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Recovery of Transplutonium Elements from Fuel Reprocessing High-Level Waste Solutions

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Zusammenfassung

Ein Verfahren zur Abtrennung von Americium und Curium aus den hochaktiven Abfall-Lösungen des Purex-Prozesses befindet sich in Karlsruhe in der Entwicklung. Der flüssige hochaktive Abfall wird denitriert und der entstehende Spaltprodukt-Niederschlag zur Waste-Verfestigung geleitet. Die Actiniden-Lanthaniden-Fraktion wird während der Denitrierung in Lösung gehalten und anschließend durch einen Extraktionszyklus mit HDEHP als Extraktionsmittel getrennt. Die Lanthaniden werden zur Waste-Verfestigung geleitet, während Americium und Curium durch Kationenaustausch weiter gereinigt werden. Die Ergebnisse von "kalten" Fließschema-Tests werden diskutiert.

Abstract

A process for the recovery of americium and curium values from Purex-type high-level waste solutions is under development at Karlsruhe. The high-level waste is denitrated and the resulting fission product precipitate sent to HLW-solidification. The actinide/lanthanide fraction which has been kept in solution is separated by a HDEHP solvent extraction cycle. The lanthanides sent to waste solidification, while the Am/Cm is further purified by cation exchange. Results of "cold" flowsheet tests are reported.

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1. Introduction

Spent nuclear fuels from power reactors contain appreciable amounts of transplutonium nuclides. In a fuel reprocessing plant with an annual throughput of 1000 tons of light-water reactor fuel, about 47 kg of ^{241}Am, 53 kg of ^{243}Am, and 10 kg of ^{244}Cm per year can be expected (based on figures given for LWR uranium fuels of 25 000 MWd/t burn-up (1)). During fuel reprocessing by PUREX-type processes the transplutonium nuclides accumulate in the high-level waste (HLW). Recovery of these values may be of interest, provided that there is a demand, and that an appropriate and economic process can be established.

The actual high-level waste management concept of the Federal Republic of Germany provides for solidification into a borosilicate glass after an intermediate liquid storage of about five years, and final disposal of the encapsulated glass blocks into salt formations. Since HLW vitrification asks for some chemical pretreatment of the waste solution, it is preferable to combine the recovery of any valuable by-products, e.g. the transplutonium elements, with the final waste treatment. Moreover, a liquid storage time of about five years offers the advantage that the shorter-lived fission products and also the shorter-lived transplutonium isotopes (in particular Cm-242) have decayed to negligible quantities, which leads to less radiation damage to process chemicals in the recovery operations.

It is evident that any additional chemical treatment of the high-level waste must be compatible with its final solidification. From that reason, a solvent extraction flowsheet which had been developed earlier (2) had to be abandoned, since it produced a citrate-complexed high-level waste solution which upon calcination leads to vigorous, and possibly explosive, exothermic reactions. Pretreatment of the citrate-nitrate solutions with various oxidants did not eliminate these vigorous reactions. Some improvement was attained by substituting the citrate by a lactic acid - NTA mixture (NTA = nitrilotriacetic acid) (3), but the modified flowsheet which is now under development does not produce any high-level waste containing organic substances, and hence is compatible with the final solidification process.

2. Flowsheet Description

The flowsheet considered for Am/Cm recovery is presented in Fig. 1.

The high-level waste concentrate (designated by "1WW") is transferred from the storage tank to the denitrator where the nitric acid is destroyed by formic acid (4). Conditions are maintained under which the americium and curium remain in solution while part of the fission and corrosion products (in particular Zr, Nb, noble metals, Mo, Fe) are precipitated. The precipitate is separated and transferred to the solidification stage. The denitrated waste solution which contains essentially the Am, Cm, fission product rare earths (RE), Sr, and Cs, is transferred to the feed preparation stage where the acidity is adjusted to extraction conditions.

The extraction cycle consists of four multi-stage extractors designated WA (coextraction of Am, Cm, and rare earths), WB (partitioning of Am/Cm into the aqueous phase, leaving the REs in the organic solvent), WC (rare earths back-extraction), and WD (Am/Cm purification from residual REs). The organic solvent is 0.3 M HDEHP + 0.2 M TBP in a n-alcane diluent (HDEHP = di(2-ethylhexyl)phosphoric acid; TBP = tributyl phosphate; diluent = "Alkan C_{10} - C_{13}", a product of Gelsenberg AG consisting essentially of straight-chain saturated C_{10} to C_{13} hydrocarbons). Separation of Am/Cm from rare earths in the WB bank is accomplished by a "Reverse - TALSPEAK" process (2) using 0.05 M Na_5DTPA - 1 M lactic acid as the aqueous partitioning (WBX) solution (Na_5DTPA = diethylene triamine pentaacetic acid, pentasodium salt). Rare earths back extraction (WC bank) is done by nitric acid. The Am/Cm purification (WD bank) is essentially a TALSPEAK-type (5) procedure; the RE-loaded solvent (WDW) from the WD bank is immediately recycled into the WA bank where it serves as part of the extractant (WAX). The spent solvent (WCW) from the WC bank is subjected to a solvent clean-up, and the washed solvent is recycled to serve as WA and WD extractant (WAX and WDX), and WB scrub solution (WBX), respectively. A full description of the different flows is given in Appendix A.

The products from the extraction cycle are: (a) The Am/Cm product solution (WDAC), a very dilute Am/Cm solution in 0.05 M Na_5DTPA - 1 M lactic acid, which is sent to concentration and further purification by ion exchange. (b) Two high-level waste solutions, i.e. WAW containing essentially the strontium and cesium in a dilute nitric acid medium, and WCRE containing essentially the rare earths in a rather concentrated nitric acid medium. It is clear that these solutions might constitute the sources for the recovery of ß or γ emitting nuclides (e.g., ^{147}Pm, ^{90}Sr, and ^{137}Cs), should there be any interest. Here it is proposed that the two high-level waste streams are combined and concentrated in a HAW evaporator, and that the HAW concentrate (HAWC) is backcycled to the denitrator, where it is denitrated and then sent to waste solidification by spray calcination and vitrification. Alternatively, a separate denitrator unit for HAWC denitration might be considered. (c) Two medium active waste (MAW) streams, i.e. the distillate from the HAW evaporator (HNO_3 medium), and the spent aqueous wash solution from the solvent clean-up (the composition of which is not yet defined), which require a MAW treatment.

The Am/Cm product solution from the extraction cycle (WDAC) is concentrated by a cation exchange cycle (IA column, in practice two parallel columns used alternatively). At the same time this cycle serves for further purification from contaminants, and in particular for separation from the complexants (Na_5DTPA and lactic acid). The WDAC solution is adjusted to pH = ca. 0.9, and is run through the IA column. Am and Cm are sorbed on the cation exchanger (Dowex-50 or similar resin).

The loaded column is washed with 0.1 M HNO_3 (IAS solution), and the Am/Cm is eluted with ca. 3 M HNO_3. The Am/Cm product solution (IAAC; about 2.6 g/l Am/Cm, 3 M HNO_3) is sent to final purification and Am/Cm separation. The waste from the ion exchange cycle, IAW, is a strongly complexing solution (Na_5DTPA + lactic acid); further treatment of this MAW seems to present severe problems. Hence, studies on possible back-cycling and re-use of this solution are presently being done.

