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THE AMALGAMATION BEHAVIOR OF HEAVY ELEMENTS. 3. EXTRACTION OF RADIUM, LEAD AND THE ACTINIDES BY SODIUM AMALGAM FROM ACETATE SOLUTIONS.

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### THE AMALGAMATION BEHAVIOR OF HEAVY ELEMENTS. 3. EXTRACTION OF RADIUM, LEAD AND THE ACTINIDES BY SODIUM AMALGAM FROM ACETATE SOLUTIONS.

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December 1967

#### ABSTRACT

This paper presents extraction yields of radium, lead and the actinides from Ac through No, with sodium amalgam from 7 M sodium acetate solutions. The extraction yield is shown to depend upon the amount of HCl added to 7 M sodium acetate solution before extraction. Californium, einsteinium, fermium, mendelevium, radium and lead are extracted with high yields (75% to 100% relative to Cf), while the lower actinides, Ac, Th, Pa, U, Np, Am, Cm and Bk are extracted with low yield (below 10% relative to Cf). The addition of oxidizing agents such as  $H_2O_2$  or  $HNO_2$  before extraction strongly suppresses the extraction, except in the case of lead. Investigation of the extraction behavior of all actinide elements (except lawrencium, which has no isotopes suitable for conventional chemical study) and a survey of the literature of the extraction behavior of the lanthanides show cases of uncorrelated extraction properties in both series. On the whole, however, high extraction yields correlate with independent evidence for the existence of a dipositive oxidation state in the lanthanides and suggest the possible formation of this oxidation state in californium and heavier actinides.

On leave from the Institute of Nuclear Research, Řež near Prague, Czechoslovakia.

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# INTRODUCTION

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Methods for separating from rare earth mixtures those lanthanides which have a well-characterized 2+ oxidation state (Eu, Sm, Yb) by extraction with alkali-metal amalgams are well known. (1-11) Europium, Sm and Yb may be extracted from other rare earths with sodium amalgam from acetate solution (2-5,7) with yields between 50% and 100% (2-5,7) as compared with yields of less than 10% for other rare earths. The heavy rare earths (from Gd to Tm) are practically not extracted. (7)

The first application of this separation method to the actinide series was the extraction of Ac, Pu, Am and Cf from HCl - 0.03 M sodium citrate solutions.<sup>(12,13)</sup> Recent results are reported in Ref. 14. The extraction of Pu, Am, Cm, Bk, Cr, Es, Fm and Md from 7 M sodium acetate with sodium amalgam shows unexpectedly high yields of Cf, Es, and Fm, while Pu, Am, Cm, and Bk were extracted about 10 times less efficiently, similar to the higher lanthanides. In this paper the results of simultaneous extractions of <sup>223</sup>Ra, <sup>211</sup>Pb, <sup>227</sup>Th, <sup>238</sup>U, <sup>237</sup>Np, <sup>239</sup>Pu, <sup>241</sup>Am and <sup>252</sup>Cf are shown to exhibit a dependence on the amount of HCl added before extraction, or on the presence of oxidants (H<sub>2</sub>O<sub>2</sub>) or HNO<sub>2</sub>). The yields of  $^{239}$ Pu,  $^{244}$ Cm,  $^{252}$ Cf,  $^{253}$ Es and  $^{255}$ No (with ~4 min half life) on extractions carried out as described in Ref. 14 are also given. The results of this work, together with previous results, (14,15) permits the presentation of the mean extraction yield of all actinides from Ac to No and a comparison of this curve with a similar one for lanthanide extractions. The results are interpreted in terms of the tendency to form the 2+ oxidation state in both series.

#### EXPERIMENTAL

# The choice of radioisotopes and the production of <sup>255</sup>No

The extraction behavior of the actinides, Ra and Pb, was studied by using an " $\alpha$  mixture" in 0.5 M HCl consisting of <sup>239</sup>Pu<sup>4+</sup>, <sup>241</sup>Am<sup>3+</sup>, <sup>252</sup>Cf<sup>3+</sup>, <sup>238</sup>U<sup>4+</sup>, <sup>234</sup>Np<sup>4+</sup> and isotopes of the <sup>231</sup>Pa series (<sup>231</sup>Pa<sup>5+</sup>, <sup>227</sup>Th<sup>4+</sup>, <sup>227</sup>Ac<sup>3+</sup>, <sup>223</sup>Ra<sup>2+</sup> with <sup>219</sup>Rn and <sup>215</sup>Po, <sup>211</sup>Pb<sup>2+</sup> with <sup>211</sup>Bi). These isotopes were present at approximately equal  $\alpha$  activities, and for one experiment 20  $\lambda$  of  $\alpha$  solution giving about 100 detectable  $\alpha$  counts/min of each radioisotope was used.

