turbid due to Al hydroxide formation. Thus the methods of luminescent and IR-spectroscopy were used for investigation of further U(VI) transformation. Luminescence

spectra were obtained for suspensions. At pH values about 5-6 the difference between individual U(VI) spectra and U(VI)-Al(III) spectra is visible. In the absence of Al(lII) the main contribution to the luminescence spectra gives the species$(UO_2)_3(OH)_5^+$·aq. It is difficult to propose the composition of compound formed in the presence of Al(III) on the basis of luminescence spectra. The solids of U(VI) were obtained at pH 7 which luminescence is also influenced by Al(III) presence. Further pH increase leads to disappearance of luminescence of solids without Al(III) addition. In contrast, solids containing Al(III) possess the luminescence which is the more bright, the more Al(III) is presented in the precipitate up to molar ratio Al:U=10:1. This is true at least up to pH 10. The luminescence spectra of solids obtained at pH 7-10 represent the wide band with a maximum near 515-520 nm. Data of IR spectroscopy show that the precipitates formed from solutions containing Al(III)+U(VI) at pH 5 to 10 all have some distinctions from mechanical mixtures of U(VI) and Al(III) solids obtained at the same pH. In particular the slight shift of the band near 900 cm^{-1} takes place. This points out on some increasing of U-O bond length in comparison with uranate. For jointly precipitated solids the character of splitting of the band at 525-420 cm^{-1} attributed to Al-O bond vibrations is also changed. Hence, data of IR spectroscopy indicate the uranium(VI)-aluminum(III) interaction. No formation of carbonate containing species (due to CO_2 from air) was observed.

Interaction of Np(VI,IV,V), Pu(VI,IV), Nd(III) and Am(III) with Al(III) in solutions at pH 0-4 was investigated by spectrophotometric method. In the range of pH 3-4 hydrolyzed species of neptunyl and plutonyl interact with hydrolyzed Al(III). For Pu(VI) possible composition of mixed hydroxo-aqua complexes appearing at the initial step of hydrolysis was determined as: $(H_2O)_3PuO_2(î-OH)_2Al(OH)(H_2O)_3^{2+}$ or $(H_2O)_4PuO_2OAl(OH)(H_2O)_4^{2+}$. Similar complexes are detected in case of Np(VI). The formation of mixed hydroxo-aqua complexes were observed at simultaneous hydrolysis of tetravalent plutonium and neptunium with Al(III) in pH range of 1.5-2.5. Complex of Np(IV) with Al(III) could be formulated as: $(H_2O)_n(OH)Np(î-OH)_2Al(OH)(H_2O)_3^{3+}$.

EAS of Np(V) and Nd(III) do not vary both in presence, and in absence of Al (III) up to pH~4 where the aluminum hydroxide precipitation begins. In this ðÍ region the

hydrolysis of Np(V) and Nd(III) is negligible. The presence of Al(III) practically does not influence the species of these ions. The hydrolysis of ion $Am^{3+} \cdot aq$ in 0.1 mol/l $NaClO_4$ solutions starts in a noticeable degree with rising ðÍ>5 [21,22]. At the presence of Al(III) at ðÍ about 4 the very weak increase of Am(III) ion absorption in the region 505-510 nm is visible. During further ðÍ augmentation the quantity of aluminum hydroxide precipitate increase. The capture of Nd(III) ions by the precipitate up to ðÍ~5 is no more than several %, of ions Np(V) is no more than 10 %. Thus, mixed hydroxo-aqua complexes of Nd(III) and Np(V) with Al(III) in solution are not detectable by our method. In the case of Am(III), such hydroxo-aqua complexes probably arise, but their yield at the investigated conditions is very small. The interaction of trivalent f-elements with Al(III) in the precipitate arises, probably, after sorption of the hydrolyzed species of f-elements by aluminum hydroxide precipitate. Such interaction is noticeably weaker, than in case of hexa- and tetravalent f-elements taking immediate part in precipitate formation.

It is possible to make a deduction on the basis of obtained results, that most strong interaction between hydrolyzed species Al(III) and An(VI) (An=U, Np, Pu) takes place. This is promoted by likeness in hydrolytic behavior of Al(III) and An(VI), including tend to formation the polymeric species, and the proximity of ðÍ ranges, in which hydrolysis and polymerization of these ions proceeds. The interaction between hydrolyzed ions Al(III) and An(IV) is also noticeably exhibited. Both hexa- and tetravalent actinides will form with aluminum mixed hydroxo-aqua complexes. If an interaction between Al(III) and trivalent f-elements is present, it is much less and we have no warrants to speak about formation of mixed hydroxo-aqua complexes. Interactions of Np(V) with the hydrolyzed species of aluminum(III) is not revealed.