3. Results

3.1 Conditioning of 1WW by Denitration

Denitration of 1WW by formic acid, as originally developed for complete destruction of nitric acid and heavy-metal nitrates prior to HLW solidification by spray calcination and vitrification (6,7), is essentially characterized by the following chemical reaction:

$$2HNO_{3(l)} + 4HCOOH_{(l)} \longrightarrow N_2O_{(g)} + 4CO_{2(g)} + 5H_2O_{(l)} - 202 \text{ kcal}$$

The exothermic reaction proceeds rather fast. If the waste is continually introduced into the boiling formic acid, the average composition of the off-gas is 78 vol.% CO_2 (80 vol.% calc.), 16 vol.% N_2O (20 vol.% calc.), 5 vol.% NO, and 1 vol.% N_2.

With the exception of the nitrates of the alkaline- and alkaline earth groups, other metal nitrates can also be destructed. Due to the redox equilibrium of the formic acid,

$$HCOOH_{(l)} \rightleftharpoons CO_{2(g)} + 2H^+ + 2e^- \quad (E = - 0,19 \text{ V})$$

some of the cations are at the same time reduced to lower valency states. Moreover since the pH of the waste solution rises during the denitration, a fine-grained precipitate is formed which essentially contains the hydrolizable metal ions.

In contrast to the complete denitration as anticipated in the high-level waste solidification process by spray-calcination and vitrification, only a partial denitration to a final hydrogen-ion concentration of roughly 10^{-2}M is aimed at in the 1WW-conditioning for Am/Cm recovery. At the same time it is aimed at an elimination of interfering fission products (essentially zirconium) in order to enable a direct extraction of the actinides from the 1WWD stream without addition of complexing agents. Laboratory studies of such a modified denitration process were started (7) using simulated 1WW solutions spiked with radionuclide tracers.

As shown in figure 2, partial denitration of the 1WW is possible by proper selection of the formic acid to nitric acid molar ratio. Figure 3 shows that the precipitate formation stays almost constant up to pH 1 and then increases with increasing pH. Batchwise denitration has been carried out in these experiments by continuously introducing a given volume of 1WW into the appropriate amount of boiling concentrated formic acid. More than 80 % denitration is obtained during this dosing phase, the rest is achieved in a secondary reaction phase which depending on the $HCOOH/HNO_3$ molar ratio takes between one and seven hours. (Similar results can be obtained by continuous operation, i.e. by dosing the corresponding amounts of 1WW and formic acid at room temperature through a short mixing zone into a reaction vessel of sufficient volume to provide the residence time needed for the boiling reaction mixture to achieve complete reaction).

Conditioning of the 1WW to optimum Am/Cm extraction conditions, i.e. a pH region 1 to 2, can be obtained in two ways:

In the first version, the 1WW is directly denitrated to the desired pH region by selecting the appropriate $HCOOH/HNO_3$ molar ratio. In this case fission product Zr/Nb and Mo are completely precipitated ($DF > 10^3$), most probably in the form of complex zirconium molybdato compounds. At the same time roughly 80 % of the fission product noble metals, e.g. Ru and Rh, are precipitated, whereas the other fission and corrosion products, as well as the transplutonium elements, remain almost quantitatively in solution. Apart from corrosion product iron which is reduced to soluble Fe(II), the cations which strongly interfere with the extraction step are eliminated in this way.

In the second version, the denitration is performed at a higher $HCOOH/HNO_3$ molar ratio leading to a higher final pH and hence to increased precipitation. For instance, at an $HCOOH/HNO_3$ ratio of 2 a final pH between 7 and 8 is obtained, and apart from the fission products of the alkaline and alkaline earth groups all other fission and corrosion products are almost completely precipitated together with the actinide elements. After completion of the denitration reaction Fe(II) can be oxidised to Fe(III) by means of hydrogen peroxide and the pH of the 1WW solution can be decreased by controlled HNO_3 addition, which leads at the same time to a gradual redissolution of the precipitate. As demonstrated by figure 4 this version allows to eliminate also iron, which remains in the precipitate as ferric hydroxide. At pH 3, DF values of $>10^3$ are obtained for the fission products Mo, Zr, Ru, Rh, and corrosion product Fe; at pH 2 the values for Zr/Nb and Mo remain practically unchanged, and only about 1 % of Ru/Rh and 2 % of Fe are redissolved. As on the other hand roughly 50 % or 80 to 90 % of the rare earth and transplutonium elements are redissolved at pH 3 and 2, respectively, this version offers an interesting 1WW conditioning step allowing an elimination of corrosion product iron, too.

Precipitates may either be filtered on glass grain filters which can be vitrified after exhaustion, or redissolved in nitric acid which is sent to HLW solidification.

3.2 Solvent Extraction Cycle

Parameter studies were carried out in order to optimize the solvent extraction cycle (8,9), with the results given below:

1. A pH in the range 0.8 to 1.5 is optimum for Am/Cm extraction in the WA battery. The aqueous phase is not buffered, hence the equilibrium pH is determined by the amount of H^+ ions released by the liquid ion exchange reaction,

$$M^{n+}_{(aq)} + n(HA)_{2(org)} \rightleftharpoons nH^{+}_{(aq)} + M(HA_2)_{n(org)} \quad (HA=HDEHP)$$

2. The metal concentration in the feed (WAF) solution is a compromise between overall efficiency and performance of the WA battery. Good efficiency would demand that the metal concentration be high, which can lead however to precipitate formation at the feed inlet stage (due to overloading of the organic phase), and to a lowering of the pH below 0.8 (due to excessive release of H^+ ions to the aqueous phase).

3. The extractant concentration is a compromise between overall efficiency and performance of the WB battery. Overall efficiency would demand that the HDEHP concentration be high, but this is impaired by the necessity of rising the aqueous back-extractant (WBX) stream in the WB battery.

4. The Na_5DTPA concentration in the WBX solution is an upper limit with regard to solution stability. At considerably higher DTPA concentrations, e.g. 0,1 M DTPA, precipitation of DTPA occurs after several days.

5. The optimum pH of the WBX solution is 3.0 ± 0.1. Lowering the pH would lead to less effective back-extraction of Am/Cm; moreover the 0.05 M Na_5DTPA/1 M lactic acid solutions tend to become unstable at pH values below ca. 2.5 (precipitation of DTPA). Rising the pH would rise the stripping efficiency while essentially maintaining the actinides/lanthanides separation factors, but a considerable part of the HDEHP would be converted to the sodium salt, which would finally lead to a higher sodium content of the high-level RE waste (WCRE) solution.

6. The HNO_3 concentration of the rare earths back extraction (WCX) solution is essentially determined by the behaviour of yttrium, which in contrast to the fission product lanthanides is not well back-extracted by lower (e.g. 2M) HNO_3 concentrations (9).

Counter-current tests of the flowsheet were performed in mini-mixer-settlers using synthetic (inactive) denitrated 1WW solutions spiked with radioactive tracers (9). The results can be summarized as follows:

1. Am/Cm and the rare earths (Ce, Nd, Eu, Y) show "regular" behaviour in the WA, WB, and WC batteries. In particular, separation of Am/Cm from REs in the WB battery is good, with DF values ranging from 500 (Eu) to >4000 (Ce). Concentration profiles for Am and Nd are given in Figs. 5 and 6.