In the study of nobelium about 20 detectable  $\alpha$  counts of <sup>255</sup>No, caught on a platinum catcher plate from a gas jet were used for each experiment. After a 10-minute target irradiation of <sup>244</sup>Pu with <sup>16</sup>O to give <sup>255</sup>No in the HILAC (as described elsewhere), <sup>(16)</sup> the <sup>255</sup>No was dissolved on the catcher foil, using 5  $\lambda$  of HCl and sodium acetate solution containing tracer <sup>239</sup>Pu<sup>4+</sup>, <sup>241</sup>Am<sup>3+</sup>, <sup>244</sup>Cm<sup>3+</sup>, <sup>252</sup>Cf<sup>3+</sup> and <sup>253</sup>Es<sup>3+</sup>. This "No mixture" was used for extraction directly from the plate, as explained later.

# Extraction and electroplating procedure for runs with the " $\alpha$ mixture"

For every extraction experiment 20  $\lambda$  of " $\alpha$  mixture" were mixed in a 3-ml cone with 5  $\lambda$  of 8 M ammonium acetate, 150  $\lambda$  of 7 M sodium acetate, and 3  $\lambda$  of LaCl<sub>3</sub> solution, containing 300 µg La<sub>2</sub>O<sub>3</sub>. To this mixture 250  $\lambda$  of sodium amalgam were added from a short spitzer (pipette), in 3 or 4 portions about a minute apart. The amalgam and tracer-containing sodium acetate solution were mixed intensively by sucking in and out of the spitzer (40 to 50 times during 4 min). Foaming and hydrogen evolution occurred on introduction

of the first portion of amalgam, but by the fourth the foaming usually stopped and a precipitate of  $La(OH)_{\gamma}$  appeared in the solution. After 4 min of extraction, the mercury was separated from the original solution and washed twice with 300  $\lambda$  of 6 M NH, OH. The extracted elements were back-extracted from the mercury into 400  $\lambda$  of 6 M HCl. After the solution was neutralized to pH 4 (methyl red indicator) the activities were plated out on a Pt disc, flamed and counted by  $\alpha$ -pulse analysis. The electroplating was performed from ~1 ml of 3 M NH<sub>4</sub>Cl solution. The cathode was a Pt disc of about 1 cm<sup>2</sup> surface area. Electrolysis was continued for about 10 min at a current of about 500 µamp and an applied potential of 5 v. For better electrodeposition of Ra, 100  $\lambda$  of a saturated solution of ammonium oxalate were added to the NH, Cl solution. The extraction yield was measured by comparison with the same amount of " $\alpha$  mixture" electroplated directly on the platinum disc. Electroplated Pb, Th, Pa, U, Np, Am, Pu and Cf were compared to the various activities measured after direct evaporation of the same volume of  $\alpha$  mixture. Yields of 50-100% were observed in the plating of all elements except radium for which the yield was only 10-20%. When the ammonium oxalate was not present, the plating of Ra dropped to only  $\approx 5-10\%$ .

#### Extraction of nobelium with sodium amalgam

A micro-modification of the extraction method described above was used for nobelium. Five microliters of tracer solution in 3 M HCl were placed on the catcher plate on which  $^{255}$ No had been collected. The droplet was spread and stirred for 10-20 sec to dissolve the No; 10  $\lambda$  of a solution 7 M in

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sodium acetate and 0.26 M in ammonium acetate containing 10  $\mu$ g La<sup>3+</sup> were then added. After an additional 10 sec of mixing, 15  $\lambda$  of sodium amalgam (3.5 milligram equivalents of Na/ml) were added and mixed on the surface of the Pt catcher foil for 1 min. The mercury phase which adhered to the plate was washed successively with a stream of 6 M NH<sub>4</sub>OH, water, and then alcohol, for 5 sec each, with mixing. The mercury was driven off by heating with a flame and the plate counted in an  $\alpha$ -pulse chamber. Usually the start of counting was within 2.5 min from the end of bombardment.

