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EUROPEAN ATOMIC ENERGY COMMUNITY — EURATOM

THE ISOLATION OF MICROGRAM AMOUNTS OF BERKELIUM AND CALIFORNIUM FROM IRRADIATED AMERICIUM

by

J. KOOI, J.M. GANDOLFO, N. WÄCHTER and J. WIJKSTRA
(Euratom)

and

R. BODEN, R. HECQ, E. VANHOOF and M. LEYNEN
(C.E.N.)

1965

Transplutonium Elements Program

Report prepared by CEN
Centre d'Etude de l'Energie Nucléaire, Mol — Belgium
Euratom Contracts No. 001-61-2 TPUB and 006-62-10 TPUB

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Summary

This report gives a detailed description of the processing of a slug containing 2.5 grams of highly irradiated ^{241}Am from which approximately 200 milligrams of curium and microgram-quantities of berkelium and californium were isolated. The processing included the first full scale application of a newly developed extraction chromatography procedure using HDEHP loaded columns.

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—— ^{244}Cm; _ .. . . _F.P. β-activity
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—— Cm-fraction; — — — Cf-fraction

Figure 10 b : Elution of column V. Purification of berkelium.
Column volume versus β and fission activity (in arbitrary units) of same samples as of figure 10a.
—— β -activity (corrected for α's)
— — — fission activity

INTRODUCTION

By courtesy of the United States Atomic Energy Commission the U.S. Transplutonium Program Committee made an important quantity of highly irradiated material available to the Euratom Transplutonium Elements Program for processing in Europe. This not only permitted for the first time European research workers in this field to isolate and manipulate weighable quantities of the elements berkelium and californium and of several hundreds of milligrams of curium prior to the obtention of material irradiated in European high flux reactors, it also enabled them to try out a recently developed separation method on higher than tracer level. The experience gained and the materials resulting from the processing are of essential importance for sustaining and expanding European research programs on heavy elements.

The irradiated material consisted of an aluminium slug UCRL 29-3, weighing approximately 50 grams and initially containing 2.5 grams of americiumoxide ($^{241}AmO_2$). It had been subjected to an integrated flux of $5.32 \cdot 10^{22}$ nvt over a 5 year period in the Materials Testing Reactor at Idaho Falls at a thermal flux of $4.5\text{-}5.0 \cdot 10^{14}$ n/cm^{2}sec. It was taken out of the reactor in May 1963. The total heat produced was found to be 0.724 watt on April 21st,1964 in a measurement performed at the Lawrence Radiation Laboratory at Livermore 1).

Apart from the bulk material (Al) and the heavy element products (estimated at 200-250 mgs), about 2 grams of fission products are constituted by a dozen of elements. Of these zirconium, molybdenum and ruthenium are the most important, together accounting for more than 50 percent by weight. In addition, between 40 and 50 ml of fission gases are present.

Before introduction into the chemical hot cell the activity of the slug was measured in the dismantling cell of the BR-2 reactor complex at Mol. It was found that the total γ-activity amounted to approximately 8 R at 1 m, while a smeartest of the slug gave 20 cps α-activity.

FACILITIES AND EQUIPMENT

FACILITIES

The facilities available for the processing consisted of a 1000 curie 1 Mev α-β-γ-n hot cell, an α-tight lead shielded cave and radiochemistry laboratory space with glove boxes.

1. A detailed description of the 1000 curie chemistry cell will be given elsewhere. In the schematic drawing given in figure 1, three different areas are indicated.

1.1. The isolation room is equipped with C.R.L. Model 8 extended reach manipulators, while the opening, foreseen for the lead-glass window, was closed by perspex plates, 1 inch thick and filled with tap water. Visibility through this water-window, providing more than sufficient shielding, was excellent due to the addition of copper sulphate and approximately 1 kg of EDTA per 1000 liter of water. Not the slightest turbidity was observed during a three month period. Sometimes it proved to be very convenient that the upper surface of the water acted as an ideal mirror for operations on floor level close to the window.
The isolation room served for the following purposes.

1.1.1. Unpacking of the Am-slug from the inner uranium container.

1.1.2. Measuring of neutron and γ-activity of the slug and of waste and product solutions from the working cell.

1.1.3. Packing of radioactive waste prior to disposal and introduction of equipment, process solutions, etc. into the working cell. In many cases α-active material was sealed into plastic bags by a manipulator handled sealing device. At no instance contamination of the isolation room occurred, nor of the interlock system, giving access to the working cell.

1.2. The α-tight working cell is equipped with an adjustable inner α-containment box of special design. This box can be easily introduced into the cell and is connected with the isolation room and an outside α-transfer box by means of specially designed interlock systems. These are provided with a sliding table, permitting transfers under α-tight conditions or even inert-gas atmosphere. For details, see reference 2. The cold transfer lock is of the lazy-susan type and is in particular used for introduction of small equipment and extraction of samples for assay. The working cell is equipped with a leadglass window and two C.R.L. Model 8 manipulators.

1.3. The inner containment box is directly connected with two stainless steel boxes, equipped with gloves and tongs and installed in the cellar. They provide for installation of additional gas adsorption towers and for emergency storage of radioactive solutions, especially in case of mechanical failures which require maintenance workers to enter the working cell. The two containers are accessible from the isolation room through manholes, which under normal conditions are hermetically sealed.

During the processing the radioactivity was fully contained within the containment box. After the treatment also no activity could be detected in the transfer locks. After retracting of the manipulators the upper half of the α-box was lowered and the box, wrapped in plastic, taken out for final decontamination.

2. The lead shielded cave is constructed* on the same principles as the 1000 curie cell. It provides 10 cm of lead shielding to an α-tight containment box, equipped with two Hobson M 7 manipulators and simple transfer systems. Radioactive waste solutions, samples and equipment may be transferred under α-tight conditions. After removal of the bulk of the radioactive material the front panel may be slided aside, giving direct access to the gloved containment box. For final decontamination the box may be disconnected and removed.

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* The cell was manufactured by Ets. Lemer, Nantes (France)

3. Commercially available glove boxes were provided with standard equipment for sample preparation and low level column separations.

EQUIPMENT

Concerning the processing equipment in general the designs of previous workers were adapted to our needs regarding size, connections and general facilities available. In addition, a deliberate effort was made to simplify the apparatus as much as possible in order to minimize the risks of failure or breakage. We profited largely of the experience of Dr. S.M. Fried, Lawrence Radiation Laboratory, who assisted in the design of equipment and the planning of the operations and who gave us in many instances invaluable advice during his stay at Mol. We also made a good use of the experience of one of the present authors (R. Hecq) obtained during his stay at Fontenay-aux-Roses where he actively participated in a napkin ring processing carried out by Dr. R. Berger 3).

A few additional remarks may be useful. First of all we found it quite convenient and satisfactory to heat the ion-exchange columns by passing current through a bare resistance wire, immersed in the water-filled outer jacket. It was found that, by just preventing boiling of the water fairly constant temperatures of 85-90°C could be realized.

Secondly, silicon gold plated surface barrier detectors *, situated close to the tip of the ion-exchange column greatly facilitated the detection of α-active drops coming off the column. In fact the formation, the growth and falling of the drops could be observed from an integrating indicator. Care should be taken to prevent contamination of the detectors. This may be realized by exchanging the thin plastic caps, which in any case have to be used to decrease the counting rates from the most concentrated drops of the Cm-solutions.

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* Our sincere thanks are due to Prof. Dr. G. Bertolini, Nuclear Chemistry Group of the C.C.R. Ispra (Euratom) for providing us with the mini-counter detector and electronics and a 4π proportional counter which rendered us many good services.

FLOWSHEET

In recent years a fairly large number of irradiated plutonium napkin rings, slugs containing irradiated americium and/or curium and similar samples have been processed by a number of groups, especially in the United States, in view of the isolation of the higher transplutonium elements. From their experience a flowsheet has resulted which has generally been adopted, usually with only minor modifications in order to take into account the characteristics of the starting material or the special purpose of the treatment.

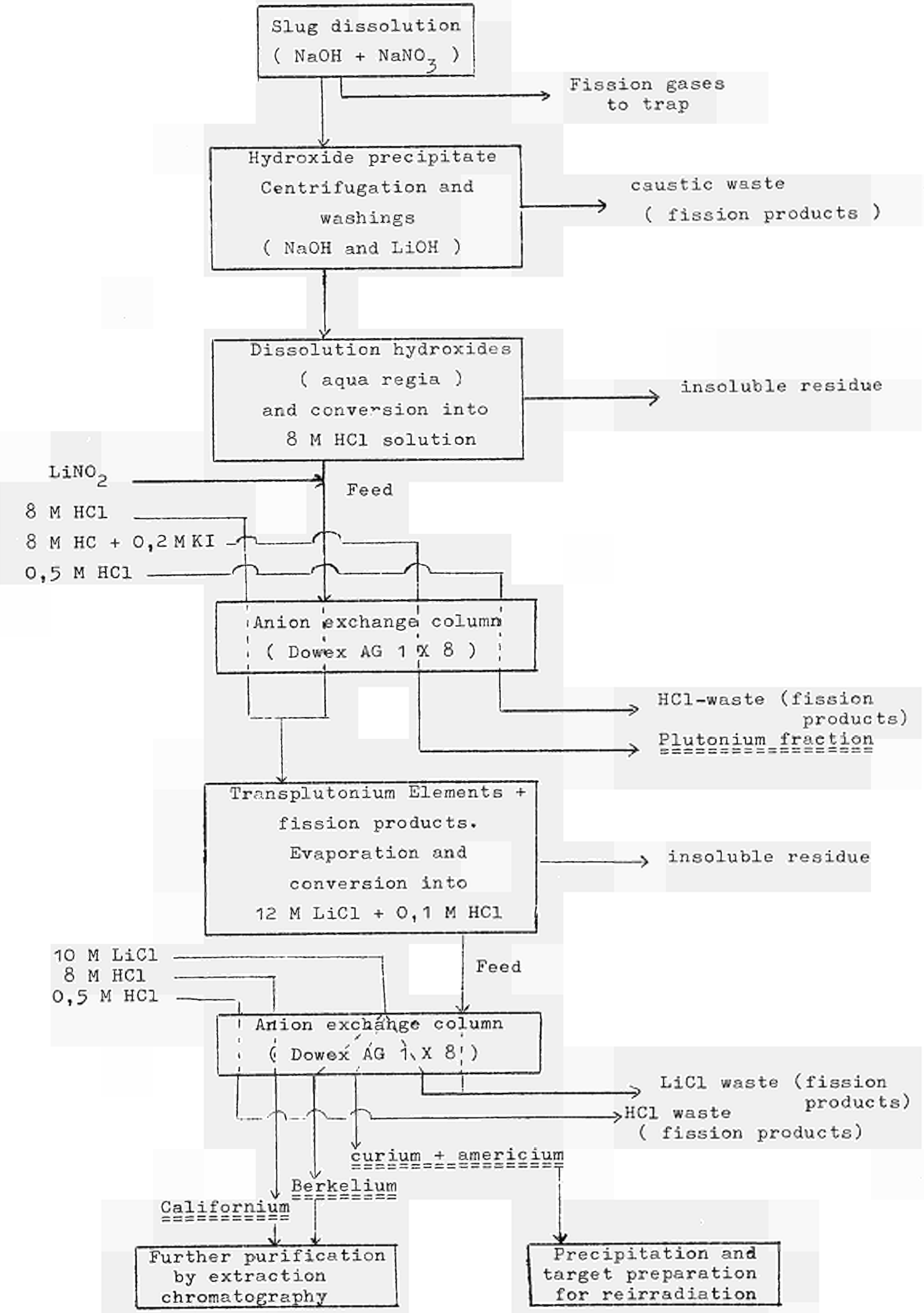
Also the processing carried out by Berger 3) and two plutonium napkin runs performed as test runs by our group, followed the same general pattern.

As the processing of slug UCRL-29-3 offered a first and up to now unique possibility to test an extraction chromatography method 4) for the separation in particular of berkelium and californium, which showed promise on a tracer scale, two major modifications were made in the flowsheet. Both concerned steps which are usually considered as being rather delicate and time consuming.

First of all, the generally employed cation-exchange procedure using α-hydroxyisobutyrate as an eluant for the separation of higher actinides was replaced by the extraction chromatography procedure. Secondly, its application also enabled to avoid a separate step for the isolation of the berkelium by solvent extraction after conversion into a HNO_3 solution and oxidation.

Although it is believed that for operations at this level of activity, extraction chromatography may partly or completely replace the usually applied anion exchange procedure with concentrated LiCl-solutions, the above mentioned separation meant already a scaling-up by a factor of 10^4 - 10^5 in the amount of e.g. californium. The scaling-up factor in respect to the level of irradiation was still much more important. It was therefore decided, to limit the modifications of the actual flowsheet to the two points mentioned above and to separately explore the potentialities of the extraction chromatography procedure and the behaviour of the extraction column under intense radiation.
Consequently table I represents the overall-flowsheet adapted.

TABLE I


Slug dissolution
(NaOH + $NaNO_3$)
Fission gases to trap
Hydroxide precipitate
Centrifugation and washings
(NaOH and LiOH)
caustic waste
(fission products)
Dissolution hydroxides
(aqua regia)
and conversion into
8 M HCl solution
insoluble residue
$LiNO_2$
Feed
8 M HCl
8 M HC + 0,2 M KI
0,5 M HCl
Anion exchange column
(Dowex AG 1 X 8)
HCl-waste (fission products)
Plutonium fraction
Transplutonium Elements + fission products.
Evaporation and conversion into
12 M LiCl + 0,1 M HCl
insoluble residue
10 M LiCl
8 M HCl
0,5 M HCl
Feed
Anion exchange column
(Dowex AG 1 X 8)
LiCl waste (fission products)
HCl waste
(fission products)
curium + americium
Berkelium
Californium
Further purification by extraction chromatography
Precipitation and target preparation for reirradiation

DISSOLUTION AND COLUMN FEED PREPARATION

The dissolution of the americium slug was carried out in a cylindrical vessel of about one liter with a coneshaped bottom and provided with a reflux condensor. The off-gases were, by means of a slight underpressure (- 5 cm Hg), passed through a glass frit, acting as a water trap and through two towers, one filled with molecular sieves, the other with active charcoal, in order to eliminate remaining water vapor, NH_3 and fission gases. After passing two additional charcoal towers installed in the basement box the gases were vented into the active ventilation duct. All absorption towers were degassed at 200°C before use.