2. In the WD battery, Am behaves as expected, with Am losses of <0.2 %. From the rare earths tested, cerium behaviour is "regular", with an additional DF >200. Europium, however, showed "irregular" behaviour, yielding an additional DF of only 2. This is evidently due to the fact that europium extraction by HDEHP from DTPA/lactic acid solutions is rather slow (2). The use of long-residence-time contactors in the WD battery would thus presumably improve europium decontamination. Concentration profiles are given in Fig. 7.

3. A number of metal ions, among them Fe(III), Zr(IV), Nb(V), and RuNO(III), exhibit slow extraction and back-extraction kinetics under the conditions of this process (3,8,10). Hence "splitting" of these metals occurs in the WA and WB batteries, i.e. partial extraction in WA and partial back-extraction in WB. The amount which is extracted in WA or back-extracted in WB and hence accompanies the Am/Cm, or in other words the decontamination factor, is dependent on the particular extraction kinetics of the respective metal ion and on the hydromechanic conditions in the extraction apparatus (i.e. intensity of mixing, residence time, etc.) Use of fast contactors would presumably improve the decontamination of Am/Cm from those metals, provided that Am/Cm extraction kinetics is fast enough (2). In the WC battery, the above-named metals are essentially retained in the organic phase. Pu(IV) and Np (introduced as Np(V)) exhibit similar behaviour. As an example the behaviour of iron(III) in laboratory mixer-settlers is shown in the concentration profile, Fig. 8.

4. Uranium(VI) is essentially completely extracted in the WA battery, but is retained in the organic solvent in WB and WC, thus leading to a good decontamination factor.

5. Cesium and strontium are essentially not extracted in WA.

The overall decontamination factors of the Am/Cm product obtained in the solvent extraction cycle using mixer-settlers were: Ce, $>8\cdot10^5$; Eu, $1\cdot10^3$; Zr(IV), 20; Nb(V), 20; RuNO(III), 50; Fe(III), 420; Sr, $>10^3$; Cs, $>10^4$; Pu(IV), 250; Np (introduced as Np(V)), 800; U(VI), >650. For Nd and Y, DF values up to now have been measured for the WA-WB-WC cycle only; these values, which do not include the expected benefits from the WD battery, were 870 for Nd and 1300 for Y. Measured Am losses to the different waste streams were 1.2 %, which corresponds to an americium recovery of 98.8 %.

3.3 Ion Exchange Cycle

The optimum pH region for sorption of Am^{3+} from 0.05 M DTPA/ 1 M lactic acid solutions by strongly acidic cation exchange resins (Dowex-50 X 8 and AG 50 X 12) is at pH = 0.9 - 1.0 (11), with distribution coefficients of $\geq 10^3$. The Am/Cm product solution (WDAC) from the solvent extraction cycle is adjusted to pH = ca. 0.9, and is passed through the cation exchange column. Due to the appreciable Na^+ concentration of this solution the H^+ form of the resin is partly converted to the Na^+ form, and at the same time some DTPA is sorbed on the resin (as H_6DTPA^+?). The Am^{3+} which is more strongly bound to the resin displaces part of the Na^+, but the full capacity of the resin is not available for the Am^{3+}. A sharp Am zone boundary is formed which moves down the column with a "constant pattern" (see Fig. 9; Eu^{3+} which behaves very similar to Am^{3+} (11) has been used to simulate americium in many of the ion exchange experiments). That part of the resin capacity which under equilibrium conditions is available for Am sorption is designated "maximum usable capacity"; it depends on the chemical conditions of the solution and on the resin but not on column dimensions or flow rates. Maximum usable capacities for

<u>Table I.</u> Maximum usable capacities for Dowex 50 and Lewatit SP (macroporous) resins of different cross-linkage. Aqueous feed 0.001 M Eu(NO$_3$)$_3$, 0.05 M Na$_5$DTPA, 1.0 M lactic acid, pH = 1.0 (adjusted by HNO$_3$). Column cross-section 5.07 cm^2, bed height 20 cm (21°C) or 10 cm (60°C), flow rate 1.6 ml/cm^2·min, particle size 20 ÷ 50 mesh. Capacities given in meq per gram oven-dried resin or meq per ml wet resin.

Resin	temp. °C	maximum usable capacities	
		meq/g	meq/ml
DOWEX 50 X 4	21	1.02	0.26
	60	1.37	0.35
DOWEX 50 X 8	21	1.42	0.54
	60	2.00	0.76
LEWATIT SP 100 (8 % DVB)	60	2.01	0.71
DOWEX 50 X 12	21	1.94	0.88
	60	2.42	1.10
LEWATIT SP 112 HL (12 % DVB)	60	2.35	0.83
LEWATIT SP 120 (18 % DVB)	21	2.00	0.59
	60	2.73	0.80

different resin types and temperatures are given in Table I. Break-through curves measured under the same conditions as indicated in Table I are represented in Fig. 10. Resins of high cross-linkage have considerably higher capacities for Eu^{3+} than low cross-linkage resins but sorption is slower with the former; 8 % cross-linkage seems to be the best compromise. Rising the temperature from 21 to 60°C increases the usable capacities by a factor of about 1.5 and also increases the sorption rate.

After loading the column is washed with 2 - 3 bed volumes of 0.1 M HNO_3 (IAS). The Am/Cm is then eluted by HNO_3 (IAX solution). The optimum HNO_3 concentration with respect to minimum elution volume and minimum HNO_3 consumption is at about 3 M HNO_3 (Fig. 11). Minimum elution volumes and therefore highest concentration factors were obtained at flow rates of 0.3 to 0.4 ml/cm^2 min, and with resin cross-linkages of 8 % (Fig. 12). That part of Na^+ and DTPA which was sorbed on the column (cf. Fig. 9) is eluted together with the Am/Cm product (IAAC) solution. The Am/Cm can be separated from these contaminants by adjusting the H^+ concentration to 0.2M and precipitating the actinide oxalates.

A study on fission product behaviour during the ion exchange cycle is under way. Measured distribution coefficients under loading conditions were 110 for Ru and 2 for Zr/Nb. Part of the Ru was irreversibly sorbed, and was not desorbed during elution. Decontamination factors obtained in preliminary column experiments were 18 for Ru and 1100 for Zr/Nb.

4. Conclusions

The process which has been outlined above seems to provide a reasonable way for routine recovery of transplutonium isotopes from fuel reprocessing high-level waste solutions. In the laboratory studies performed so far, high Am/Cm recoveries and high decontamination factors from fission and corrosion products have been experienced. DF values measured for the different process stages and overall DF values, as far as they are available, are given in Table II. It is clear that high-level pilot-plant tests will be required before a final decision on full-plant use can be made. Moreover, a number of auxiliary process steps (e.g., medium active waste treatment, solvent clean-up, re-use and recycle of process streams, etc.) have to be worked out.

Table II. Decontamination factors obtained in cold tests of the different process stages. (a) = not determined. (b) = without WD battery.