#### $\alpha$ -pulse analysis and errors of measurement

Counting was done with a standard grid  $\alpha$  chamber, connected with an  $\alpha$ -pulse analyser. In the experiments with nobelium four successive 3-min counts were performed; " $\alpha$  mixture" counts were for one or two 30-minute (or longer) intervals. The  $\alpha$  spectrum of the " $\alpha$  mixture" before extraction is shown in Fig. 1. Yields before and after extraction were computed from the sums of the  $\alpha$  counts in 6 or 10 channels near characteristic  $\alpha$  peaks in the spectra.

As shown, the spectrum exhibits good resolution, except for  $^{227}$ Th and  $^{252}$ Cf, especially after extraction. The usual counting error was ~5% to 10%, but in the case of  $^{227}$ Th (and also  $^{227}$ Ac) it was 30-50%. The relative error in repeated measurements of the same quantity was usually within ±50%; in the case of  $^{227}$ Th; reproducibility was ±70%. By taking the average of two to four results, the relative errors were reduced to about ±30% and ±50%, respectively.

#### Chemicals

The sodium amalgam, containing 3.5 to 4.0 milliequivalents Na per ml, was prepared by dissolving small pieces of freshly-cut sodium metal in mercury. Chemicals used were "chemically pure" grade.

#### RESULTS

#### Extraction of No and heavy actinides

Extraction conditions were similar to those described for experiments 8 and 9 in Ref. 14. Results of a typical experiment which compares the tracer mixture (a) with the extract (b) are shown in Fig. 2. The separation of Es and Cf on the one hand from Am, Cm and Pu on the other, is good with a separation factor  $\approx 10$ , in a total time of 2.5-3.0 min. All extraction data (including 27 separate experiments with No) are summarized in Table 1 along with comparative data from Ref. 14. Comparison of extraction ratios found in the present work with those from Ref. 14 shows good agreement for Pu and Am, but substantial variation for Cm. The extraction of No is lower than Cf, but substantially higher than that of lighter actinides. In Ref. 16 it was found that No exhibits a stable 2+ state in solution, with basic behavior similar to Ca<sup>2+</sup> or Sr<sup>2+</sup>. This explains the lower extraction yield of No, which resembles the extraction yield of Ra rather than Cf.

#### Extraction of lower actinides, radium and lead

The " $\alpha$  mixture" was extracted by the same procedure described above. Conditions were identical except for the addition of various amounts of HCl

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to the solutions just prior to adding the amalgam, as noted in Fig. 3. The volume of added HCl was in all cases 75  $\lambda$ ; the concentrations used were 0.5 M, 0.25 M, 1 M, 2 M and 3 M. The initial pH was 6.0 to 7.0, without any addition of HCl; with the addition of the 75  $\lambda$  of 3 M HCl, the pH dropped only to 5.0 to 6.0. Larger amounts of hydrogen were evolved from the more acid solution. In all cases, the final pH was 10 to 11 and a clearly visible precipitate of La(OH)<sub>3</sub> appeared.

The data of Fig. 3 clearly show that Cf, Ra, and Pb are extracted with sodium amalgam with very high yield, similar to Yb and the other lanthanides which can exhibit 2+ valency. The actinide homologs of Sm and Eu--Pu and Am--are extracted poorly (~10%) relative to Cf, but comparably to the light lanthanides, like La, Ce, Pr and Nd. <sup>(11)</sup> The results in Fig. 3 are presented in terms of the ratio of yield to the yield of californium ("californium numbers") in the same experiment in Fig. 4.

Figs. 3 and 4 show that  $Pu^{4+}$  is extracted best after Cf (0.122 of Cf yield),  $Ac^{3+}$  and  $Am^{3+}$  somewhat less (0.099 of Cf yield),  $U^{4+}$  and  $Th^{4+}$  still less (0.077 to 0.060 of Cf yield), and  $Pa^{5+}$  least of all. From Fig. 3, it appears that the percent extraction of Ac, Th, Pa, U, Np, Pu, Am and Pu is approximately linear with the amount of HCl added; the yield approximately doubles for every 100  $\lambda$  of added 1 M HCl (or for 0.0001 milligram equivalent of evolved hydrogen). However, the extraction values for only 4  $\lambda$  of added HCl are substantially higher than for 20  $\lambda$ . The reason for this effect is not known; it may be connected with the rapid increase in pH near the beginning of extraction. The reaction of the amalgam with NH<sub>4</sub><sup>+</sup> present in extraction solution (from 5  $\lambda$  of 8 M NH<sub>4</sub> acetate):

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 $\operatorname{NaHg}_{X} + \operatorname{NH}_{4}^{+} \longrightarrow \operatorname{Na}^{+} + \operatorname{XHg} + \operatorname{NH}_{3} + 1/2 \operatorname{H}_{2},$ 

raised the pH more rapidly than the reaction with H<sup>+</sup> present in more acid solutions.