The actual dissolution took place in two steps, which yielded some unforeseen data. To start with a dissolver solution was used having the following composition : 5 M NaOH, 2 M $NaNO_3$ (for converting the hydrogen evolved into NH_3) and 1 M $NaNO_2$ (to provide NO_2^- -ions). Cold runs on non irradiated aluminium had given quite satisfactory results. About 25 minutes after turning on the heating coil of the waterbath a violent reaction started which was completed in 15 minutes. Only very little water was collected in the watertrap while 33 liters of gas passed the absorption towers. After disconnecting the absorption towers the solution was allowed to cool. Later it was observed that these two towers together, measured at the outer surface, gave about 1 R/h γ-activity. After cooling the rather viscous black solution was centrifuged and the precipitates washed twice with a small volume of 1 M NaOH-solution and once with 0,5 M LiOH.

Just before transferring, the last fraction of the solution into the centrifuge cone it was observed that two lumps of metal, together representing an appreciable part of the slug had not gone into solution.

These two pieces covered by a thick black crust, were subsequently successfully dissolved in a solution, 7 M in NaOH and 3 M in $NaNO_3$. In this case it took 50 minutes, from the moment the heat was turned on, to initiate the reaction. Total reaction time again amounted to roughly 15 minutes. 5 liters of gas were evolved and absorbed on the charcoal towers in the basement box. The activity of the first one, measured at the outer surface, increased from 2.3 mr/h to 11 mr/h, that of the second one remained unchanged. ($\sim$1.8 mr/h)

The resulting solution was centrifuged and the dissolver vessel and precipitates carefully washed with 1 M NaOH (twice) and 0,5 M LiOH.

Assays were made on the two supernatants and the combined washings. (See table II).

It may be concluded that only a small fraction of the α-activity was liberated in the first dissolution operation, together with essentially all of the fission gases. The supernatant of the second dissolution (of the core of the slug) contained ~0.5 % of the total α-activity, that of the first dissolution and the combined washings each contained less than 0,005 % of the total α-activity. γ-spectroscopy on samples of the two supernatants showed the γ-activity to be due to ^{137}Cs. A small percentage of ^{134}Cs was also detected.

After centrifuging and washing aqua regia was added to the black precipitates. Very little foaming occurred. After transferring the solutions to other tubes the aqüa regia treatment was twice repeated.

The black crud left behind showed considerably β/γ-activity but zero neutron count ($\leq 3.10^{-4}$ of the counting rate of the initial slug). Later spectrographic analysis* showed it to consist mainly of Ca,Fe,Mg, other components being Al,Ba,B,Si,Mo,Pd,Y,Ru,Sr with traces of Mn and Ag.

The combined aqua regia solutions were evaporated almost to dryness and taken up in 8 M HCl. After a final centrifugation a total volume of about 33 ml resulted, 100λ of which were set aside for later analysis (sample S 3). The residue left behind by this final centrifugation was still fairly β/γ-active but no neutrons were observed. Later spectrographic analysis revealed the presence of Ba and Sr as main components, other elements present being Fe,Si,Ca, Mg,Ag,Na, Pd,Ru with traces of B,Al,Mo,Mn,Y.

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* Our sincere thanks are due to the Analytical Chemistry Group of the C.E.N. under the direction of Mr. F. LIEVENS for carrying out these analyses.

TABLE II

Sample	neutron activity	total α-activity	% of Sum total α-activity
Supernatant 1st dissolution	—	$3.4 \cdot 10^{7}$ dps	0,006 %
Supernatant 2nd dissolution	0 ($\leq 3.10^{-2}$ %)	$3 \cdot 10^{9}$ dps	∼0,5 %
Combined washings NaOH + LiOH	—	$3.3 \cdot 10^{7}$ dps	0,006 %
HCl solution column feed	n.d.	$5\text{-}6 \cdot 10^{11}$ dps	99.5 %

Material balance of dissolution step expressed in total α-activity

Table II summarizes some of the measurements on samples taken at various points of the dissolution procedure. All samples were taken by remotely pipetting 10 or 25 λ (or occasionnally 5λ) from the process solutions. Total volumes were approximately read from the graduated centrifuge tubes. Care was taken to ensure that samples were representative and volumes correctly adjusted. No duplicate samples were made, except in one case in which a deviation of <2 % was found. The micropipettes were emptied into bottles containing 20 ml of a 1 M HCl solution which were taken to the laboratory for sample preparation. All samples obtained during the run were counted in one and the same set of counters.

Successive dilutions were made to reduce or eliminate self absorption losses and anticoincidence corrections.

In the case of α-samples and applying an average counting efficiency the cpm countings have been converted into dps to indicate roughly the actual quantities obtained, mainly for reasons of comparison and for roughly checking the material balance. throughout the procedure. These data must be considered to be approximative, since in the course of the operations and due to the large number of assays made, it was generally not possible to prepare the samples according to standards required for absolute counting.

Finally fission countings were performed on the same samples as used for α-measurements in a 2π proportional flow counter yielding a first and very preliminary indication of the total activity of ^{252}Cf to be expected : $\sim 9,4 \cdot 10^{6}$ fps.

PLUTONIUM SEPARATION
HCl - COLUMN

Although only a few milligrams of plutonium might be expected to be contained in the slug it was nevertheless decided to run a so called HCl column. First, plutonium consisting mainly of the isotopes 238 and 240 is still rather scarce and may be advantageously used in a number of cases e.g. in combination with other isotopic mixtures. Second, a major fraction of fission products, in particular Zr and Ru in their IV-state could be eliminated by adsorption on the columns.

The column (inner diameter ~12 mm) was filled to a height of ~8 cm with Dowex AG 1 - X 8 resin, 300-400 mesh, from 8 M HCl solution and rinsed, prior to use, also with 8 M HCl. Dropsize as measured without pressure was found to be 185 λ. During operation of the column the outer water jacket was heated to just below uncipient boiling at the heating coil. Pressure was applied and adjusted from the outside of the hot cell. During the run, which lasted for 27 hours, pressures varied from zero to 20 cm of mercury, elution rates from 8 seconds to ~40" (with an exceptional short period of upto ~100 seconds) depending on eluting elution, gas evolution and other difficultly adjustable factors. A temporary blocking of the columns, presumably due to gas evolution, could be easily overcome by exerting pressure on the upper part of the resinbed.

After loading the column with the solutions prepared as described in the preceding chapter, and to which several hundreds of milligrams of $LiNO_2$ had been added, elution was carried out with a preheated solution of 8 M HCl. Fractions of 20-30 ml were collected in graduated centrifuge tubes and assayed for α, γ and fission counts. The two counters, installed close to the tip of the column fairly clearly indicated the appearance of α-active eluate drops and their growth and falling. However, no valid quantitative indication of the concentration of the droplets was obtained. Also radiolysis occasionnally caused sputtering of the droplets, leading to an appreciable background count from the protective covering foil.

After essentially all α-activity had been collected, the elution of the plutonium fraction was effected by passing a solution of 8 M HCl to which 0,2 M of KI had been added. Finally a solution of 0,5 M HCl was passed over the column, mainly in order to verify the nature of the fission products adsorbed.

A summary of the results of this column operation is given in Table III and graphically shown in Figure 2.

TABLE III

Eluting Agent	Tube N°	Volume in ml	α-activity		γ-activity		fission activity	
			10^{10} dps 1)	in %	10^{9} cpm 2)	in %	10^{6} fps 3)	in %
8 M HCl	→ 1	29	42.43	72.14	14.0	24.0	8.01	73.5
	2	28	14.25	24.25	11.1	19.1	2.54	23.3
	3	28	1.57	2.67	7.2	12.3	0.34	3.1
	4	28	0.05	0.09	2.8	4.8	(0.02)	(0.05)
	5	29	0.03	0.05	0.9	1.5	(0.01)	(0.03)
8 M HCl + 0,2 M KI	→ 6	31	0.03	0.05	1.3	2.2	(0.01)	(0.03)
	7	29	0.01	0.02	0.2	0.3	-	
	8	18	0.01	0.02	0.0(5)	0.1	-	
0.5 M HCl	→ 9	19	0.32	0.54	0.8	1.4	-	
	10	26	0.01	0.02	1.4	2.4	-	
	11	28	0.02	0.03	1.5	2.6	-	
	12	25	0.01	0.02	0.5	0.9	-	
Dry column after elution 4)					16.6	28.5		
Total			58.74		58.3		10.9 3	

1) See page 10 , 2nd paragraph

2) γ-activity reported as measured in a well type NaI(Tl) counter

3) fission activity in fissions per second as measured by fission fragment counting in 2π proportional flow counter

4) Measured by comparing readings obtained from combined fractions 10-12 and from columns by means of GM counter installed at the outside of the perspex window of inner α-box.

TABLE III - HCl column elution data

As indicated by arrows in table III successive eluting agents were introduced into the top of the column while changing collecting tubes, after assaying the previous one. This means that tubes 6 and 9 still contained about one column volume of the preceding eluant.

The contents of tubes 1, 2 and 3 were combined for further treatment as transplutonium fraction. It contains 99.9 % of the total fission activity and 99.1 % of all α-activity but only about half of the fission product γ-activity. A considerable fraction thereof remained on the column, the activity measured indicating only a lower limit (due to geometry- and absorption factors). The contents of tubes 4 and 5 were combined and checked for neutrons. At contact a zero neutroncount was obtained. Fractions 6 to 9 were combined and later treated for recuperating the plutonium. Fractions 10-12 were also combined and later, together with the column, stored as waste, after they were checked for the absence of neutron-emitters.

At the end of the elution the anion exchange column looked dark brown to black. We have, however, no indication that this blackening by irradiation had any influence on its performance.

α-spectra* taken from samples of the solution obtained on dissolving the americium slug had given only faint indications of the presence of californium. Spectra obtained from a sample of fraction 1 very clearly revealed the ^{252}Cf - α rays. No evidence for plutonium-238 was obtained. Samples of tube 6, on the other hand, gave a spectrum indicating, besides ^{244}Cm, in particular ^{238}Pu and ^{239,240}Pu. No energies corresponding with ^{252}Cf were detected. A γ-spectrum of this sample revealed ^{106}Ru/^{106}Rh. The 3.53 Mev β-ray of ^{106}Rh was also identified.

Finally an α-spectrum was taken of tube 9. The only isotope unambiguously identified was ^{244}Cm, although the presence of a trace of ^{238}Pu could not be excluded. No indications for ^{252}Cf were obtained.

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* Our thanks are due to Mr. J. Colard (Physics Department of the C.E.N.) and his collaborators for carrying out an appreciable part of the spectrometric measurements. We are also indebted to Mr. P. Beullens (L.M.A.) for his assistance in taking a number of spectra.

From this the conclusion seems to be justified that the plutonium indeed was eluted in a few column volumes after the addition of KI and that essentially all of the curium-"tail", still remaining on the column, was stripped with the first few column volumes of 0.5 M HCl.

The total activity data, both for α's and fissions eluted from the column correspond very well with the figures obtained during the dissolution step. Total α-activity was found to amount to $5.9 \cdot 10^{11}$ dps, total fission activity to $10.9 \cdot 10^{6}$ fps as compared to $9.4 \cdot 10^{6}$ fps found before. As regards the absolute values of these quantities the same comments apply as those made on page 10 , 2nd paragraph.

LITHIUM CHLORIDE COLUMNS

As explained before a group separation between the lanthanide fission products and the actinides was effected by anion exchange in lithium chloride medium, following the procedure of Hulet, Gutmacher and Coops5). The combined fission and α-active fractions from the HCl-column were converted into a LiCl-solution by evaporation to a volume of about 6 ml, addition of a known volume of a solution containing 12 M LiCl and 0,1 M HCl and concentration to a volume equal to that of the added LiCl-solution. Prior to use the LiCl had been purified by passing it over an anion exchange column.

Following a remark made by Cunningham 6) on the capacity of the anion exchange columns used in this procedure and in view of the fact that we were unfamiliar with LiCl-column operations with large amounts of radioactive materials it was decided to run two columns, each with approximately half the amount of material.

The resin used was Dowex AG 1 - X 8, 300 - 400 mesh. Column dimensions were 12 mm diameter and 14 cm and 13,5 cm height resp. Prior to use the columns were equilibrated with 12 M LiCl- 0,1 M HCl solution. The water jacket was heated by an immersed heating coil to 85 - 90° C. Pressure was applied from the outside, the elution rate varying from 10 to 40 seconds per drop depending on viscosity of the eluant, gas evolution etc.

Both columns performed very smoothly, the main difference being that in the first one essentially all gas evolved escaped through the top of the column in a very steady manner, while in the second run the gas was carried along with the eluting solution and escaped through the tip of the column. In the second case the maximum intensity of gas escape was even indicative for the elution peaks.

The first column run, in which 417 ml were eluted took 35 hours, the second one 28 hours for 386 ml. The corresponding elution rates were 12 and 14 ml per hour resp., or 0,18 and 0,21 ml/cm^2 per minute.

The course of the loading of the column (from 12 M LiCl) and its subsequent elution with 10 M LiCl, 8 M HCl and 0,5 M HCl resp. as well as the elution data are given in tables IV-a and IV-b and graphically presented in figures 3a and 3b.