Element	Denitration stage	DF for Solvent extraction cycle	Ion exchange cycle	Total DF
Cs	negligible	$>10^4$	(a)	
Sr	negligible	$>10^3$	(a)	
Ce	negligible	$>8 \cdot 10^5$	negligible	$>8 \cdot 10^5$
Nd	negligible	$8.7 \cdot 10^2$ (b)	negligible	
Eu	negligible	$1 \cdot 10^3$	negligible	$1 \cdot 10^3$
Y	negligible	$1.3 \cdot 10^3$ (b)	negligible	
Zr	$>10^3$	20	$1.1 \cdot 10^3$	$>2 \cdot 10^7$
Nb	$>10^3$	20		$>2 \cdot 10^7$
Mo	$>10^3$	(a)	(a)	
Ru	ca. 10^2	50	18	ca. $9 \cdot 10^4$
U	(a)	$>6.5 \cdot 10^2$	(a)	
Pu	(a)	$2.5 \cdot 10^2$	(a)	
Np	(a)	$8.0 \cdot 10^2$	(a)	
Fe	1 to 10^2	$4.2 \cdot 10^2$	(a)	

Appendix A

Reference Flowsheet for Am/Cm Recovery from High-Level Waste

Reference values (approximate figures) for the flowsheet presented in Fig. 1 are given. Flows are relative values based on f(1WW) = 100 vol. Abbreviations used: FP = fission product, RE = rare earth (including yttrium), CP = corrosion product, HDEHP = di(2-ethylhexyl)phosphoric acid, TBP = tributyl phosphate, Na_5DTPA = pentasodium diethylenetriaminepentaacetate, HLac = lactic acid, dil = diluent (Alkan $C_{10} - C_{13}$, Gelsenberg AG).

1WW:	27 g/l FPs, 4 g/l CPs, 0.12 g/l Am, 0.01 g/l Cm, 5 g/l Na, 4 M HNO_3, f = 100
FP Precipitate:	Contains essentially the fission and corrosion products without REs, Rb, Sr, Cs, Ba, Ni. Estimated amount: ca. 11 kg of metals per 1000 l of 1WW.
WAF:	5 g/l REs, 3 g/l other FPs (essentially Rb, Sr, Cs, Ba), 0.06 g/l Am, 0.005 g/l Cm, 0.2 g/l Ni, 2.5 g/l Na (all metals as nitrates), pH = 2 ± 0.5, f = 200.
WAX:	0.3 M HDEHP, 0.2 M TBP, 0.002 g/l REs, dil, f = 500.
WAS:	0.02 M HNO_3, f = 134
WAW:	1.8 g/l FPs, 0.12 g/l Ni, 1.5 g/l Na (all as nitrates), pH = 1.1 ± 0.2, f = 334.
WAP=WBF:	2 g/l REs, 0.024 g/l Am, 0.002 g/l Cm, 0.3 M HDEHP, 0.2 M TBP, dil, f = 500.
WBX:	0.05 M Na_5DTPA, 1 M HLac, pH = 3.0 ± 0.1, f = 166

WBS: 0.3 M HDEHP, 0.2 M TBP, dil, f = 166.

WBAC=WDF: 0.072 g/l Am, 0.006 g/l Cm, 0.006 g/l REs, 0.05 M Na_5DTPA, 1 M HLac, pH = 3.0 ± 0.1, f = 166.

WBRE=WCF: 1.5 g/l REs, 0.3 M HDEHP, 0.2 M TBP, dil, f = 666.

WCX: 5 M HNO_3, f = 166

WCRE: 6 g/l REs, 4.9 M HNO_3, f = 166

WCW: 0.3 M HDEHP, 0.2 M TBP, dil, f = 666.

WDX: 0.3 M HDEHP, 0.2 M TBP, dil, f = 125.

WDS: 0.05 M Na_5DTPA, 1 M HLac, pH = 3.0 ± 0.1, f = 42

WDAC: 0.058 g/l Am, 0.0048 g/l Cm, 0.05 M Na_5DTPA, 1 M HLac, pH = 3.0 ± 0.1, f = 208.

WDW: 0.008 g/l REs, 0.3 M HDEHP, 0.2 M TBP, dil, f = 125.

IAF: 0.056 g/l Am, 0.0047 g/l Cm, 0.049 M H_5DTPA, 0.98 M HLac, 0.24 M $NaNO_3$, pH = 0.9 ± 0.1, f = 213.

IAS: 0.1 M HNO_3, f = 10.

IAW: 0.047 M H_5DTPA, 0.93 M HLac, 0.23 M $NaNO_3$, pH = ca. 0.9, f = 223.

IAX: 3 M HNO_3, f = 5.

IAAC: 2.4 g/l Am, 0.2 g/l Cm, 0.5 g/l Na, 0.012 M H_5DTPA, 2.9 M HNO_3, f = 5

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List of Figures

Fig.12. Concentration factors in relation to cross-linkage (the Lewatit SP resins are macroporous).(^{x}Disturbance of the elution step caused too low value of concentration factor).