Bouissières et al. (12,13) observed high yields of Ac, Cf, Fu and Am and very low yields of Pa and Th by extraction with lithium amalgam from 0.03 M sodium citrate solution with HCl addition in the range between pH = 1 to pH = 8 or 11. This behavior differs substantially from that found in the work reported here using acetate solution with HCl addition in a pH range 6-11, and sodium amalgam extraction. In other work (2-11) carried out under conditions similar to ours it was found that Sm is the most extractable element (see Fig. 6). Réf. 12, on the contrary, indicates that Sm is less extractable than Eu, and in the pH range 6-11, about 5-10 times less. Similar experiments (12,13) indicate that La is extracted with high yield (more than Sm at pH 2-3) as are Ac and Cf. Other results (see Fig. 6), however, confirm that under conditions similar to those used in the present work, La is poorly extracted. It would seem that extractions using sodium amalgam and acetate solution are not comparable to those using diluted citrate solutions and lithium amalgam.

# Dependence of extraction yield on amount of evolved hydrogen

Marsh<sup>(2)</sup> first suggested an explanation for the high yield in Eu, Sm and Yb amalgam formation in terms of a two-step reaction in which these

elements are first reduced to the 2+ state and then extracted by an exchange reaction similar to the alkaline earths,  $Me^{2+} + 2Na \xrightarrow{(in Hg)} 2Na^+ + Me metal(in Hg)$ . This suggestion is supported by other workers also. (7,9-11) In the work reported here, the extraction yields were checked under conditions of extraction where the " $\alpha$  mixture" contained the oxidants, HNO<sub>3</sub> or H<sub>2</sub>O<sub>2</sub>. The extraction was performed using the same concentration of  $H^+$  in sodium acetate solution and the same procedure as when no oxidant was present. Four extractions were carried out in the presence of oxidants (15  $\lambda$  of 5 M HNO<sub>2</sub>, 5  $\lambda$  of 5 M HNO3, 34  $\lambda$  of 39% H<sub>2</sub>O<sub>2</sub> and 7  $\lambda$  of 39% H<sub>2</sub>O<sub>2</sub>) and one extraction without oxidation. In all 150 milligram equivalents of  $H^+$  (HC1 or HC1 + HNO<sub>2</sub>) were added before extraction to a 7 M sodium acetate solution of the " $\alpha$  mixture." The results are shown in Fig. 5. It is clearly evident that the addition of oxidant drastically suppressed the extraction of all elements, with the exception of Pb<sup>2+</sup>. Hydrogen evolution was not observed when 15  $\lambda$  of 5 M  $HNO_3$  or 34  $\lambda$  of 30%  $H_2O_2$  were present. In both cases reduction of the oxidant can consume 600 micromoles of  $H^+$  in comparison to 230 micromoles  $H^+$ added. Suppression of the extraction yield was approximately the same for all elements from Ac to Cf. Cf in the presence of H20, was still extracted about 10 times better than the other actinides. Different results were observed in extractions in the presence of only 5  $\lambda$  of 5 M HNO, or 7  $\lambda$  of 30%  $H_2^{0}$  (consumption of only 120 micromoles  $H^+$ ). In the case of 5  $\lambda$  of 5 M  $\mathrm{HNO}_{\mathrm{Q}}$ , the suppression of the extraction was substantial; in the case of 7  $\lambda$  of 30%  $\text{H}_{2}\text{O}_{2},$  only the yield of Ac was suppressed substantially, while the yields of U and Np (oxidized to  $UO_2^{2+}$  or  $NpO_2^{2+}$ ) were slightly increased. It was remarkable that the Ra yield was also suppressed by the presence of