TABLE IV-a

Eluting Agent	Tube Number	Volume in ml	α-activity		γ-activity		fission activity	
			10^9 dps	%	10^8 cps	%	10^4 fps	%
	101	20	0.00(2)	-	3.8	3.6	0	-
10 M LiCl →	102	12.5	0.02	0.01	8.2	7.9	3.7	0.8
	103	9	1.12	0.48	39.2	37.7	1.20	0.3
	104	25	0.08	0.03	25.8	24.8	0	-
	105	21	20.72	8.87	0.5	0.5	0	-
	106	14	62.30	26.66	0.8	0.8	(8.4)	(1.8)
	107	12	106.81	45.71	1.3	1.2	10.4	2.2
	108	17	30.40	13.01	0.5	0.5	(7.9)	(1.7)
	109	15	6.66	2.85	0.1	0.1	(2.0)	(0.4)
	110	17	2.86	1.23	0.2	0.2	0	-
	111	18	0.91	0.39	0.1	0.1	0	-
	112	17	0.42	0.18	0.0(5)	-	0	-
	113	19	0.29	0.12	0.4	0.4	0	-
8 M HCl →	114	18	0.87	0.37	0.4	0.4	424.6	92.14
	115	22.5	0.01	-	0.4	0.4	((0.7))	((0.1))
	116	24.5	0.00(5)	-	0.2	0.2	((0.2))	-
	117	27	0.02	0.01	0.3	0.3	((0.4))	((0.1))
	118	19	0.00(8)	-	0.2	0.2	((0.1))	-
0.5 M HCl →	119	8	0.01	-	3.8	3.6	0	-
	120	22	0.06	0.03	1.6	1.5	((0.8))	((0.1))
	121	27	0.01	-	7.0	6.7	0	-
	122	23	0.02	0.01	6.8	6.2	0	-
	123	9	0.01	-	3.1	3.0	0	-
Total		417	233.62		104.7		460.4	

TABLE IV-a - First LiCl column

To all values indicated the same remarks apply as to those of table II. Arrows indicate addition of eluting agents to top of column. Values within brackets ( ) are uncertain, those within double brackets (( )) are very uncertain, due to low or very low counting rates.

TABLE IV-b

Eluting Agent	Tube Number	Volume in ml	α-activity		γ-activity		fission-activity	
			10^9 dps	%	10^8 cps	%	10^4 fps	%
	201	21.5	0.01	-	8.2	4.3	0	-
10 M LiCl →	202	21.5	0.04	0.01	47.2	24.8	(0.6)	(0.1)
	203	15	0.00(5)	-	29.6	15.5	0	-
	204	21	0.01	-	24,5	12.8	0	-
	205	25	0.21	0.06	10.5	5.5	0	-
	206	21	217.46	64.91	46.0	24.2	19.0	3.0
	207	18	88.42	26.40	17.8	9.3	10.0	1.5
	208	18	23.22	6.93	0.5	0.3	(2.8)	(0.4)
	209	16	3.68	1.10	2.0	1.0	(0.7)	(0.1)
	210	15	0.77	0.23	0.1	-	((0.1))	-
	211	19	0.34	0.10	0.1	-	0	-
	212	16	0.08	0.02	0.1	-	0	-
8 M HCl →								
	213	17	0.59	0.18	0.6	0.3	604.0	94.7
	214	22	0.00(2)	-	0	-	0	-
	215	23	0.02	-	0.6	0.3	0	-
	216	21	0.00(5)	-	0.4	0.2	0	-
0.5 M HCl →								
	217	23.5	0.04	0.01	1.2	0.6	0	-
	218	23	0	-	0.7	0.4	0	-
	219	29.5	0	-	0.9	0.5	0	-
Total		386	334.90		191.0		637.2	

TABLE IV-b Second LiCl column

To all values indicated the same remarks apply as to those of table II. Arrows indicate addition of eluting agents to top of column. Values within brackets ( ) are uncertain, those within double brackets (( )) are very uncertain, due to low or very low counting rates.

During the run the subsequent elution of the main lanthanide fraction and of the curium + americium fraction and the remaining fission products could be clearly observed by their caracteristic glow. Especially during the second run it was noted that a green bluish zone (as observed through the window) preceded the bulk of the loading solution, followed by well separated zones in different shades of brown and black brown. In both runs a slight bluish-green precipitate was observed in the second fraction collected, which later dissolved very easily on the addition of a few drops of 0.5 M HCl.

Also in the tubes in which the bulk of the curium was collected a white our greenish precipitate appeared on standing. In one instance, the supernatant was assayed and found to contain only $\sim$1 % of the total α-activity. These precipitates dissolved instantaneously in 0.5 M HCl.

From the data of tables IV it is apparent that the second column, operated more smoothly than the first one although in both cases all californium, representing about 95 % of the fission activity, was isolated in a small volume in one tube only. The data obtained during the first run permitted to fairly accurately program the second one. The second column had a much less variable elution rate, which therefore could also be better controlled. A smaller fraction of the initial load of α-activities passed through with the first few column volumes, which contain the bulk of the γ-activity. In the second run it was possible to "cut" the different fractions in a more precise way with less tailing occuring.

In the second run, however, relatively more γ-activity was present. For both α's and fissions the ratio of the loadings of second and first columns being 1.4 a ratio of γ-loadings of 1.8 was found. The additional γ-active fission products were found back in the main α-active fractions. We have reason to believe that this activity was mainly due to cerium and europium, dissolved in a second treatment of the evaporated α-fraction of the HCl column with 12 M LiCl - 0.1 M HCl. The insoluble residu was later spectrographically analysed. The main component was aluminium, other elements present being Ca, Mg, B, Sr and Si while traces of Fe, Ag, Na and Y were also found.

On a number of samples α-spectroscopy was performed. Only in samples of tubes 114 and 213 strong α-peaks corresponding with ^{252}Cf and ^{250}Cf were detected, accompanied by the ^{244}Cm-peak. In all other cases only curium-244 with a ~1 % curium-242 content was registered. In the sample of tube 114 (Cf-fraction) a γ-spectrum revealed the presence of ^{106}Ru and may be of ^{144}Ce. ^{106}Ru was also found to be the main γ-active constituent in tubes 110,111 and 112, a trace of ^{137}Cs being found in tube 110.

At the end of the run considerably γ-activities were still fixed on the columns, giving readings of the order of 30 R/h or more at the outer Gm-counter. Both columns were only slightly discolored.

It may be pointed out that the total activities recovered from both columns, amounting to $5.7 \; 10^{11}$ dps α's and to $11 \cdot 10^{6}$ dps fissions agree very well with the values found previously. The α to fission ratio observed in the main α-fraction (Cm+Am) corresponds within experimental error with the value from literature.

Finally the contents of different tubes were combined to fractions for further treatment as described in the following chapters.

EXTRACTION CHROMATOGRAPHY SEPARATIONS

GENERAL

Recently an extraction chromatography procedure was worked out[4)] with tracer quantities of transplutonium elements. This method exists in the successive elution of these elements (in their trivalent state) with dilute hydrochloric acid from Kieselguhr columns loaded with di-(2ethylhexyl)orthophosphoric acid (HDEHP).

The use of oxidizing and/or reducing agents is not required, while loading of the column is achieved by simply passing a solution 0,1 M in HCl over the column.

More in particular, it was found that excellent separations could be achieved between americium + curium, berkelium and californium while it was also demonstrated that cerium could quite easily be separated from berkelium. As in our case the combined americium + curium fraction was destined for reirradiation in the reactor, the application of this method to the separation of weighable quantities of transplutonium elements seemed indicated. From a chemical point of view we were especially interested in the behaviour of several fission products which were not extensively tested in prior experiments and the purity of the transplutonium element fractions separated from much more complex and more concentrated starting solutions. We also wanted to verify the elimination of relatively important quantities of LiCl under actual operating conditions.

From an operating point of view, our main interest concerned the radiation stability of the columns and a proof that columns of this type can be easily and reproducibly manipulated under actual hot run conditions.

Three types of extraction chromatography experiments were carried out. As the generally used extraction method for checking the presence of berkelium in the different LiCl-columns fractions gave no unambiguous results where applied to small aliquots of our solutions, some columns were run with a view to analyse the fractions.

Secondly, a first scaling up was effected by carrying out separations on the material obtained from some intermediate fractions, after concentration and removal of the bulk of the LiCl by an iron precipitation, followed by an iron column.

Third, the californium fractionsas obtained from the two LiCl-columns were treated.

Final purification and concentration were also effected by extraction chromatography columns.

EXPERIMENTAL

Introductory

All separations were performed on a total of three columns of 4 mm diameter and 12-14 cm height and one column of 7 mm diameter and 10 cm height. Two additional experiments were carried out with columns of 2 mm diameter and 5 cm height and of 1 mm diameter and 3,5 cm height, respectively.

All columns were prepared following the procedure given in reference 4). The column volumes were determined by passing through a solution of curium in 6 M HCl. Typical elution curves obtained during these determinations are shown in fig. 4.

Prior to use the columns were conditioned by passing a few column volumes of 0,1 M HCl. All columns were operated at a temperature of 80-90° C as in the case of the LiCl-columns and by applying a slight overpressure, regulated from outside the lead cave. One of the major advantages of these columns - from an operating point of view - is that the drop rate (negligible without pressure) could be very easily regulated and maintained constant. The elution could be stopped and (re)started at will and the fractions were collected rather by checking time than by directly counting drops or measuring volumes. It did not prove necessary to run the columns continuously, so that even normal working hours could be followed. Also prolonged interruption (overnight) with the highest loads used did not produce any disturbing effects. Similarly cooling to room temperature (e.g. overnight) and reheating after prolonged interruptions had no influence on column performance.

The experimental data on all runs are presented in table VI in chronological order. In all cases loading of the column took place from 0,1 M HCl solutions. The concentration of the eluting solution was chosen with respect to the load and the separation wanted.
In a number of cases analysis of the fractions collected was carried out during the run, but in most instances the runs were programmed in advance.

Of course the ultimate purifications were usually carried out by elutions with lower concentrations of HCl, while first separations were often limited to the removal of fission products and curium from the combined berkelium and californium. In this case the last two elements were stripped off the column with a few column volumes of 2 M HCl and passed over a second column. Also in the Bk-Cf separation californium was always stripped with 2 M HCl. Usually any remaining activity was stripped off the column with 6 M HCl before reconditioning it with 0,1 M HCl.

The fractions collected from the two LiCl-columns were combined together with the washings of the tubes and labelled as indicated in table V

TABLE V

Fraction	1st LiCl column tubes	2nd LiCl column tubes
Fission Products I	101,104	201,203,204
Fission Products II(x)	102,103	202,205
Curium I	105,106,107,108	206,207,208
Curium II	109,110	209,210
Intermediate	111,112,113	211,212
Californium	114	213
Fission Products III	115-123	214-219

Fractions collected from LiCl-columns

The transplutonium elements contained in a number of LiCl-solutions were isolated by co-precipitating them with 0,2 mg of added Fe^{+++} per ml with 10 M NaOH. The mother liquids of these precipitates contained always $<$0,5 % of the total activity present. After combining them a second precipitation was carried out. After centrifugation and washing the precipitates were dissolved in a small volume of 8 M HCl and the resulting solution passed over a pretreated anion exchange column (Dowex AG1-X8). The iron being retained by the column, the solutions were evaporated to dryness and the residues taken up in 0,1 M HCl for feeding the extraction columns.

Results

Table VI presents the results of the different column runs, in the same order as they were actually carried out. Figures 5 to 10 show experimentally determined elution curves of a number of runs, representative for the elutions carried out and separations achieved. In the figures arrows indicate when the successive eluting solutions were added to the top of the column. All activities are expressed in relative values. In the table are indicated the column feed, the eluting solutions used and the fractions isolated. From this table the actual separation flow sheet can be closely followed. The elution velocity was usually 13-15 minutes per column volume. The eluate was collected in portions of several column volumes, in graduated tubes, usually following a programme established on the basis of prior experience and the known position of the elution peak of the different elements as a function of the molarity of the eluting solution. The contents of the different tubes were combined to fractions according to the countings (α, β, γ, fissions, neutrons) obtained on samples of the tubes. The figures give representative examples. Additional details on the runs are given below, together with pertinent analytical data.

TABLE VI

Column run	Column diameter	load	successive eluting solutions (molar HCl)	Fractions isolated
A	4	100 λ intermediate fractions table V	0.3;2	Cm - Int.-strip
B	4	conc. intermediate fractions table V	0.3;2	Cm-Int.-strip
C	4	conc. curium II 2nd LiCl col.	0.3;2	Cm-Int.-strip
D	4	strip col. B(+A)	0.5	Cm-Bk-F.P.-Cf
E	4	conc. curium II 1st LiCl col.	0.3;2;6	Cm-Int.-strip
F	4	strip.col. C+E	0.5;2	Cm-Int.-FP I- Cf-FP II
G	4	100 λ curium I fractions 1st and 2nd LiCl col.	0.3;2;6	Cm-Int.-strip
H	4	25 λ Cf-fractions 1st and 2nd LiCl col.	0.5;2	Cm-Bk-F.P.-strip
I	4	strip col. G	0.5;2	samples only
K	4	Bk fr. col. H	0.3;1	samples only
L	4	conc. mother liquids Fe col.'s Int. fr.'s col A,B,C,E,G.	0.1;0.3;2;6	Cm-Int.-strip
M	7	Cf-fractions col. D,F,H	0.5;2	F.P. - Cf
N	4	Cm-fractions col.A,B,C,D,L	0.1;0.2;2	F.P. - Cm
O	7	part Cf-fraction 2nd LiCl col.	0.1;0.3;0.5;2	Cm-Bk-Cf-Cf(2)
P	4	Cm-fraction col. E	0.1;0.5;2;6	Cm-F.P.
Q	7	Cf-fraction col. O	0.1;0.3;2	Cf-F.P.
R	7	part Cf-fraction 2nd LiCl col. + Cf(2) fr. col. O	0.1;0.3;2	F.P.-Cm-Bk-Cf
T	7	Cf-fraction 1st LiCl col.	0.1;0.3;2	F.P.-Cm-Bk-Cf
V	4	Bk fractions col.'s O,R,T,D,F	0.3; 2	Cm-F.P.-Bk-Cf
Z	2	Bk fraction col. V	0.3;2	Cm-Bk
AA	4	Cf fraction col. V	0.5;2	F.P.-Cf
BB	4	Cf fraction col AA	0.3;2	Cm-Bk-F.P.-Cf
CC	4	Cf fraction col. BB	0.3;2	Cm(trace)-F.P. (trace)-Cf
DD	1	Cf fraction col CC	6	Cf sample

Abbreviations used:
Int. : intermediate fraction
conc. : concentrate of total fractions table V after precipitation with iron
col. : column
F.P. : fission product fraction
fr. : fraction

Columns A and B Well defined curium fractions were obtained. Countings on the strip solution indicated the presence of traces of Bk and californium (total $< 10^4$ fps). A sample taken from the beginning of the strip peak indicated the presence of ^{154}Eu and ^{155}Eu

Column C. Fission count of curium fraction corresponding to halflife. No fission count in strip.