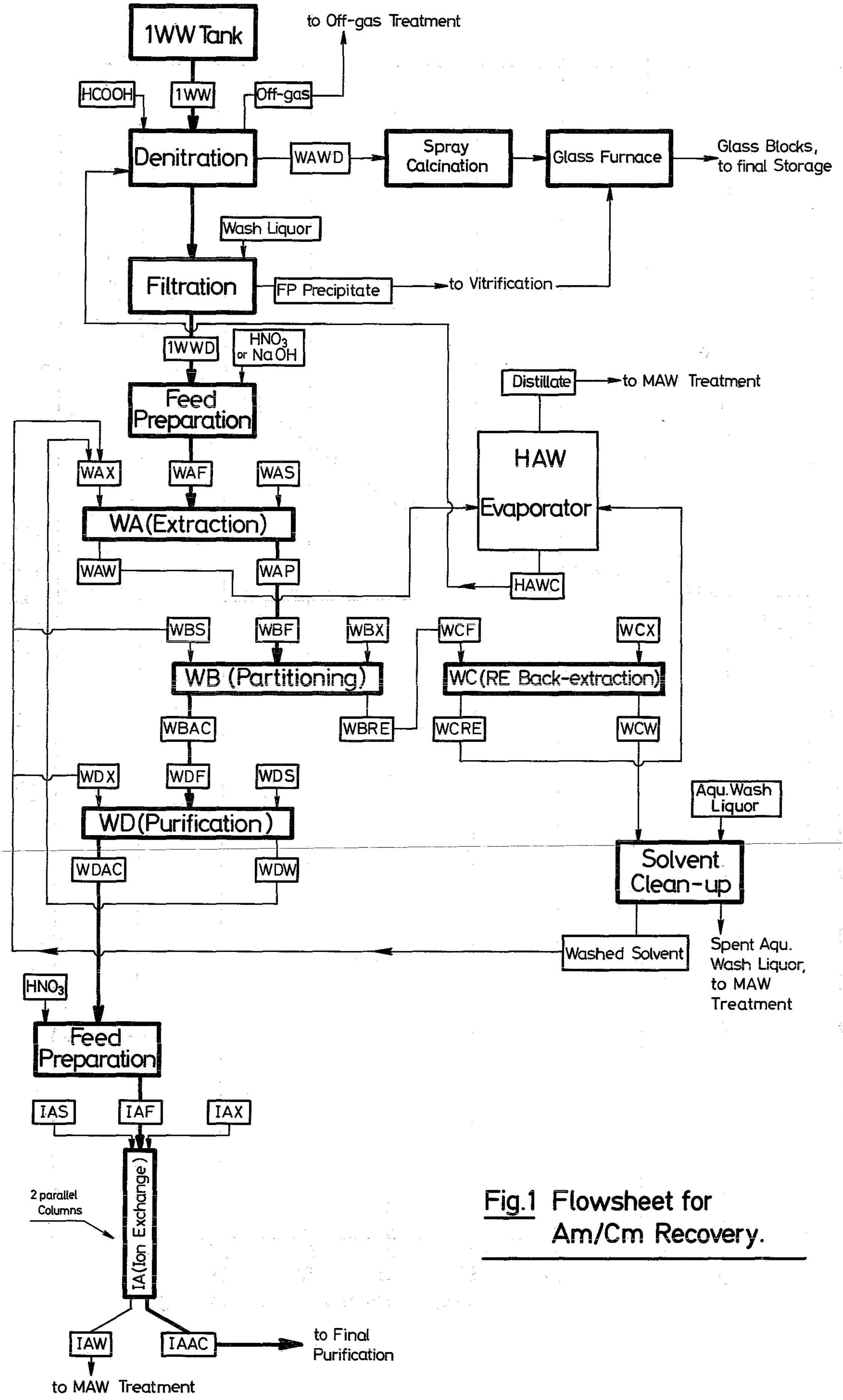

Fig.1 Flowsheet for Am/Cm Recovery.

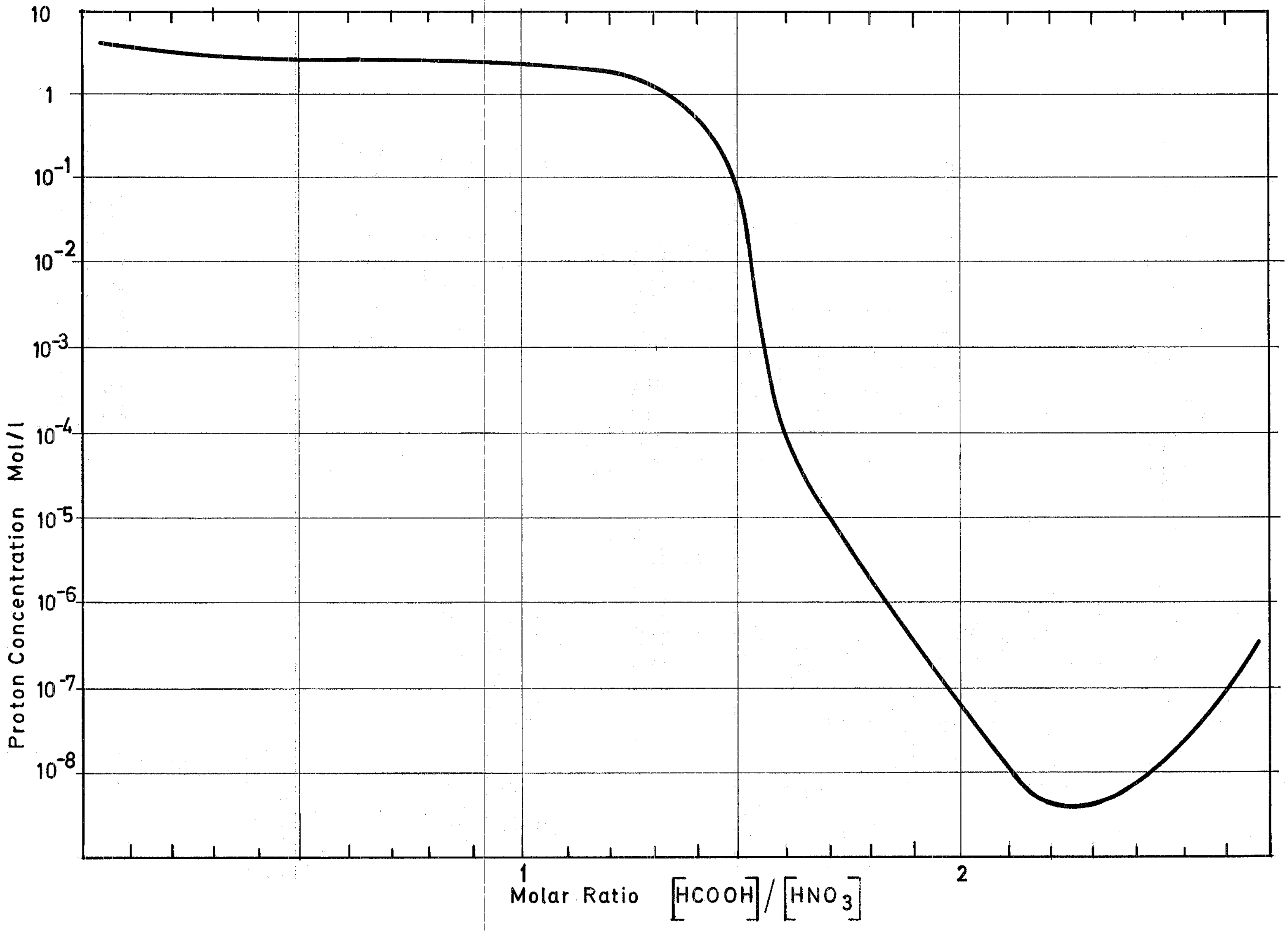

Fig. 2 Hydrogen – Ion Concentration in a 1 WW Solution after Denitration with various $HCOOH/HNO_3$ Molar Ratios.