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oxidants. The conclusion from these experiments--the strong suppression of extraction yield when no hydrogen is evolved--and also, the lack of effect on the yield of Pb<sup>2+</sup> (which exists as such even in the presence of the oxidants) strongly supports Marsh's suggestion, that first the 3+ ions are reduced to 2+ and then are easily extracted. The same effect was observed qualitatively in the extraction of ~500 µg of Yb from a solution containing 15  $\lambda$  of 5 M HNO<sub>3</sub>, added to 7 M sodium acetate. The green color of Yb<sup>2+</sup> was not observed and Yb was not recovered from the amalgam. A parallel experiment in the absence of HNO<sub>3</sub> showed the green color of Yb<sup>2+</sup> in solution during the extraction and  $\approx$ 50% of the Yb was recovered from the amalgam by treatment with HCl and subsequent precipitation of Yb(OH)<sub>3</sub> with KOH. The stronger suppression of extraction by HNO<sub>3</sub> than by H<sub>2</sub>O<sub>2</sub> suggests that the reaction of nascent H with H<sub>2</sub>O<sub>2</sub> is probably slower than with HNO<sub>3</sub>.

#### Californium numbers

From Fig. 4 it may be seen that "californium number" (yields relative to the californium yield) for the actinides remains approximately constant over the whole range of added HCl. This shows that the "mean californium number" (mean value from Fig. 4) may be used as a base value to define the extraction behavior of other actinides. Table 2 shows these "mean californium numbers" from the present work (column 1) and from the preceding work (column 2) and also (in parentheses) the number of individual experiments used to calculate them. Weighted mean values from all experiments are given in column 3. Californium numbers for No, Md, Fm and Es are less reliable than the values for other elements, since they were obtained for only one or two volumes of added HCl. Hence, as shown in Fig. 4, this can lead to  $\approx 50\%$ error in the californium number. However, this will not affect the classification of these elements as highly or poorly extractable actinides.

### DISCUSSION

The dependence of the "californium numbers" on atomic number is plotted in Fig. 6 for all actinides from Ac to No. and the plot also includes Pb and Ra (lower scale). On the figure (upper scale) are plotted the mean "californium numbers" calculated in Table 3 from the experimental results reported in Refs 2 and 11, which give relative extraction yields of La, Ce, Pr, Nd, Sm, Eu, Gd and Yb. Upper limits for the extraction of Ho, Er, Tm and Lu were taken from Ref. 7. The experimental work was performed similarly to the measurements made in these laboratories for Dy (the authors of Ref. 7 show zero extraction yield for Tb, Dy, Ho, Er, Tm and Lu). The percentage yields of the lanthanides were recalculated from the data given in Refs.  $\frac{1}{4}$ , 7 and 11 by using the value 0.748 for Yb as measured in this laboratory (see Table 2). Comparison of the curves for the lanthanide and actinides reveals obvious differences in extraction behavior. The lanthanides exhibit high extraction yields (Cf number = 1 or approximately 66% of extraction) at Sm and Eu in the light lanthanides and at Yb in the heavy lanthanides. The actinides give one broad peak from Cf to No, and possibly a slight maximum between Pa and Bk. However, this maximum is about ~15 times lower than the

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maximum of the heavy actinides at Fm. The extraction yields plotted in Fig. 6, which have californium numbers above 0.70, indicate that these have 2+ states in solution which are relatively more stable in elements the solution than the others which have low extraction yields. This correlation is well established in the lanthanide series, where the least negative reduction potentials for the Me<sup>3+</sup> + e  $\longrightarrow$  Me<sup>2+</sup> couple are for Eu (-0.43 v), (17) Sm  $(-\sim 0.9 \text{ v})^{(17)}$  and Yb  $(-0.58 \text{ v}^{(17)} \text{ or } -1.15 \text{ v} \text{ calculated from } \Delta F_{f} \text{ values})^{(18)}$ and correspond to maximum extraction into Na-Hg, as shown in Fig. 6. The reduction potentials of all other lanthanides for  $Me^{3^+} + e \longrightarrow Me^{2^+}$  couple are very close (within +0.1 v or below) to the reduction potentials for  $Me^{3^+} + 3e \longrightarrow Me$  couples, as shown in Refs. 19 and 20, and indicate that in this situation the extraction yield is low. The experiments with the actinide series summarized in Fig. 5 along with the experiment with macroamounts of Yb show the extraction yield probably is related to the presence of the. extracted element in the 2+ state, after addition of NaHg<sub>x</sub> to the " $\alpha$  mixture" solution and strong nascent hydrogen evolution.