Column D A sample of the (intermediate) Bk-fraction gave a β-spectrum identical to that of a Bk-sample originating from the decay of pure ^{253}Es. Its activity, however, was too low to permit accurate α-growth measurements. A F.P.-fraction sample contained europium isotopes 154 and 155.

Column E Same remarks as for column C

Column F Figure 5 presents the results of this run.

Column G No fission or neutron counts were obtained from the strip samples. No indications were found for the presence of berkelium.

Column H A definite Bk-peak was observed, a sample of which gave the same spectrum as that of the pure ^{249}Bk-sample (see column D)

Column I A trace of curium eluted in the proper position as a very sharp peak. The strip gave no fission count. No Bk peak was observed, the elution of a small amount of europium activity was observed in a position intermediate between those of the Bk- and Cf peaks. The data of columns I and G showed that $< 10^{-3}\,\mu g$ of Bk was present in the total intermediate fractions of the two LiCl-columns.

Column K The elution curves of this run are presented in figure 6. In the first two column volumes the Ru106-Rh106 couple was detected. Its elution peak overlapped the curium peak. In the curium fraction also ^{144}Ce (and probably ^{141}Ce) were observed. In the strip no α's were found.

Column L During this run the first indications were obtained that a major fraction of fission products could be separated from the curium by eluting with 0,1 M HCl.

Column M The combined Cf-fractions of columns D,F and H were used to test the 7 mm column. No curium was detected. The α and fission peaks coincided completely. The measured β-activity which is mainly due to α's registered in the thin window GM-tube showing a peak coinciding with the α and fission peaks, had a shoulder in front of the peak maximum. γ-measurements revealed that these β's were due to europium isotopes, eluted (with 0.5 M HCl) at a peak maximum position 1 - 1.5 column volumes before that of californium. (see fig. 7)

Column N This experiment was especially carried out to verify the results of column L. After passing 3 column volumes of 0.1 M HCl the elution was continued for 15 column volumes with 0.2 M HCl, followed by a 2 M strip. Column volumes 1-5.5, with a sharp peak maximum at 2 column volumes, constitute a fission product fraction, consisting of Ru106/Rh106. See also column P. Curium started coming off at column volume 6, its elution being completed at column volume 18.

Column O Since this experiment constituted the first run with non-tracer quantities of californium only part of the Cf-fraction of the second LiCl-column was used. Separation of the LiCl still present was achieved by elution with 0,1 M HCl. The fission products present in this fraction of eluate were again found to be ^{106}Ru and ^{106}Rh. Neutron counting of the different collecting tubes confirmed the fission countings. α-Spectrometry measurements indicated a ^{244}Cm/^{242}Cm ratio of 100/0.6 and a ^{252}Cf/^{250}Cf ratio of 100/7-10 as found before. No γ-emitters were detected in the Cm-fraction.

Complete elution data are presented in figures 8^{a-d}.

Column P The results of this run are presented in figure 9. They confirm those obtained in run N

Column Q The Cf-fraction from column O was again passed over the column to verify the purification achieved. Only a very minor amount of fission products was separately collected.

Columns R and T These runs could be programmed on the basis of the results from columns O and Q. As in run Q the elution with 0,5 M HCl could be omitted. Results are presented in figures 8.

Column V The combined berkelium fractions of previous runs were subjected to further purification on a 4 mm column. The elution curves are given in figure 10. Samples of the Bk fraction of this column were measured for ingrowth of α-activity which followed the curve calculated from the halflives of ^{249}Bk and ^{249}Cf.

Column Z In particular in view of testing the performance of a very small column the Bk was subjected to a final purification on a 2 mm X 5 cm column (column volume 5 drops) A trace of Cm could still be eliminated in a peak, two column volumes wide at its base. The berkelium was thereafter stripped in 2-3 column volumes.

Columns AA and BB These two columns were run in order to further purify the californium from the berkelium purification column. The ratio of activities at peak maximum for α's of curium and californium resp. was $<$ 1/100. Also a complete separation of traces of europium from the californium was achieved.

Column CC This smallest column (1 mm diameter, 3,5 cm height, column volume $\sim$1 drop) was finally used to concentrate the californium in a very small volume.

CONCLUSIONS

From the data presented above first of all it may be concluded that the excellent separation properties of extraction chromatography columns loaded with HDEHP observed when working on a tracer scale have been fully confirmed with quantities of transplutonium elements in the 10 micrograms range (for ^{252}Cf) to several milligrams for ^{244}Cm. In addition, it has been found that purification of the curium + americium from the fission products coeluted (in particular ^{106}Ru/^{106}Rh and Cs) from a LiCl-column can be easily and effectively performed. Separation of europium from californium,eluting just before the last element in 0.5 M HCl,can also be achieved, either by repeated extractions or by extraction with less concentrated acid. Elimination of LiCl is very easy. The columns can be very easily reconditioned for use.

By simply varying the concentration of the eluting agent between 0.1 and 1 M (HCl) and by selecting the moment for stripping the column, many different separation and purification cycles and combinations of a number of transplutonium and rare earth elements can be realised. Given the considerable influence of HCl-concentration on the elution peak distance($E_a^o \sim$inverse third power) and the large number of plates per unit length almost unlimited possibilities for separations exist provided a minimum difference in extraction coefficient between elements. From known extraction coefficients there is every reason to conclude that this technique will also be useful for separating elements with $Z > 98$. Elsewhere a more detailed discussion of these questions will be presented.

As may be derived from the above the columns behaved ideally from an operational point of view. They showed extreme stability in performance, even after some 15-20 micrograms of californium-252 had been passed. The 4 mm column showed no deterioration after about 10 milligrams of curium had been treated. Up to now we have no reason to believe that batches of the order of 100 milligrams of curium-244 could not be conveniently handled by a small HDEHP column.

To our opinion the results obtained during this processing would fully justify, for quantities of material comparable to those handled here, to attempt to replace the conventional LiCl-columns by extraction chromatography columns. Some of the major advantages would be the following:

1. The replacement of the delicate and time consuming LiCl-columns and conversions into and from the LiCl-system by a straightforward and simple treatment (chemically and operationally) of the solutions resulting from the dissolution step or from a HCl-column.

2. A simpler flow sheet and consequently simpler apparatus and a considerable reduction of the number of different operations, solutions etc.

3. Very likely, and at an early stage of the processing, a better fractionation in well defined fractions, which in turn can be easily prepared for further treatment, or other uses.

4. In addition to the fact that the elements separated are quantitatively recovered, all remaining activity can either be stripped off with more concentrated HCl or recovered in a very small volume by passing an organic solvent. Waste disposal of active material and column material is therefore extremely simple.

ADDITIONAL EXPERIMENTAL RESULTS

The plutonium fraction obtained from the HCl-column run was evaporated to dryness, taken up into 8 M HNO_3 and converted to Pu(IV) with potassium nitrite. It was purified from other activities by ion exchange separation in the usual way : fixed on a Dowex AG 1 column, rinsed with about 10 column volumes of 7.5 M HNO_3, stripped, after addition of potassium iodide. From the eluate a pure plutonium fraction was obtained by two successive extractions into TTA.

Mass-spectral analysis* showed the composition to be (in atom percent)

238 : 1.04
239 : 0.16
240 : 20.74
241 : 0.69
242 : 76.74
244 : 0.62

From this follow activity ratios for 238/239+240/242 of 100/26.6/1.7. From α-spectra the same ratios were found to be 100/27.2/1.8. The ^{244}Cm/^{238}Pu mass ratio was found to be $< 2 \cdot 10^{-6}$.

The curium+americium fractions from the LiCl-columns were precipitated as the hydroxides, separated by centrifuging, taken up into hydrochloric acid and stored after evaporation to dryness. Mother liquids were recovered and again treated in the same way.

Aliquots of the HCl-solutions were assayed for total α-counts. The total activity, expressed in the same relative units as used before amounted to $6.1 \cdot 10^{11}$ dpsα. This figure corresponds very well with those found earlier.

Preliminary mass spectrometry measurements on a sample of californium were in accordance with preliminary data derived from

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* Our sincere thanks are due to Dr. G. DEBUS and Mr. P.J. DE BIEVRE of the Central Bureau for Nuclear Measurements at Geel who very kindly carried out these measurements.

α-spectrometry indicating a value of ∼0.07 for the $^{250}Cf/^{252}Cf$-α ratio.

Preliminary data on the total quantity of californium isolated calculated from neutron counting were obtained by comparing neutron counts on a portable commercial neutron counter of the different batches isolated with a reading obtained from a sample which had been measured at the Euratom Transuranium Institute in Karlsruhe*. Since at the moment of writing an accurate calibration of this counter still had to be carried out the data obtained still need further accurate verification. Nevertheless, the total neutron count corresponds to within 10 percent with the total fission count. They indicate a total quantity of californium-252 isolated of 13-13.5 micrograms (May 1965). Equally preliminary data indicated that the total quantity of ^{249}Bk isolated (May 1965) amounted to ∼0.7 μg. This would give an initial $^{249}Bk/^{252}Cf$ ratio of 0.19, in agreement with data obtained by others as reported by Cunningham$^{6)}$.

At the time of writing all measurements on yields and composition still have to be considered preliminary and subject to modifications resulting from more detailed measurements.

We examined a sample of the condensate collected before the molecular sieves during the evaporation step. Only traces of α-activity were found. Isotopes identified included ^{144}Ce, ^{144}Pr, ^{134}Cs, ^{106}Ru and ^{156}Rh.

All liquid waste originating from this processing was solidified by adding it to mixtures of vermiculite and cement. A careful check was made of the activity level of the containers and in particular care was taken that neither waste nor contaminated equipment or apparatus contained any neutron activity. All glassware was carefully washed before ultimate disposal and counting samples were recovered, yielding a few minor fractions of the different elements which were subsequently purified and used for preparing samples for experimental uses.

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* Our thanks are due to Dr. M. GEERLINGS for use of his counter and his assistance with the measurements.

DISCUSSION

A few additional remarks of a more general nature will be made here.

First of all it may be pointed out that all radioactivity measurements made in the course of this run were found to be fully consistent. The experimental losses therefore must have been very small. The major loss occurred in the dissolution step, most likely by occlusion in insoluble material. It, however, did not surpass 0.5 %. In all other steps losses have been considerably less. We estimate that total overall losses do not amount to more than in the order of 2 %.

Besides enabling us to recover the plutonium at an early stage, the hydrochloric acid column served a very useful purpose by eliminating roughly half of the fission product activity.

The flow sheet followed proved to be satisfactory and reliable. Although both lithiumchloride columns worked very smoothly, this part of the procedure certainly was the most delicate and time consuming one. The good results obtained with the extraction chromatography columns indicate, to our opinion, that they very likely may replace the LiCl-columns although a few details still need to be checked.

One of the advantages of their use will be that the berkelium fraction can be easily obtained at a well defined point and in good purity. That in our case the bulk of the berkelium was coextracted with the californium in the LiCl-column runs was probably rather due to simultaneous stripping than to real co-elution. Even so their separation is easy.

Since it is difficult to identify ^{249}Bk easily and unambiguously, it is also very difficult to check its quantitative recovery. Therefore, it might be most useful to run first of all, after dissolving the slug, a small extraction column on an aliquot of the original slug-solution. If successful this would also prove that the flow sheet could be drastically simplified. An experiment of this kind is being planned. In addition, it may be pointed out that in all cases the Bk-samples obtained by extraction chromatography had a purity sufficient to permit following the α-ingrowth.

ACKNOWLEDGEMENTS

First of all our most sincere thanks are due to the United States Atomic Energy Commission, the United States Transplutonium Program Review Committee and all its members and participating research laboratories for making the irradiated americium available to Euratom's Transplutonium Program. It not only permitted us to acquire invaluable experience at a much earlier stage than otherwise would have been possible, it also enables Euratom to fill many requests filed by European Laboratories for samples of elements, which otherwise would not have been available.

Amongst the many american colleagues who in some way or another gave us our support, technically and morally, we may mention in particular Dr. S.M. Fried, Berkeley, who came up with the original suggestion and subsequently gave us much good advice during his stay at Mol. We are also particularly grateful to Drs. R. Leuze (Oak Ridge) and E. Shank (Oak Ridge,p.t. Mol) for their fine collaboration and much valued support as observers for the U.S.A.E.C. and to the late Dr. J.C. Wallmann (Berkeley) for his stimulating interest, willingness and support.

We gratefully acknowledge the collaboration of Messrs. Coppens, Slegers and Hermans of CEN/SCK Mol, which greatly contributed to the successful completion of this run. Our thanks are also due to the other members of the transplutonium group at Mol who took their share of the physical burdens.