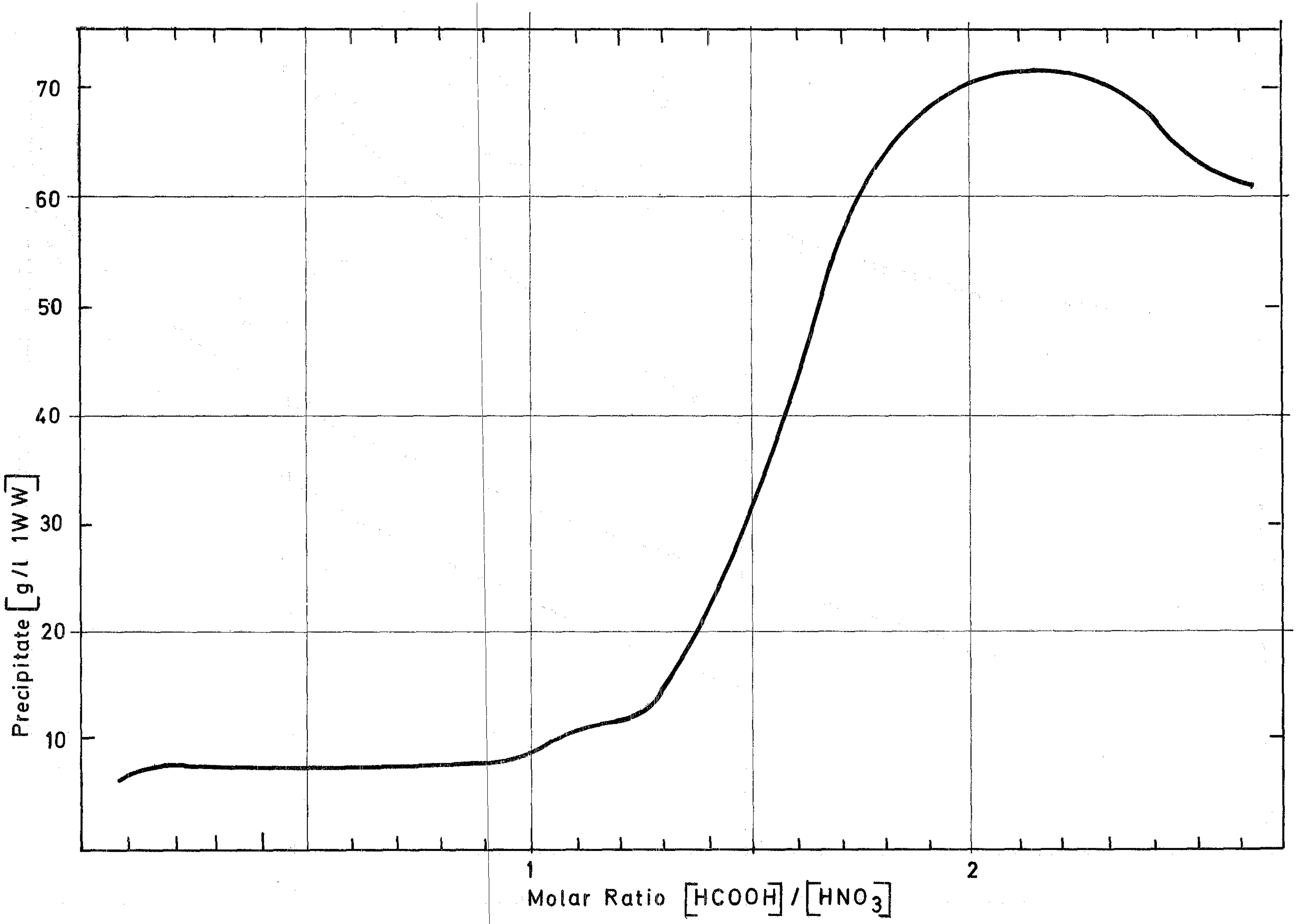

Fig. 3 Precipitate Formation during Denitration of 1WW with various HCOOH/HNO$_3$ Molar Ratios.

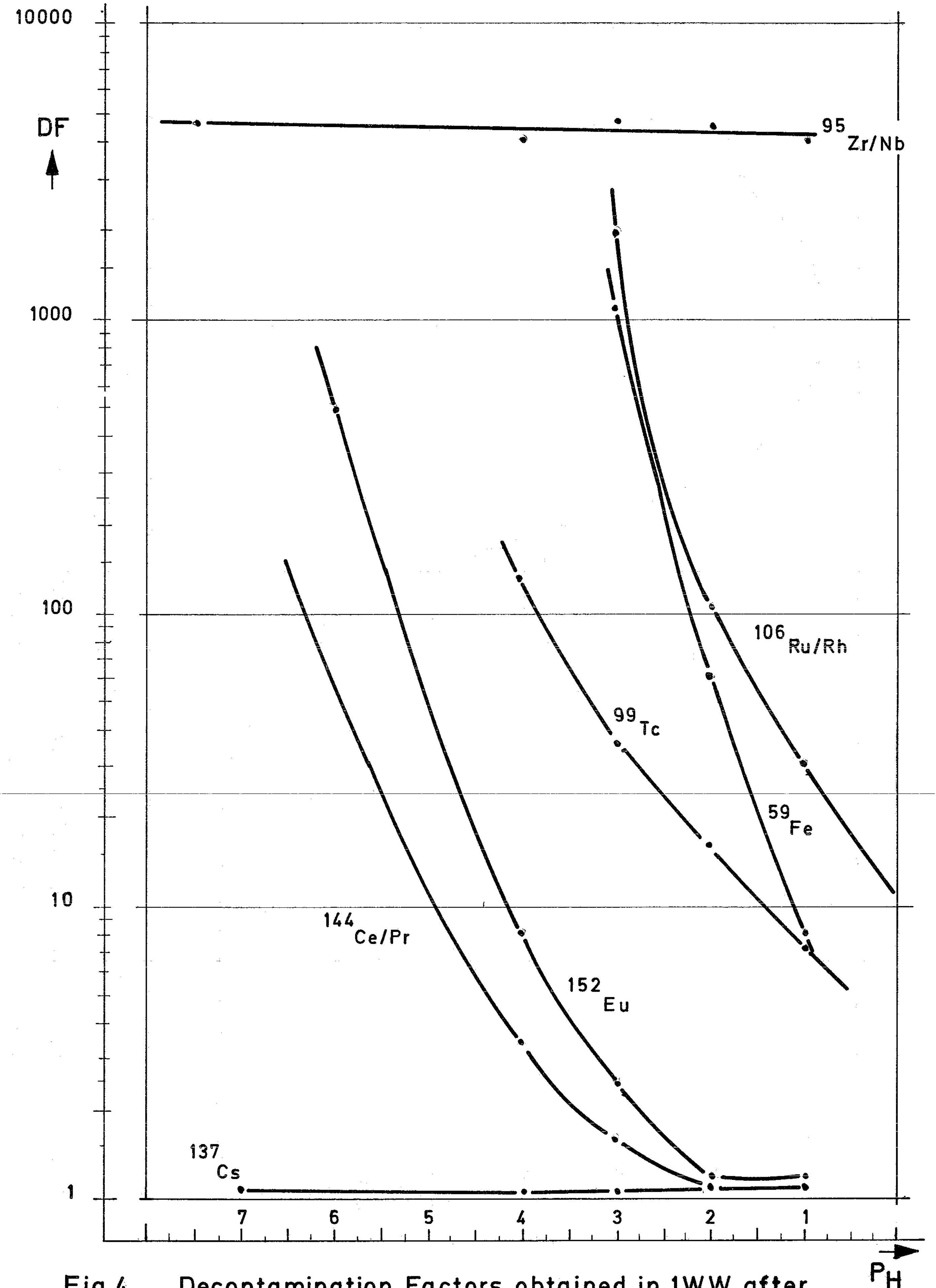

Fig. 4 Decontamination Factors obtained in 1WW after Denitration and Reacidification.