The results with Md indicate the formation of Md<sup>2+</sup>, with a reduction potential of about -0.1 v<sup>(15,21)</sup> for the Md<sup>3+</sup> + e = Md<sup>2+</sup> couple; those with nobelium indicate the No<sup>3+</sup> + e = No<sup>2+</sup> couple has a reduction potential of about +1.2 v.<sup>(16)</sup> Fig. 6 shows that the californium numbers for Md (1.02) and No (0.74) are the same or higher than for Yb (0.748) and that these three elements probably form relatively stable Yb<sup>2+</sup>, Md<sup>2+</sup> and No<sup>2+</sup> ions. The californium numbers of Cf (1.0), Es (1.12) and Fm (1.73) are higher than the number for Eu (0.768) or Sm (0.895), which definitely form EuCl<sub>2</sub> and SmCl<sub>2</sub>. It seems very probable, therefore, that solid CfCl<sub>2</sub>, EsCl<sub>2</sub> and FmCl<sub>2</sub> as well as other divalent compounds of these elements can be prepared. Comparison of oxidation potentials and extraction yields in the lanthanide series (for Eu, Sm and Yb) and the actinide series (Md and No) suggests that the oxidation potentials of Cf, Es and Fm will be below the oxidation potential of the  $\text{Sm}^{3+} + e \longrightarrow \text{Sm}^{2+}$  couple, and that  $\text{CfCl}_2$ ,  $\text{EsCl}_2$  and  $\text{FmCl}_2$  will be more stable in aqueous solution than  $\text{SmCl}_2$ .

From the theoretical point of view, it will be very interesting to perform the extraction experiments with Lw, to be able to compare the complete actinide and lanthanide series. According to the Seaborg actinide hypothesis,<sup>(22)</sup> the end of the series occurs at Lw, and No<sup>2+</sup> is the actinide analog of Yb<sup>2+</sup>. Then Lw should be strongly trivalent and its extraction yield should drop substantially in relation to No. This experiment would be an interesting test of the Seaborg actinide theory.

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Extracted	211 <sub>Pb</sub>	223 <sub>Ra</sub>	222 <sub>Th</sub>	239 <sub>Pu</sub>	241 Am	244 <sub>Cm</sub>	252 <sub>Cf</sub>	253 <sub>Es</sub>	255 <sub>No</sub>	Extraction Time	Number of Runs
% Extracted				8.2	5.1	5.9	4.7	59	35	l min <sup>a</sup>	
Ratio to Cf				0.179	0.109	0.126	1.0	1.26	0.74		27
% Extracted	72.5	30.8	12	6.2	10.1		73.7	•		2 min <sup>a</sup>	
Ratio to Cf	1.01	0.424	0.134	0.072	0.12		1.0				4
% Extracted	85.5	46.2	7	16.5	11.0		96 <b>.</b> 5			4 min <sup>b</sup>	- -
Ratio to Cf	0.89	0.48	0.073	0.171	0.119		1.0				<b>Z</b>
% Extracted (14)		+	-	16.6	7.8	5.5	97.5			4 min	
Ratio to Cf <sup>(14)</sup>	(			0.170	0.08	0.056	1.0				2

Table 1. SODIUM AMALGAM EXTRACTION OF ACTINIDE ELEMENTS

<sup>e</sup>Flamed on plate <sup>b</sup> Electroplated. ដូ

	Element	This Work	Ref. 14	Mean value
<del></del>	Pb <sup>2+</sup>	1.72 (12 exp.)		1.72
	Ra <sup>2+</sup>	0.72 (10 exp.)	· .	0.72
	Ac <sup>3+</sup>	0.095 (12 exp.)		0.095
	Th <sup>4+</sup>	0.061 (12 exp.)		0.061
	Pa <sup>5+</sup>	0.013 (12 exp.)		0.013
	U <sup>4+</sup>	0.077 (12 exp.)		0.077
	Np <sup>4+</sup>	0.060 (12 exp.)		0.060
	Pu <sup>4+</sup>	0.122 (12 exp.)	0.102 (9 exp.)	0.113
	Am <sup>3+</sup>	0.094 (12 exp.)	0.062 (9 exp.)	0.080
	Cm <sup>3+</sup> .		0.050 (9 exp.)	0.050
•	Bk <sup>3+</sup>		0.018 (3 exp.)	0.018
	Cf <sup>3+</sup>	1.0 (12 exp.)	1.00 (9 exp.)	1.00
	Es <sup>3+</sup>	a 1.26 (2 exp.)	0.98 (2 exp.)	1.12
	$Fm^{3+}$		1.73 (2 exp.)	1.73
	Ma <sup>3+</sup>	(15) 1.02 (2 exp.)		1.02
	No <sup>2+</sup>	0.74 <sup>a</sup> (2 exp.)		0.74
	•		•	
	Yb	•	0.748 (6 exp.)	0.748
	ТЪ		0.0025 (2 exp.)	0.0025
	Dy		0.0006 (2 exp.)	0.0006