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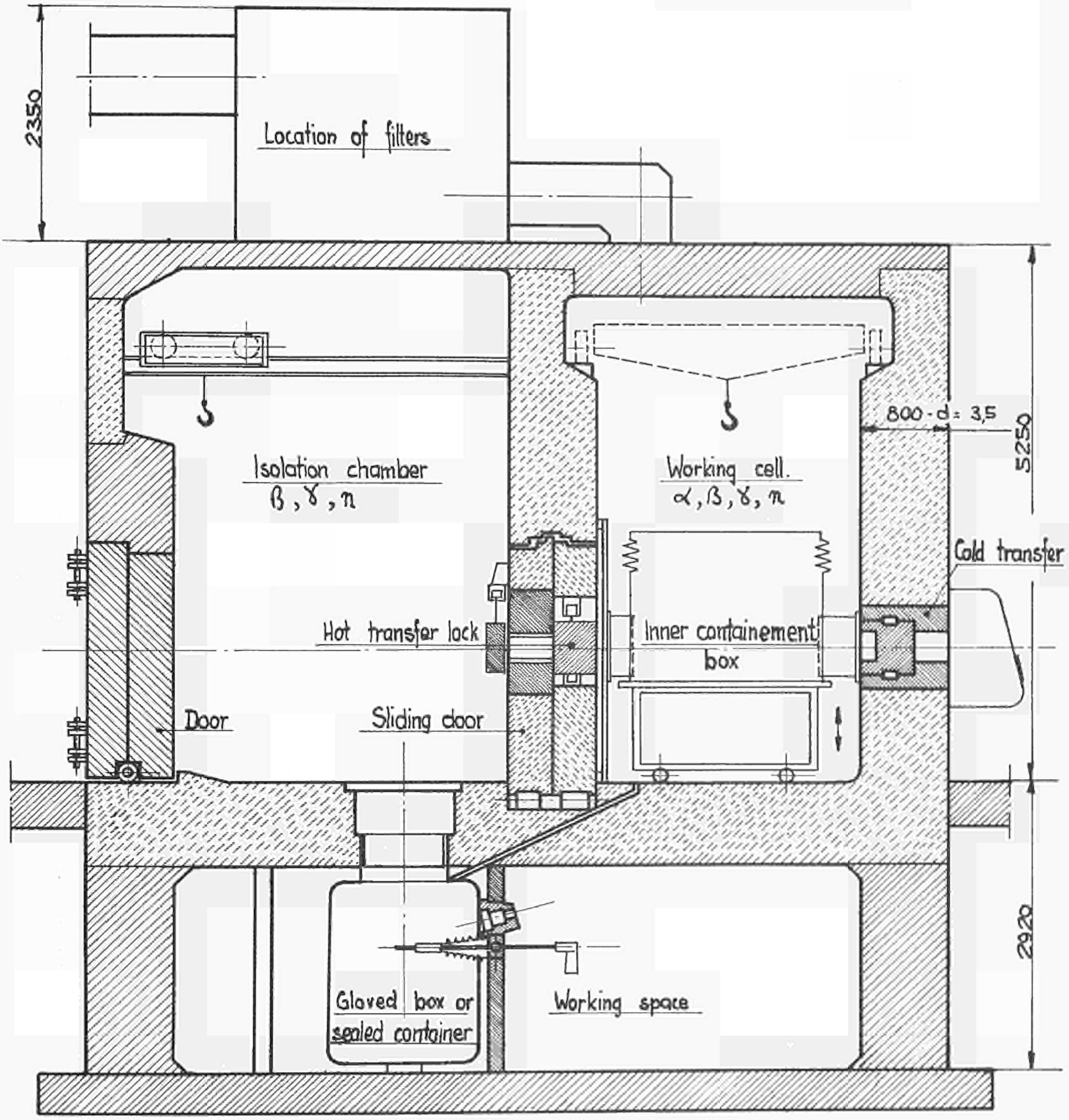
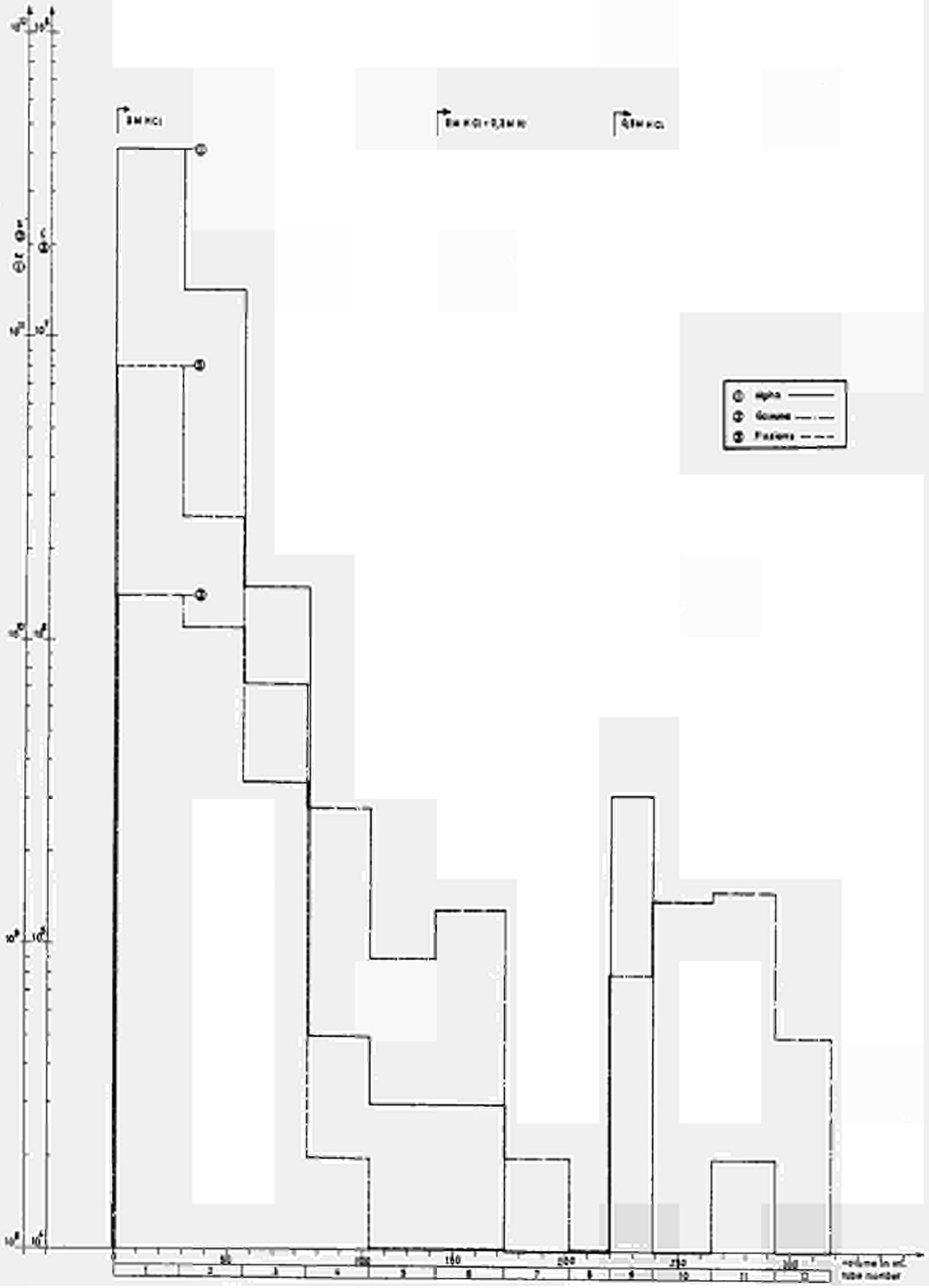
TRANSPLUTONIUM CELL