5. Interaction of actinides with complex aluminate-ions.

Complexes of the trivalent f-elements with heteropolyanions (HPÀ) of the composition $XMo_6O_{24}^{n-}$ -where X=Mo(VI)[23], Te(VI)[24], I(VII)[24,25] are known. The listed HPA have planar Anderson structure. HPA of Anderson structure, which are very close on a composition to listed above, but having 6 additional protons per HPA formula

unit: $Ìe(OH)_6Z_6O_{18}{}^{m-}$ (where Z = Mo,W) can form when Me = Al, Cr, Fe, Ga, Ni [26]. The complexation study of molybdoaluminate-ion $Al(OH)_6Mo_6O_{18}{}^{3-}$ with ions of f-elements could represent some practical significance, as it is possible to assume a possibility of formation of such HPA in some objects of a storage of liquid radioactive wastes containing significant amounts of molybdenum and aluminum at various ðÍ of liquid phases[3]. f-elements interaction with some of listed HPA was studying in the works [27,28]. Their stability constants with $MnMo_9O_{32}{}^{6-}$, $TeMo_6O_{24}{}^{6-}$, $CrMo_6(OH)_6O_{18}{}^{3-}$ were determined by the method of lanthanide's micro concentration extraction [28]. Coordination modes between HPA and lanthanide ion were considered there and it was found that stability of the complexes with $CrMo_6(OH)_6O_{18}{}^{3-}$ anion is much lower than stability of $TeMo_6O_{24}{}^{n-}$ or $MnMo_9O_{32}{}^{6-}$ complexes. In spite of week interaction between lanthanide ions and $XMo_6(OH)_6O_{18}{}^{3-}$ (X=Al,Cr) in solution, we succeeded in isolation of crystalline complexes of the following common composition: $Ln[Al(OH)_6Mo_6O_{18}]\bullet nH_2O$, where Ln =La, Ce, Sm, Gd or Am(III), as well as $Eu[Cr(OH)_6Mo_6O_{18}]\bullet nH_2O$ complex. Their structures have been solved and coordination of ions of f-elements with polymolibdate ligands has been demonstrated.

Complexation of Nd(III) and Am(III) with $AlMo_6(OH)_6O_{18}{}^{3-}$ and also with $CrMo_6(OH)_6O_{18}{}^{3-}$ in solution was studied spectrophotometrically. Calculated values of stability constants for Am(III) complexes are: â=18±6 for $AlMo_6(OH)_6O_{18}{}^{3-}$ and â=25±5 for $CrMo_6(OH)_6O_{18}{}^{3-}$ (ì=0.1, pH 3.90 and 4.55 respectively). The last one is about 10 times lower than given in [27] for Eu(III) complex with $CrMo_6(OH)_6O_{18}{}^{3-}$. The study of Eu(III) and Cm(III) behavior in solutions of $AlMo_6(OH)_6O_{18}{}^{3-}$ was carried out by luminescence method. Solutions $AlMo_6(OH)_6O_{18}{}^{3-}$ with europium(III) concentration up to 10^{-3} M do not exhibit luminescence under nitrogen laser irradiation and this indicates absence of complex formation. The proper Eu(III) absorption at the such wave length is negligible and no energy transfer from polymolybdate ion to Eu(III) does not occurs. At such excitation $3 \cdot 10^{-5}$ mole/L solution of $Cm(ClO_4)_3$ has very weak luminescence. Addition of $AlMo_6(OH)_6O_{18}{}^{3-}$ up to 0.02M leads to noticeable increasing of curium luminescence, its maximum is situated at 594 nm and lifetime was found 63±4 μs. Theses luminescent

parameters are close to ones of Cm(III) aqua-ion [29], but the fact of sensitizing indicate a weak complex formation Cm(III) with $Al(OH)_6Mo_6O_{18}^{3-}$ anions (it should be noted that intermolecular energy transfer from HPA is extremely improbable).