Table 2. THE "CALIFORNIUM NUMBERS" FOR Ra, Pb AND THE ACTINIDES

<sup>a</sup>Statistically 27 duplicate experiments with the small amount of nobelium used in each experiment is equivalent to two experiments with the other elements.

an La Carlor A Carlor a La	Ref. 10	Ref. 2	Ref. 7	Mean Californium Numbers
La <sup>3+</sup>	0.334	0.221		0.278
Ce <sup>3+</sup>	0.207	0.213		0.210
$Pr^{3+}$	0.207	0.101		0.154
Na <sup>3+</sup>	0.111	0.081		0.0%
Pm <sup>3+</sup>		·		
Sm <sup>3+</sup>	1.05	0.764		0.895
Eu <sup>3+</sup>	0.796	0.740		0.768
Ga <sup>3+</sup>		0.0016		0.0016
ть <sup>3+</sup>	<b></b>			0.0025
Dy <sup>3+</sup>		<b>~~~</b>		0.0006
<sub>Но</sub> 3+	· · · · · · · · · · · · · · · · · · ·	<b>.</b>	< 0.002	< 0.002
Er <sup>3+</sup>			< 0.002	< 0.002
Tm <sup>3+</sup>	÷ = =	· · · · · · · · · · · · · · · · · · ·	< 0.002	< 0.002
уъ <sup>3+</sup>	0.748	0.748	0.748	0.748
Iu <sup>3+</sup>			< 0.002	< 0.002

Table 3. THE "CALIFORNIUM NUMBERS"<sup>a</sup> FOR THE RARE EARTHS

<sup>a</sup>Californium numbers for extraction of lanthanides by sodium amalgam recalculated from extraction yield values. (11,2,7) The mean values for Yb, Tb and Dy are taken from Table 2.

#### FIGURE CAPTIONS

- Fig. 1.  $\alpha$  spectrum of " $\alpha$  Mixture." The energies (in MeV) define the peaks used for the determination of the yields of the elements U, Np, Pa, Pu, Am and Th in the extraction. For the Cf determination the 6.11 MeV peak and the SF peaks were used as a double check. For the Pb yield the 6.62 MeV peak of the short-lived daughter <sup>211</sup>Bi was used. The Ra yield was determined from <sup>219</sup>Rn and <sup>215</sup>Po peaks. The O points are valid for the left scale and the x points are valid for the right scale.
- Fig. 2.  $\alpha$  spectrum of the "No Mixture" used for the extraction of  $^{255}$ No. Curve 2a (O points) represents the  $\alpha$  spectrum before extraction. Curve 2b (x points) represents the  $\alpha$  spectrum after extraction. The range of  $^{255}$ No counts is between channels 145 and 166.
- Fig. 3. Extraction yields (in %) of the actinides, depending on the amount of HCl added before extraction (in lambda of 1 M HCl).
- Fig. 4. Californium numbers of the actinides, depending on the amount of HCl added before extraction (in lambda of 1 M HCl).
- Fig. 5. Extraction yields (in %) of the actinides in the presence of 230  $\lambda$  of 1 M HCl before the addition of 30% H<sub>2</sub>O<sub>2</sub> (7  $\lambda$  and 35  $\lambda$ ) or of 5 N HNO<sub>3</sub> (3  $\lambda$  and 15  $\lambda$ ).
- Fig. 6. Californium numbers of Pb, Ra and the actinides (O points) compared with the californium numbers of the lanthanides (x points).



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Fig. 1

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Fig. 2



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Fig. 3

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Fig. 4



# Fig. 5

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Fig. 6

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