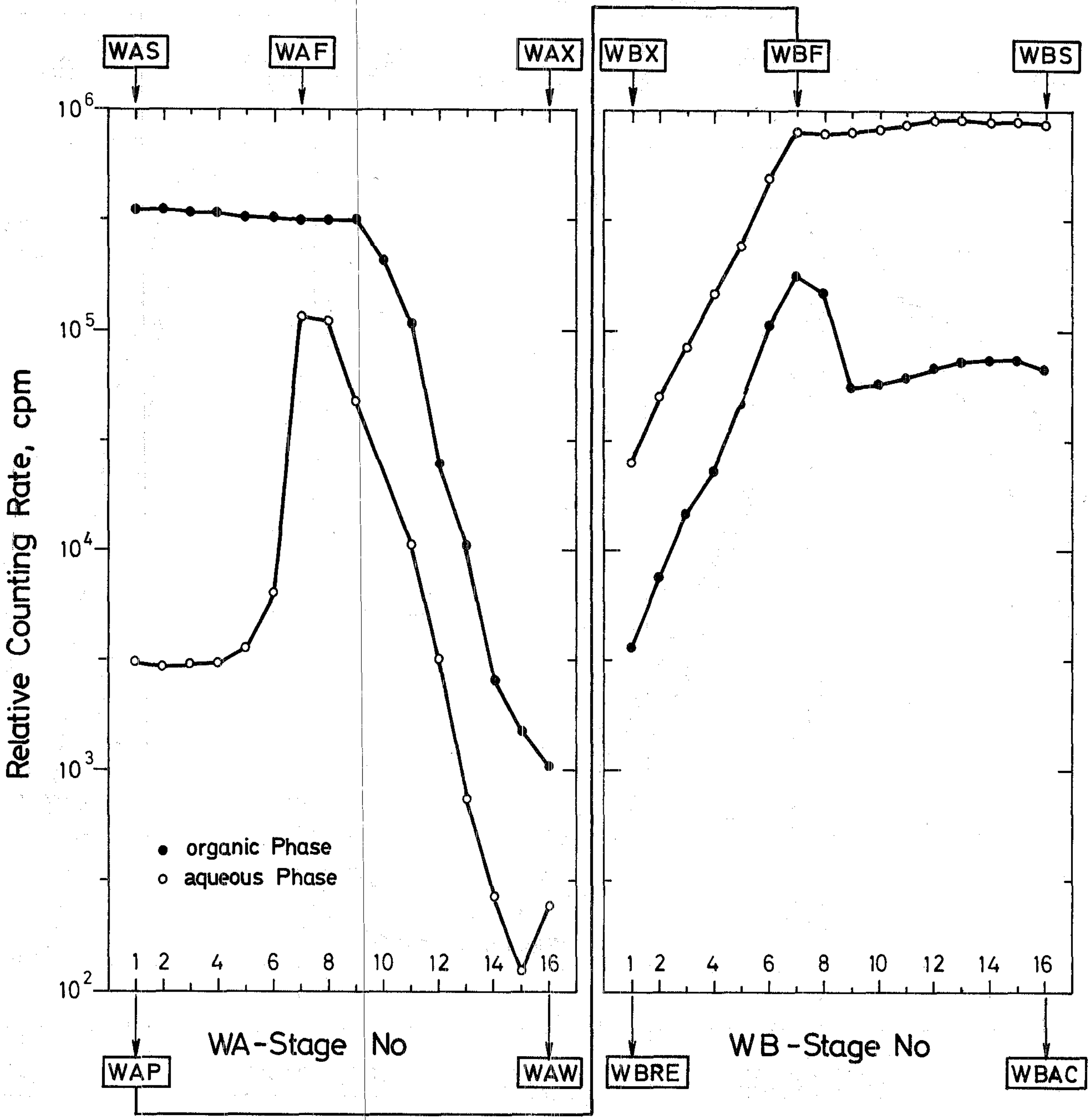

Fig. 5 Concentration profile for Am(III) in the WA and WB batteries.

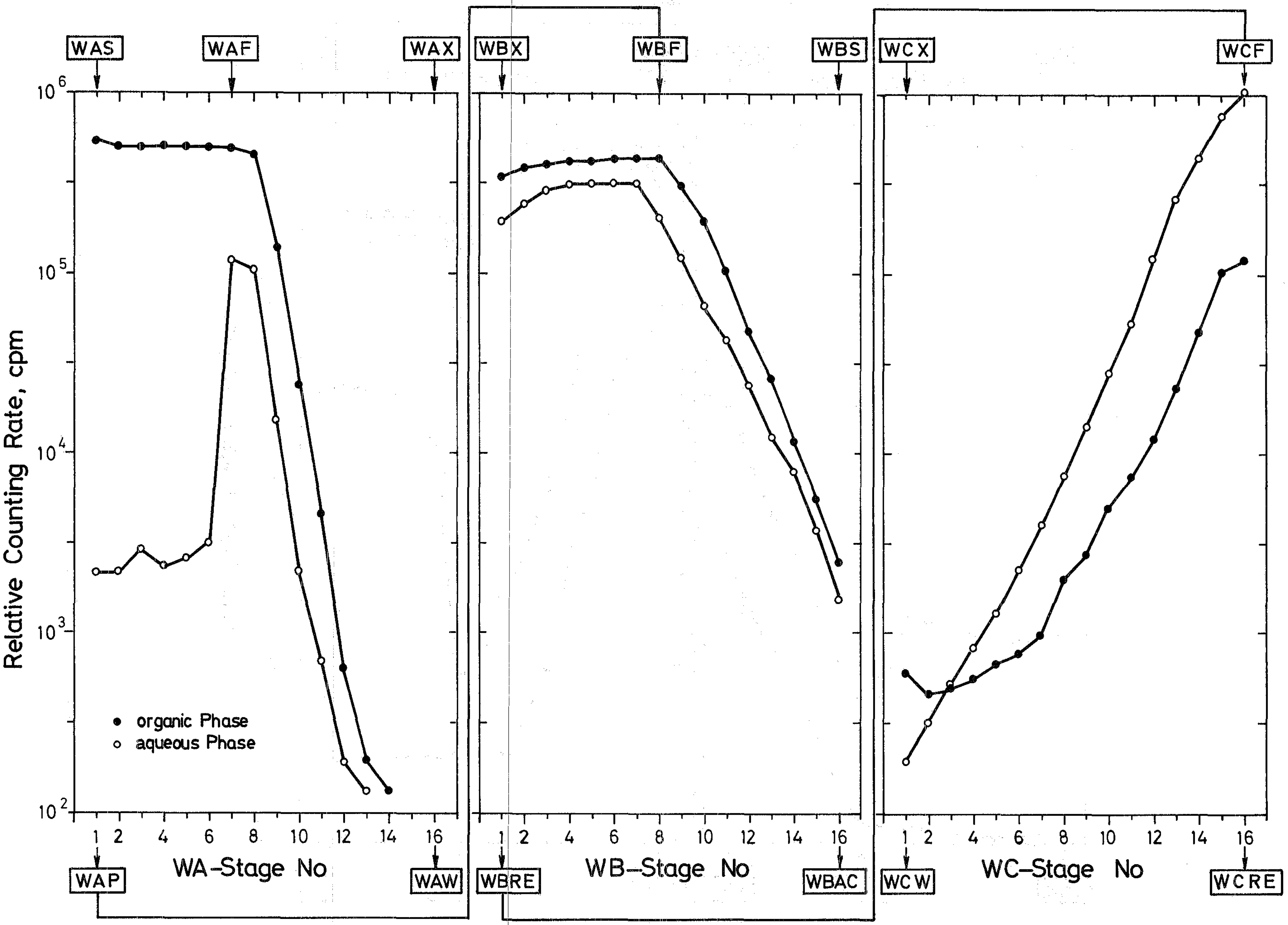

Fig. 6 Concentration profile for Nd(III) in the WA, WB, and WC batteries.