The crystals $Ln[Al(OH)_6Mo_6O_{18}] \cdot nH_2O$ [Ln= lanthanides, Am(III)] are formed for several hours at room temperature in 0.01-0.02 M $(NH_4)_3[Al(OH)_6Mo_6O_{18}] \cdot nH_2O$ solution, containing equimolar quantity of corresponding rare earth element as nitrate salt. $(NH_4)_3[X(OH)_6Mo_6O_{18}] \cdot nH_2O$ (X=Al,Cr) were prepared in accordance with [30]. Crystalline structure of $Sm[Al(OH)_6Mo_6O_{18}] \cdot 11H_2O$ is built up from infinite chains located along a direction [001] and consisting from $[Al(OH)_6Mo_6O_{18}]^{3-}$ anions, Sm^{3+} cations and molecules of coordination water. There are also crystallization water molecules in a structure, holding by hydrogen bonds (Fig.2). The basis of the anion $[Al(OH)_6Mo_6O_{18}]^{3-}$ consist of Al atom, enclosed by six Mo atoms lying in one plane. The maximum deviation of atoms from an average plane is 0.042(1) Å. The distances between the nearest Mo atoms are equal to 3.28(1) - 3.32(1) Å. Al-Mo distances are equal to 3.28(1) - 3.33(1) Å. The oxygen atoms of hydroxyl groups forming a coordination polyhedron of aluminum atom (octahedron), bind it with molybdenum atoms. The angles O-Al-O differ from ideal value 90° and are equal to 84.4(6) - 95.8(6)°. The Al-O distances are 1.90(2) - 1.91(2) Å. Coordination polyhedron of molybdenum atoms is distorted octahedron which is formed by oxygen atoms of various types: by two end, two bridge, binding adjacent Mo atoms, and two bridge, binding adjacent Mo and Al atoms. Depending on oxygen atoms executed functions Mo-O distances change: 1.65(2) - 1.75(2), 1.90(2) - 1.98(2), 2.27(2) - 2.34(2) Å. Each octahedron of Mo atom has one common edge with two adjacent MoO_6 octahedrons and octahedron AlO_6 of Al atom. The anions are united in chains by Sm atoms. A coordination polyhedron of Sm atom - trigonal tricaped prism, in tops of which there are two end oxygen atoms of two adjacent anions and seven molecules of water. The angle between the basis of a prism is equal to 7.5°, the angles between lateral faces are 58.2, 58.3 and 63.5°.
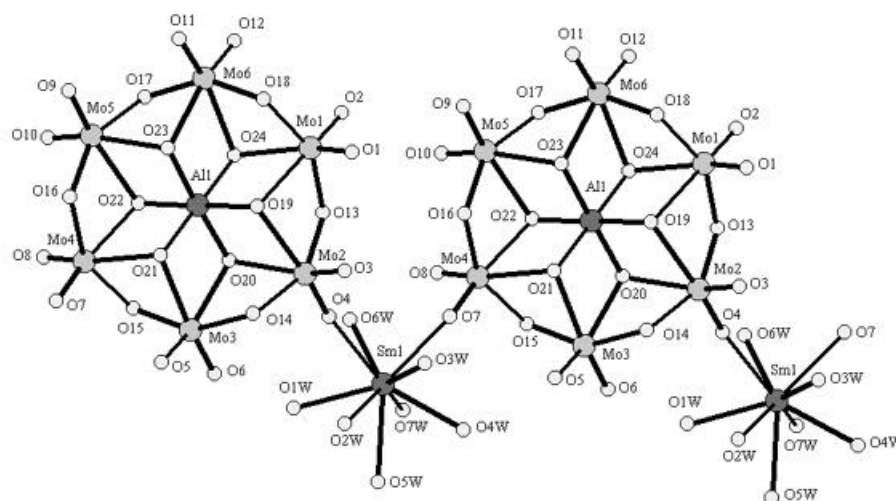
Fig.2. Crystalline structure fragment of $Sm[Al(OH)_6Mo_6O_{18}]\cdot11H_2O$.

Crystalline complexes of Eu, Tb, Sm, Dy with $Al(OH)_6Mo_6O_{18}^{3-}$ exhibit REE luminescence at nitrogen laser excitation resulting from energy transfer from ligand to f-element ions. Bright luminescence of Tb(III) in this compound is unexpected because of the already reported quenching by charge transfer between Tb(III) and Mo(VI). The hydration numbers n of REE ions in compounds $Ln[Al(OH)_6Mo_6O_{18}]\bullet nH_2O$ have been calculated using the luminescence lifetime values for solids prepared from H_2O and D_2O: $n=7.65\pm1.44$ for Eu, Tb, Sm tripositive ions; X-ray data for $Sm[Al(OH)_6Mo_6O_{18}]\cdot11H_2O$ gave $n=7$. Crystalline complexes were investigated by means of IR spectroscopy and thermogravimertry.