1000 CURIES

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8 M HCl + 0,3 M HI
0,5 M HCl
① Alpha
② Gamma
③ Fissions
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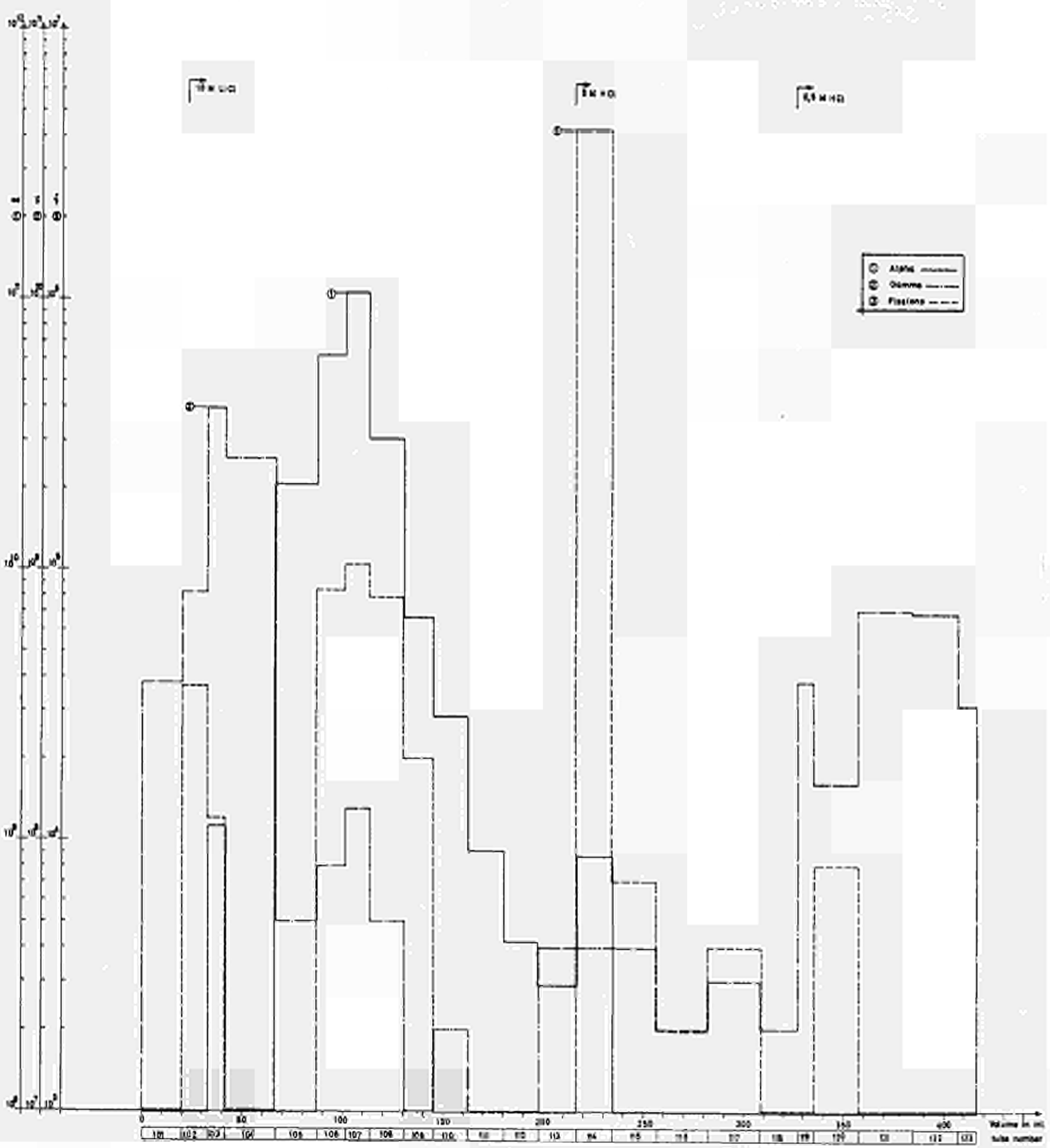
300
volume in ml
1
2
3
4
5
6
7
8
9
10
11
12
tube number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① Alpha
② Gamma
③ Fissions
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300
350
400
Volume in ml
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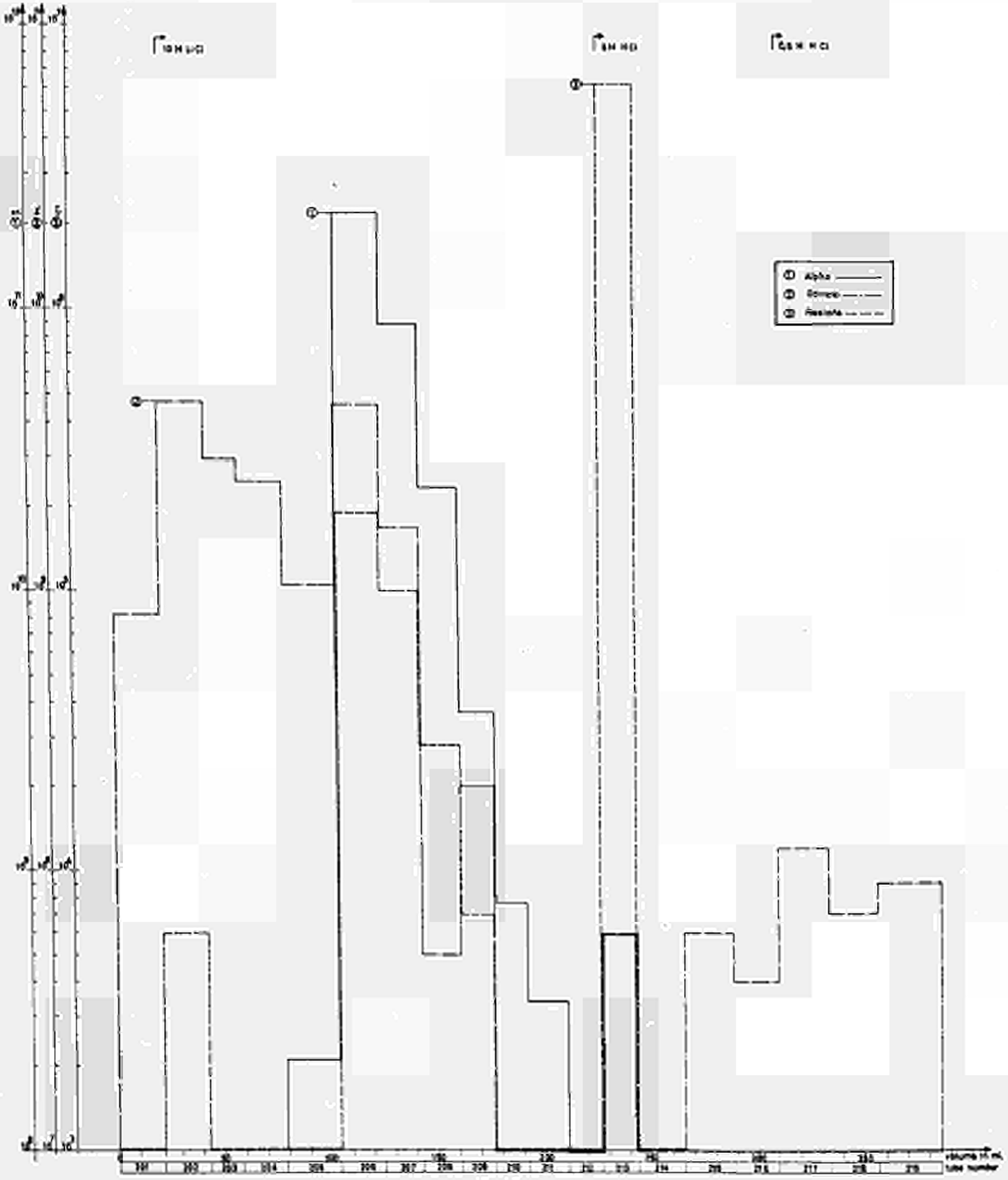
102
103
104
105
106
107
108
109
110
tube number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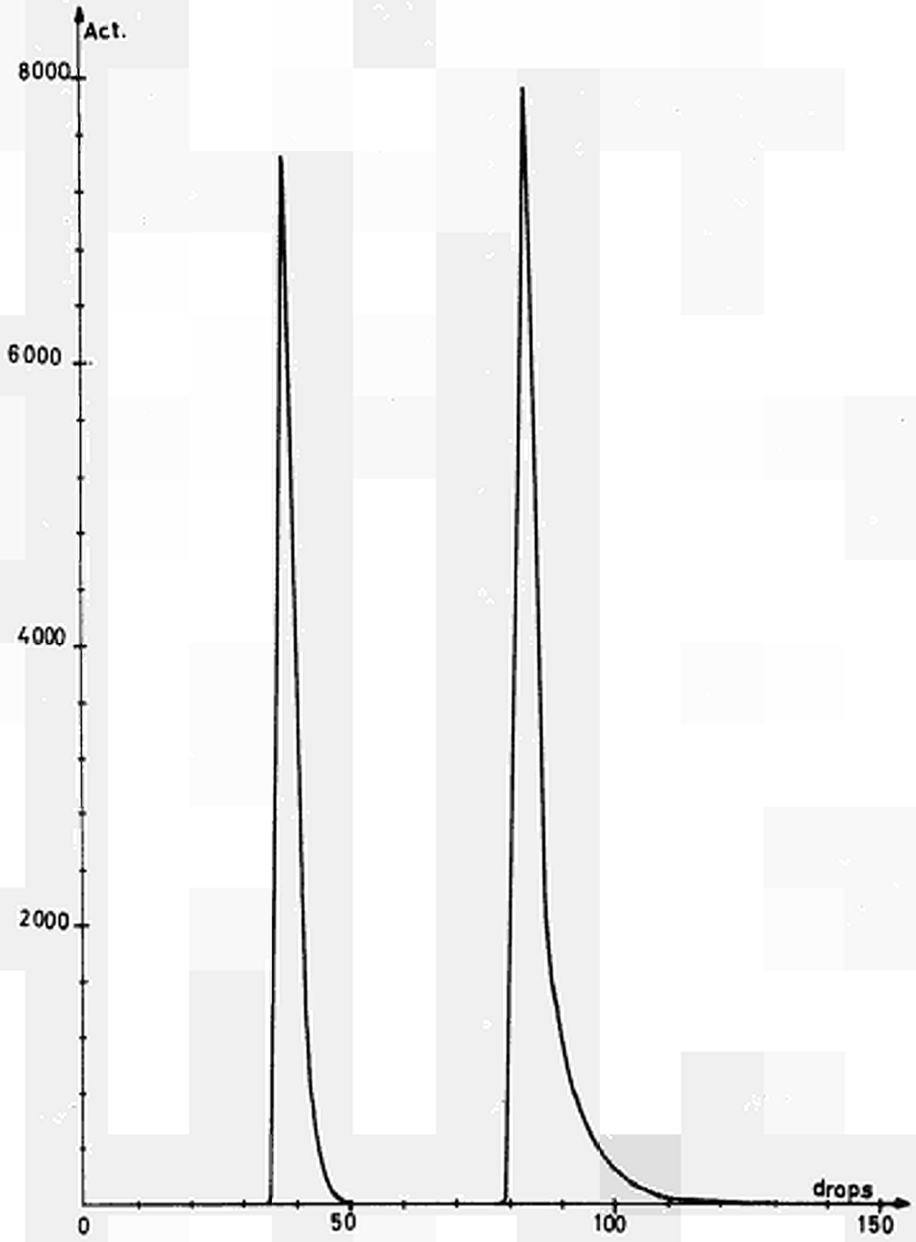
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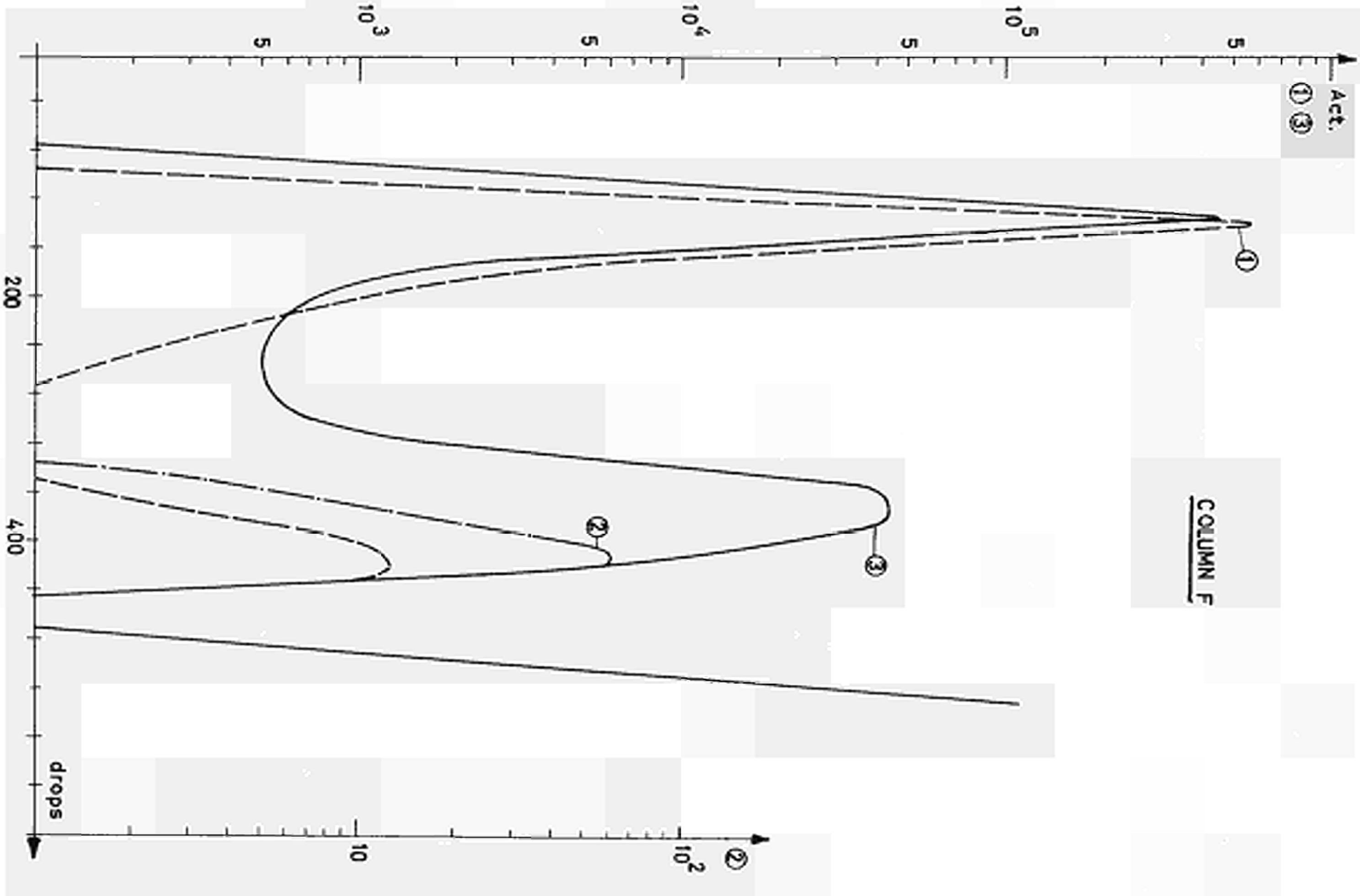
6000
4000
2000
0
50
100
150
drops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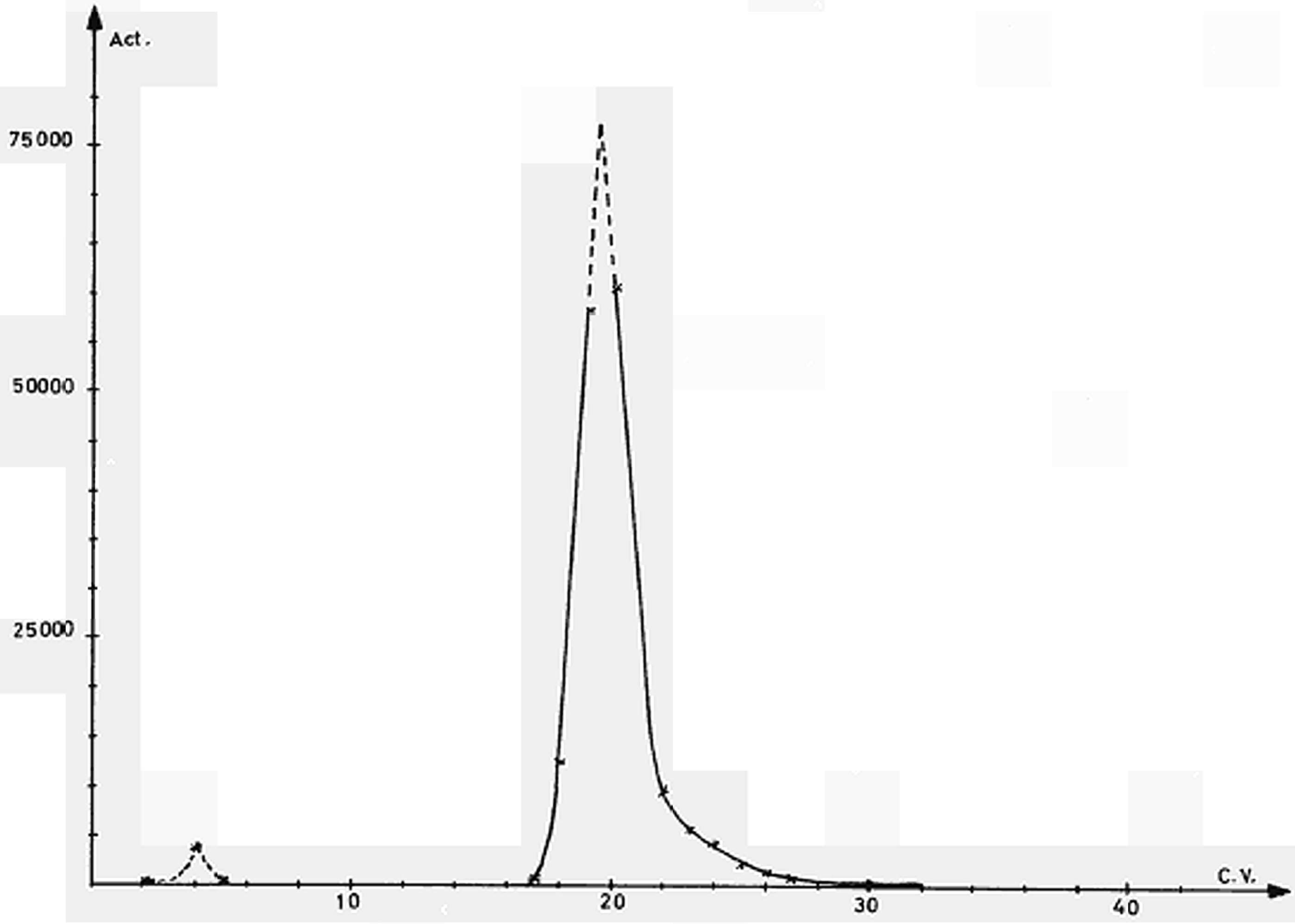
Act.
① ③
5
10^3
5
10^4
5
10^5
5
200
400
drops
10
10^2
②
①
②
③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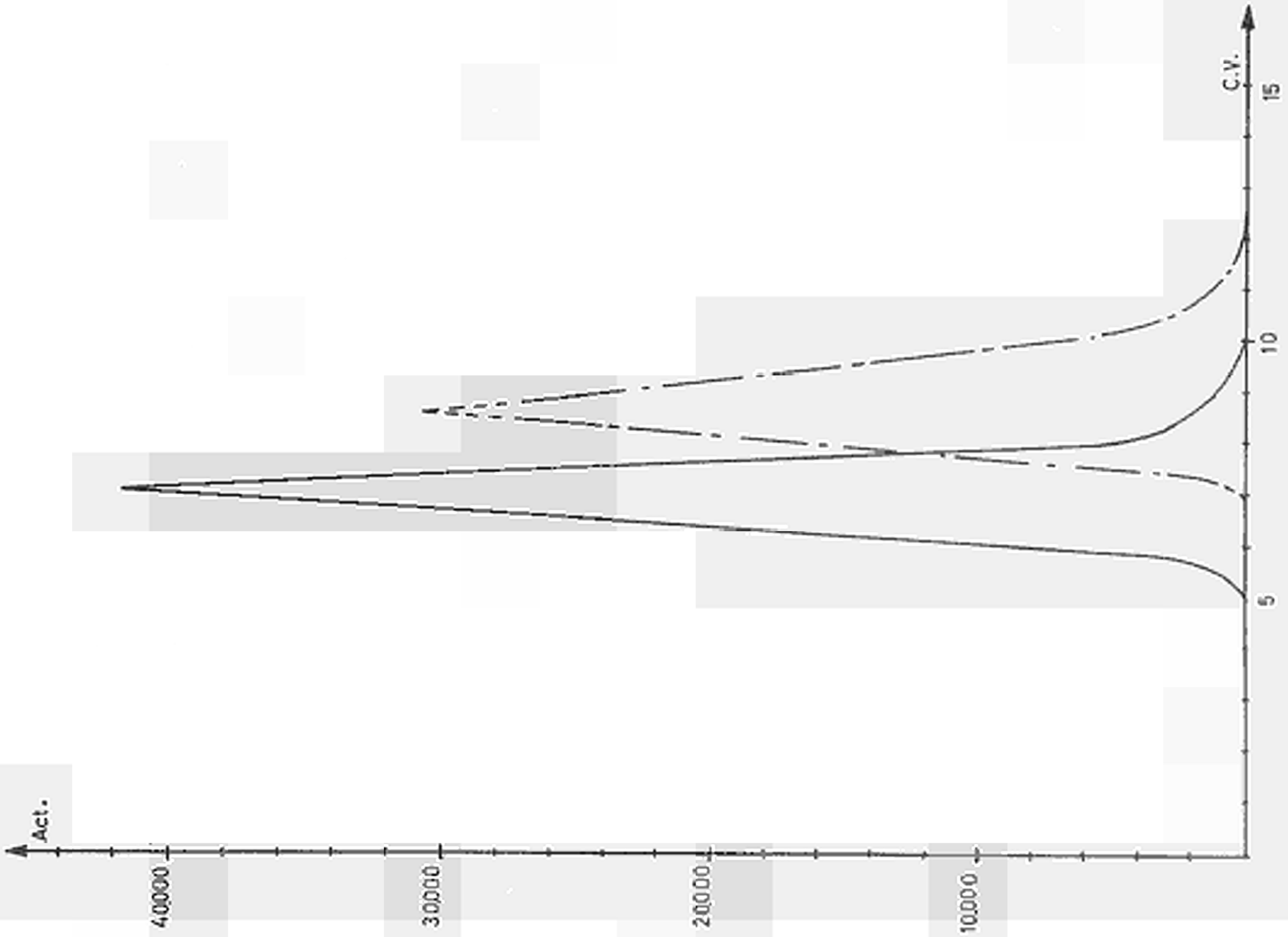
50000
25000
10
20
30
40
C.V.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10
5
Act.