Uranyl and Np(IV,V,VI) give the precipitates with $(NH_4)_3[Al(OH)_6Mo_6O_{18}]\bullet nH_2O$ of relatively low solubility - less than $10^{-3}M$.

The attempts to synthesize the compounds of Np(VI,V), Pu(VI) and uranyl with hexafluoroaluminate-ion were unsuccessful. Pu(VI) and Np(VI) are slow reduced even at low acidity of solutions. Np(V) disproportionates giving the insoluble fluoro-complexes of Np(IV), among which the complex Na_2NpF_6 with unknown structure was found.

Thus, possible influence of heteropolyanions containing Al, on the actinides species in tanks is negligible, though the solid complexes of trivalent f-elements have been separated and their structure is studied.

Relevance, Impact and Technology Transfer

a. In our opinion the one of the principal conclusion concerning the critical DOE environmental management problems is the absence of aluminate-ion influence on the species and stability of actinides in different oxidation states in alkaline liquid HLW.

b. Behavior of actinides during the technologic operations with HLW such as aluminium leaching now is more clear and could be predicted more precisely.

c. The study of trivalent f-element complexes with heteropolyanion $Al(OH)_6Mo_6O_{18}^{3-}$ as well as with $Cr(OH)_6Mo_6O_{18}^{3-}$ gave the start to intense investigation of the structure and luminescent properties of these compounds due to unusual pathway of energy and charge transfer. In our laboratory in cooperation with Professor Jean-Claude Krupa (head of laboratory of radiochemistry of Institute of Nuclear Physics, France, Orsay) this study will be developed.

d. Our work deals with more scientific aspects of HLW management so we could say nothing about the trial scale.

e. Unfortunately we had not any collaboration with American research group.

f. It is difficult to estimate importance of our own work but it seems to us that results concerning the mechanism of actinides complexation with aluminium in course of simultaneous hydrolysis and interaction of actinide ions with heteropolyanions are quite interesting.

g. There are no evident obstacles in using of obtained results in DOE Environmental Management problems.

h. We had not possibilities to discuss the obtained results with American research group or representative of DOE.

6. Project Productivity

7. Personnel Supported

G.B.Andreev was supported during preparing of his Ph.D. thesis.

8. Publications

a. Published in peer-reviewed journals:

1. A.B.Yusov, N.A.Budantseva, A.V.Anan'ev, A.M.Fedosseev, "Coprecipitation of aluminum with hydroxides of tetra-, penta-, and hexavalent actinides", Radiokhimiya, 2000, V.42, №5, P.413-416. (English Translation Radiochemistry, 2000, V.42, №5, P.456-460).

2. Alexander B.Yusov, Alexander M.Fedosseev, Grigorii B.Andreev, Irina B.Shirokova, "Luminescence properties of solid Eu, Sm, Tb and Dy compounds with the molybdoaluminate ion $Al(OH)_6Mo_6O_{18}{}^{3-}$". Mendeleev Commun., 2001, №2, P.86-88.

3. Mikhail V.Nikonov, Nina A.Budantseva, Alexander M.Fedosseev. "Synthesis of neptunium(VII) compounds with aluminium and europium in alkaline solutions". Mendeleev Commun., 2001, №2, P.102-104.

4. A.B.Yusov, N.A.Budantseva, M.V.Nikonov, I.A.Charushnikova, I.B.Shirokova, A.M.Fedosseev, "Interaction of aluminum with Am(III) and certain REE(III) ions in alkaline solutions", Radiokhimiya, 2001, V.43, №4, P.333-336. (English Translation Radiochemistry, 2001, V.43, №4, P.379-383).

5. A.B.Yusov, N.A.Budantseva, A.M.Fedosseev, L.N.Astafurova, "Interaction of aluminun(III) with uranyl in course of simultaneous hydrolysis", Koordinatsionnaya Khimiya, 2001, V.27, №11, P.824-830. (Russian Coordination Chemistry).

b. Proceedings, technical reports etc.:

1. M.V.Nikonov, N.A.Budantseva, "Synthesis of heptavalent actinide solid compounds with Al and Eu in alkaline media", 5th International conference on nuclear and radiochemistry, Pontresina, Switzerland, September 3-8, 2000, Extended Abstracts, V.1, P.138-139.