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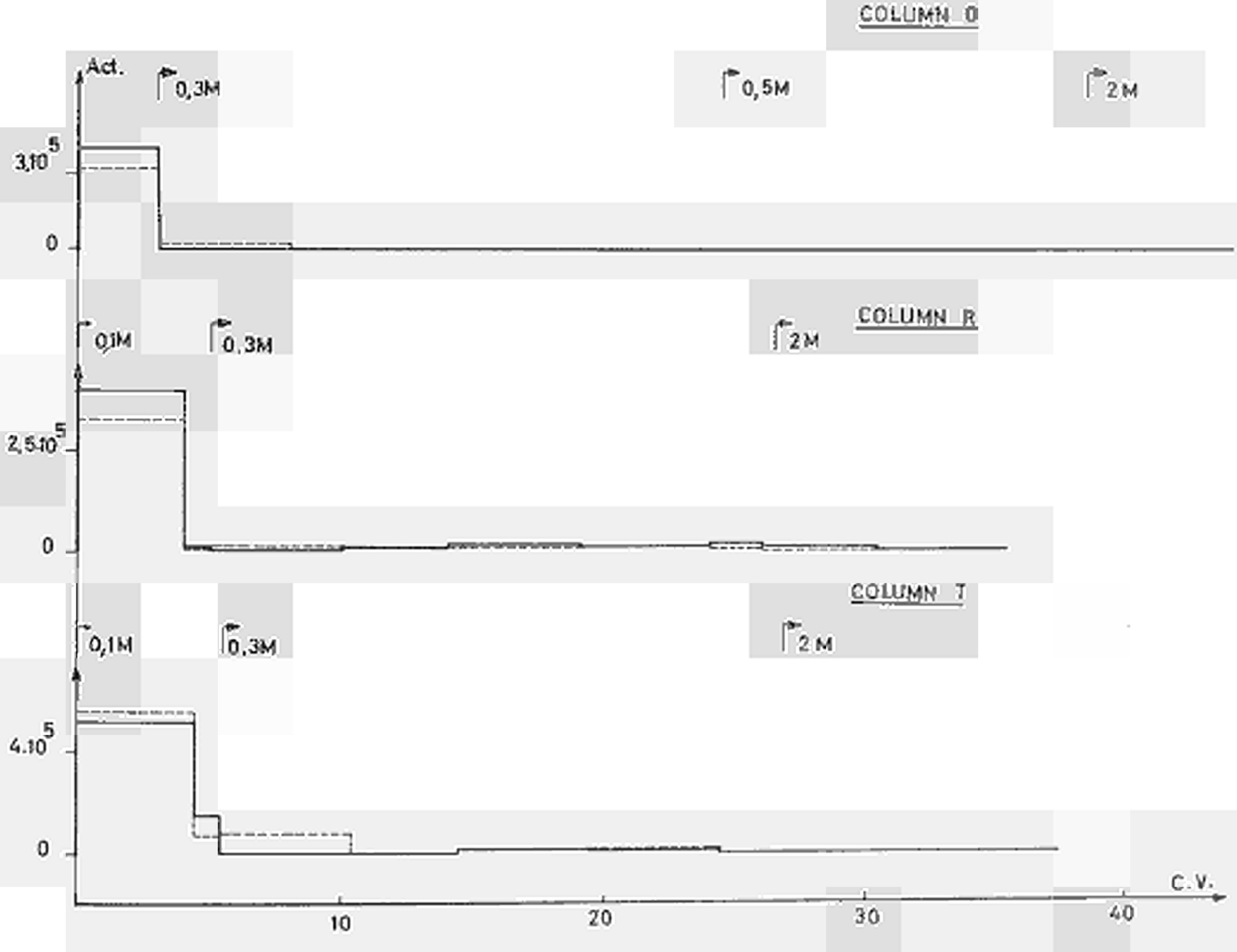
30000
20000
10000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2M
3.10^5
0
COLUMN R
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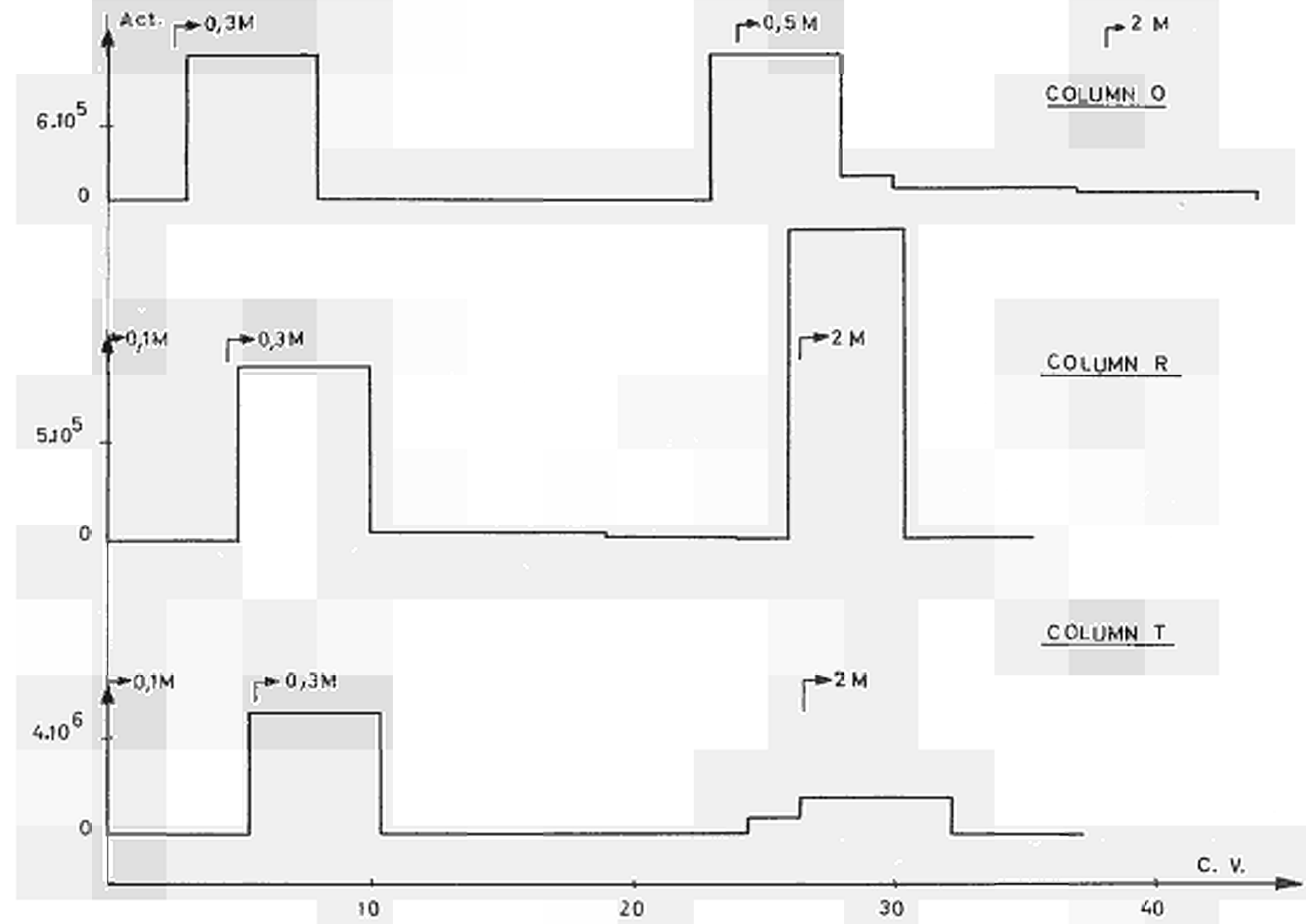
0,3M
2M
$2,5.10^5$
0
COLUMN T
0,1M
0,3M
2M
4.10^5
0
10
20
30
40
C.V.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0,5M
2 M
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0,1M
0,3M
2 M
COLUMN R
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2 M
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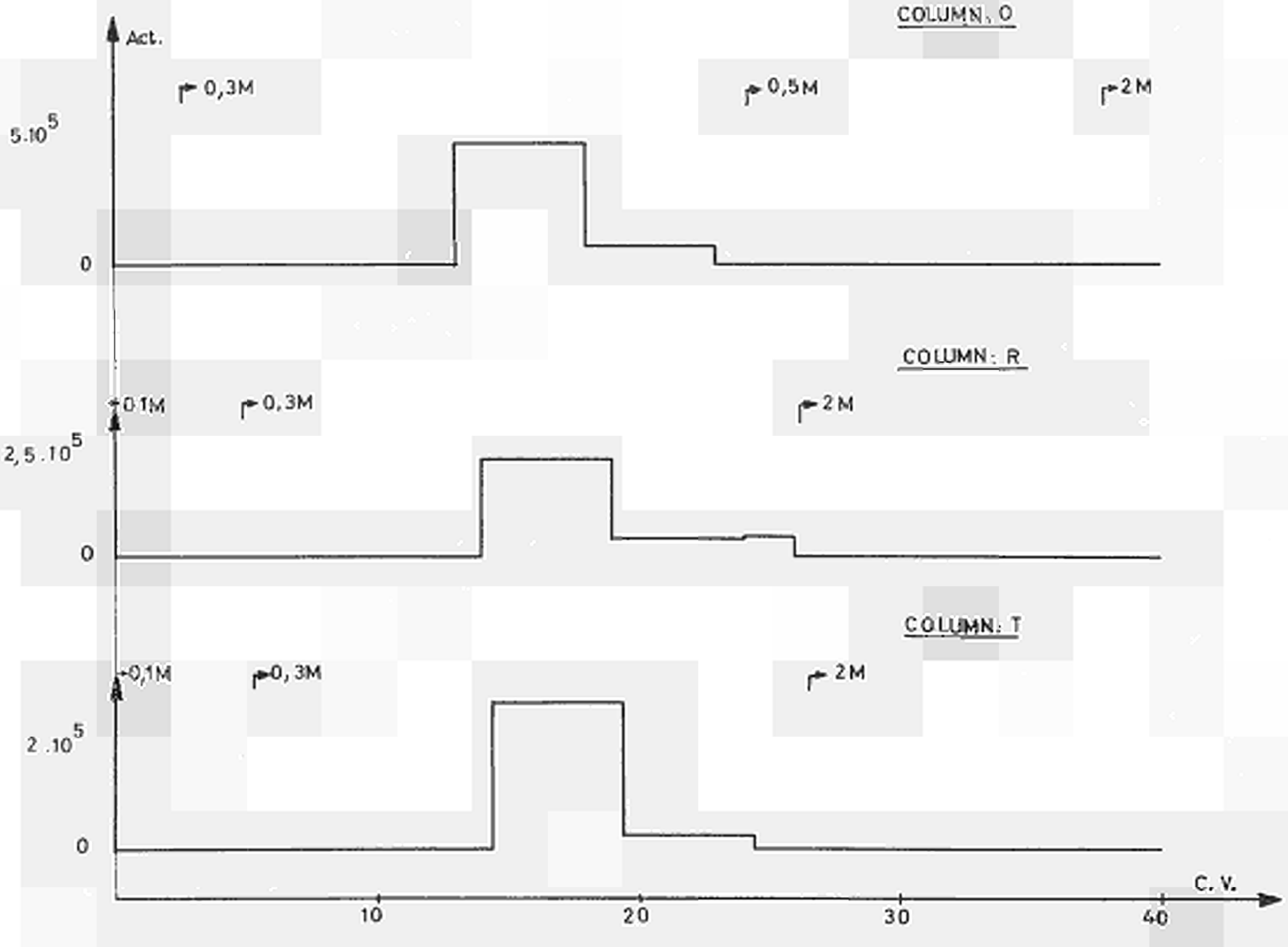
C. V.
10
20
30
40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Act.
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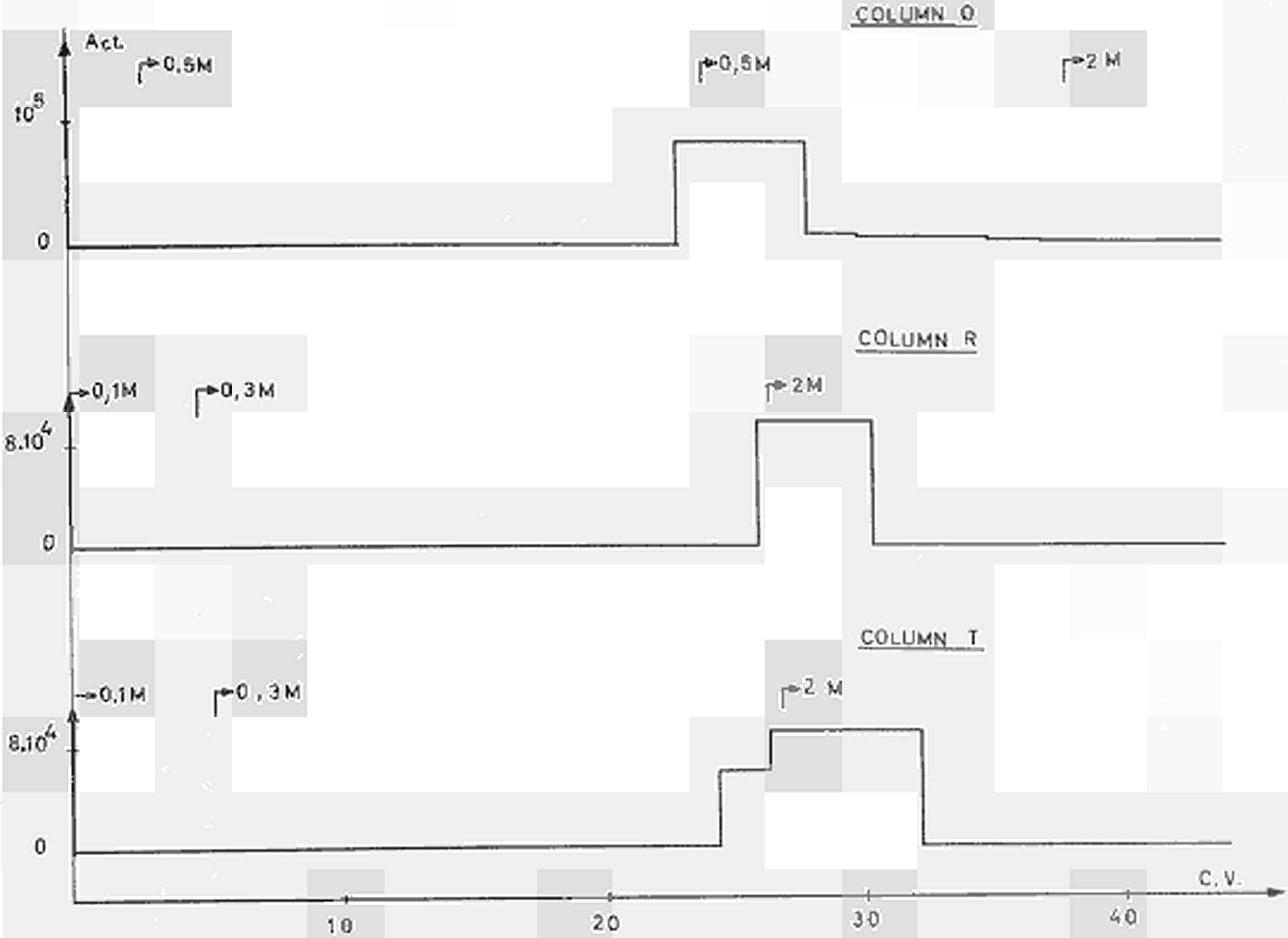
0,5M
2M
5.10^5
0
COLUMN: R
0,1M
0,3M
2 M
2,5.10^5
0
COLUMN: T
0,1M
0,3M
2 M
2 .10^5
0
C. V.
10
20
30
40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Act.
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2 M
10^5
0
COLUMN R
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0,3M
2M
8.10^4
0
COLUMN T
0,1M
0,3M
2 M
8.10^4
0
C.V.
10
20
30
40


C.V.
15
10
5
0
2M
0,5M
0,1M
Act.
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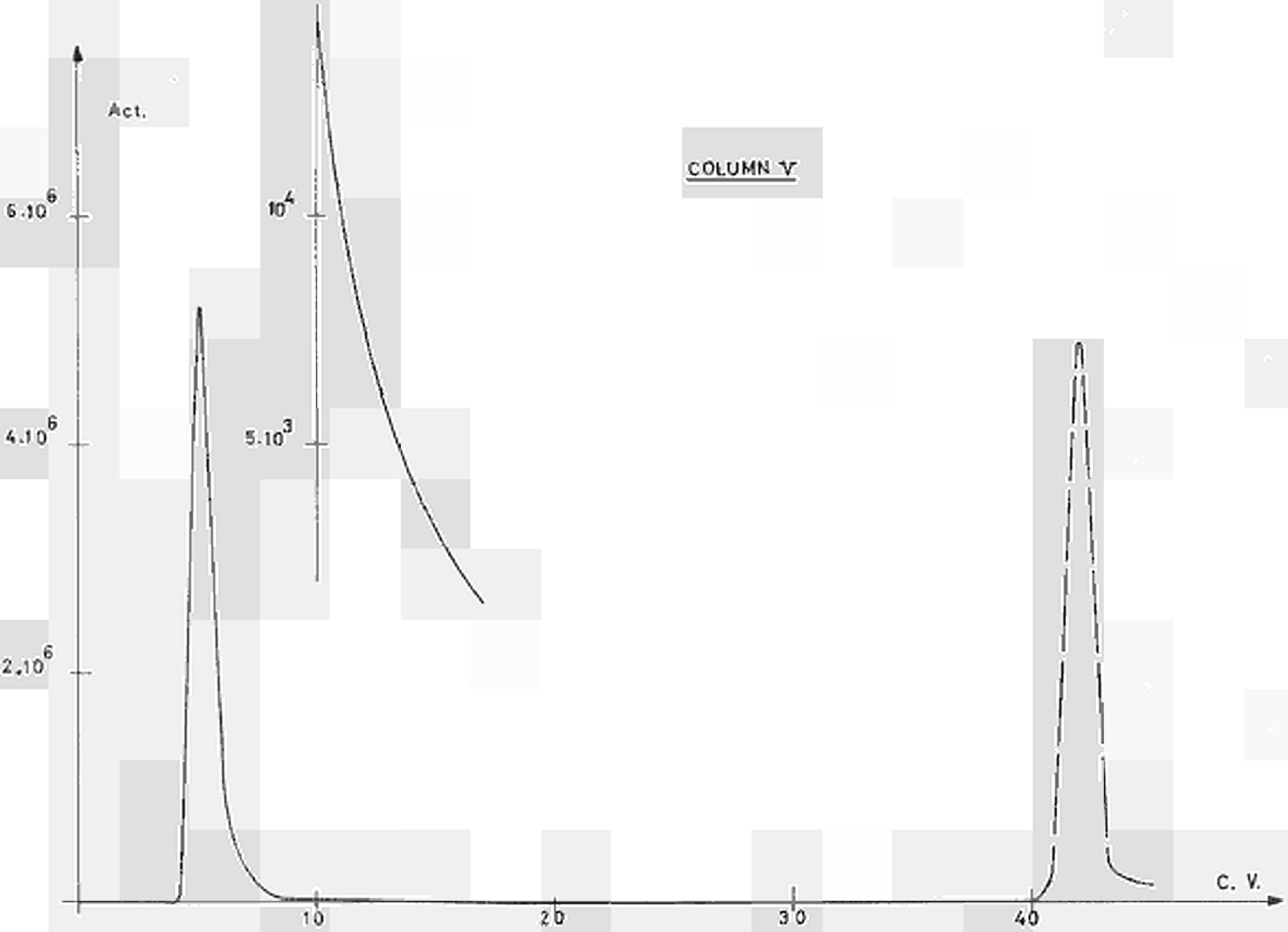
4.10^6
2.10^6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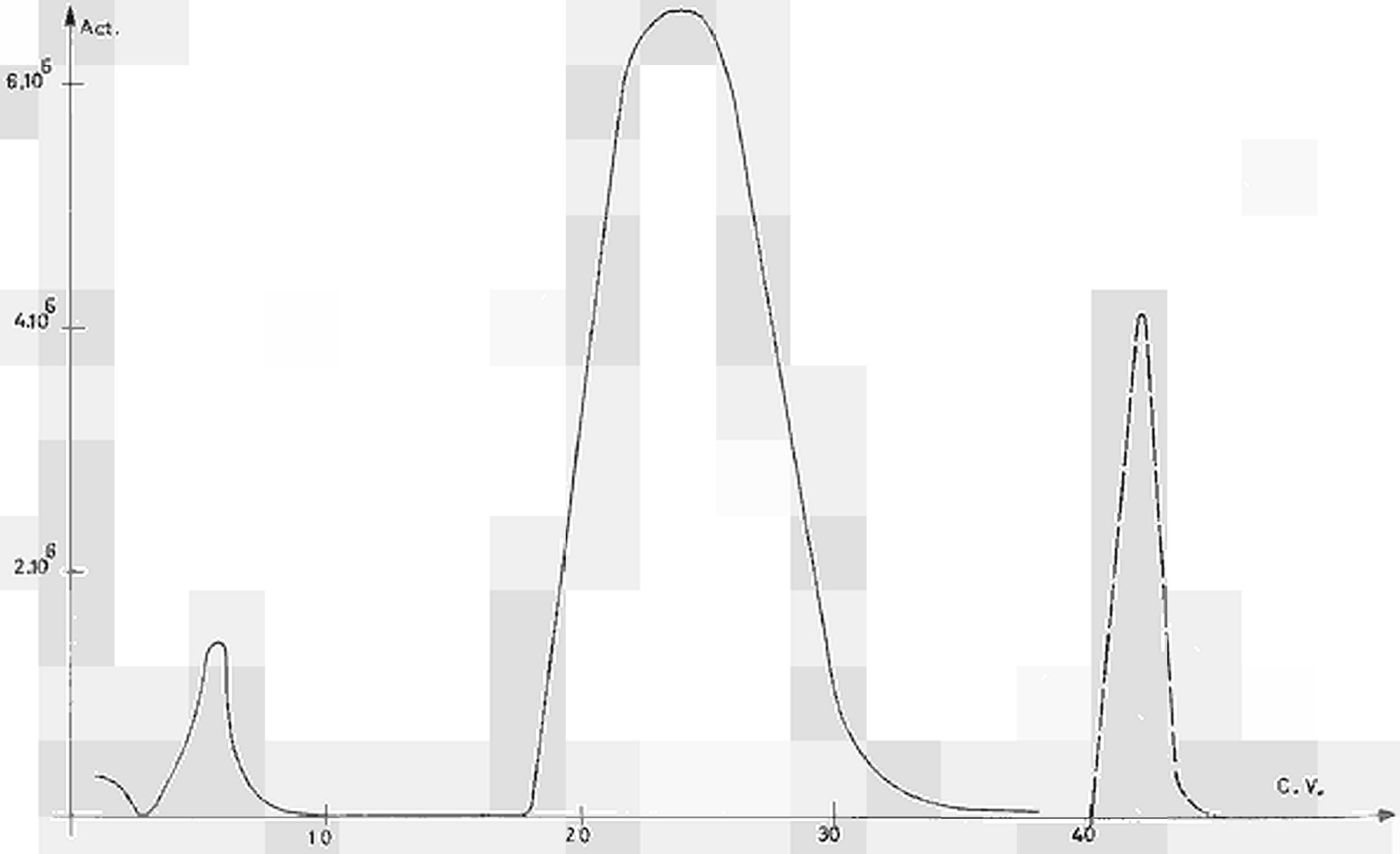
10
20
30
40


Act.
6.10⁶
4.10⁶
2.10⁶
C. V.
10
20
30
40

To disseminate knowledge is to disseminate prosperity — I mean general prosperity and not individual riches — and with prosperity disappears the greater part of the evil which is our heritage from darker times.

Alfred Nobel

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