2. A.B.Yusov, N.A.Budantseva, T.C.Lapitskaya, I.A.Charushnikova, M.V.Nikonov, "Study of aluminum interaction with Am(III) and certain REE(III) ions in weakly alkaline solutions", Third Russian conference on the radiochemistry. Radiochemistry-2000. Abstracts, Sankt-Petersburg. 2000, P.20.

3. A.M.Fedosseev, A.B.Yusov, S.I.Pisarev, "Luminescence of crystalline complexes of lanthanides with molybdoaluminate-ion $Al(OH)_6Mo_6O_{18}{}^{3-}$". Third Russian conference on the radiochemistry. Radiochemistry-2000. Abstracts, Sankt-Petersburg. 2000, P.50.

4. N.A.Budantseva, A.M.Fedosseev, A.B.Yusov. "Aluminium(III)-Uranium(VI) and Aluminium(III)-Plutonium(VI) interaction during hydrolysis". Global 2001. International conference on: "Back-end of fuel cycle: from research to solutions". Paris, France, September, 9/13, 2001. Book of Abstracts. Abstract 041.

5. A.B.Yusov, N.A.Budantseva, A.M.Fedosseev, J.-C.Krupa. "Aluminium(III) interaction with actinides(III), (IV), (V), (VI) during simultaneous hydrolysis". Actinides-2001. International Conference. Hayama, Japan. November 4-9, 2001. Final Program & Abstracts. 3P38, p.119.

6. A.B.Yussov, A.Ì.Fedosseev, G.B.Andreev, I.B.Shirokova, J.-C.Krupa, "Luminescence Study of New Ln(III) Complexes with Anderson's Heteropolyanions $Al(OH)_6Mo_6O_{18}^{3-}$ and $Cr(OH)_6Mo_6O_{18}^{3-}$", Rare Earths'-2001. Brazil, Campos do Jordão, 22-26 September 2001, Abstracts Book. p. 119.

c. Accepted papers in peer-reviewed journals:

1. A.M.Fedosseev, N.A.Budantseva, A.B.Yusov, G.B.Andreev, I.B.Shirokova, "Hexamolybdoaluminates and hexamolybdochromates of certain trivalent lanthanides and americium", Koordinatsionnaya Khimiya, 2002, V.28, ¹4 (Russian Coordination Chemistry).

2. A.B.Yusov, A.M.Fedosseev, L.N.Astafurova, "Interaction of aluminun(III) with f-elements ions {Np(VI), Pu(VI), Np(V), Np(IV), Pu(IV), Nd(III), Am(III)} in course of simultaneous hydrolysis", Koordinatsionnaya Khimiya, 2001, V.28, ¹4, (Russian Coordination Chemistry).

3. A..B.Yussov, A.Ì.Fedosseev, G.B.Andreev, I.B.Shirokova, J.-C.Krupa, Luminescence Study of New Ln(III) Complexes with Anderson's Heteropolyanions $Al(OH)_6Mo_6O_{18}^{3-}$ and $Cr(OH)_6Mo_6O_{18}^{3-}$, J. Alloys.Comp., 2002.

9. Interactions.

Part of the work dealing with the luminescence of lanthanides complexes with $(NH_4)_3[X(OH)_6Mo_6O_{18}] \cdot nH_2O$ (X=Al,Cr) was accomplished in the collaboration with Professor Jean-Claude Krupa – head of laboratory of radiochemistry of Institute of

Nuclear Physics, France, Orsay. Some samples prepared in Moscow were sent to Orsay for detailed study of their luminescence by means of more appropriate apparatus.

10. Transitions

11. Patents

None

12. Literature Cited

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4. A.M.Fedoseev, N.N.Krot, N.A.Budantseva, A.A.Bessonov, M.V.Nikonov, M.S.Grigoriev, Yu.A.Garnov, V.P.Perminov, L.N.Astafurova. Interaction of Pu(IV, VI) Hydroxides with Metal Hydroxides/Oxides in Alkaline Media. Report PNNL-11900, UC-2030. Pacific Northwest National Laboratory. Richland, Washington. August 1998.

5. N.A.Budantseva, A.M.Fedoseev, I.G.Tananaev, A.A.Bessonov, C.Delegard. Investigation of the Behavior of Plutonium in Alkaline Media. Report PNNL-11624. Pacific Northwest National Laboratory Richland. USA. Sept.1997.

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13. Feedback

14. Appendicies

15. Quantities/Packaging