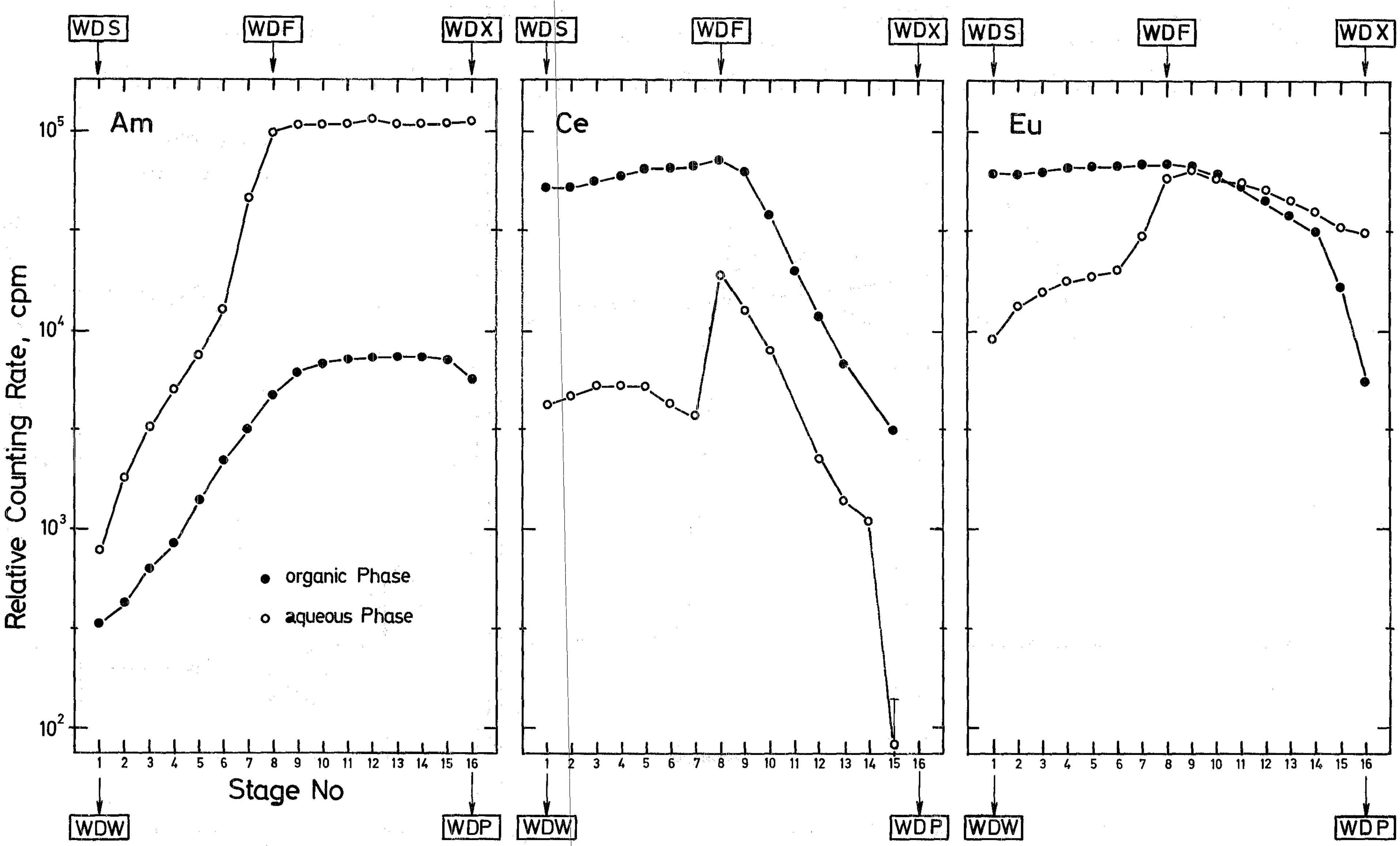

Fig. 7 Concentration profile for Am(III), Ce(III), and Eu(III) in the WD battery.

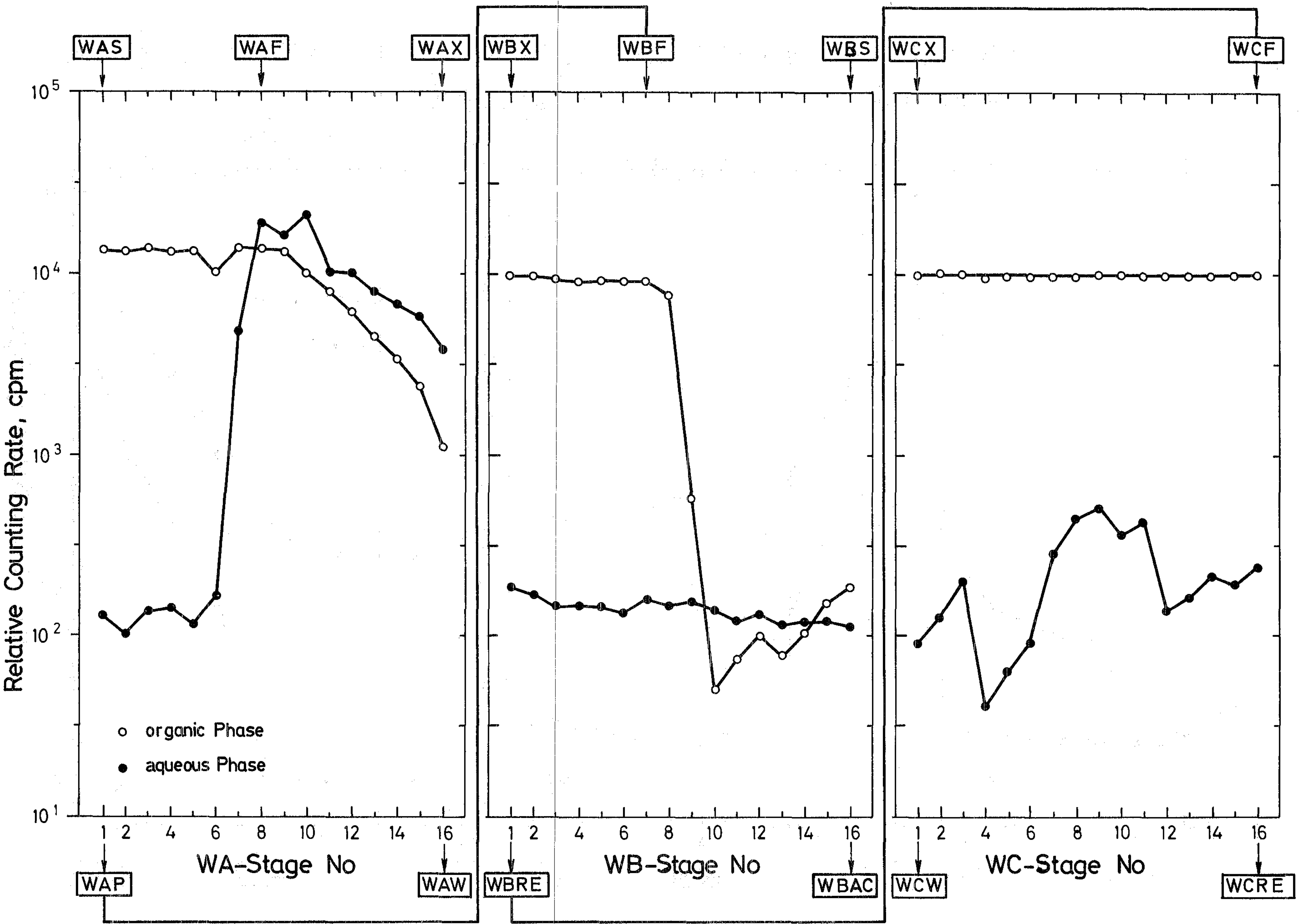

Fig. 8 Concentration profile for Fe(III) in the WA, WB, and WC batteries (Mixer-settlers)

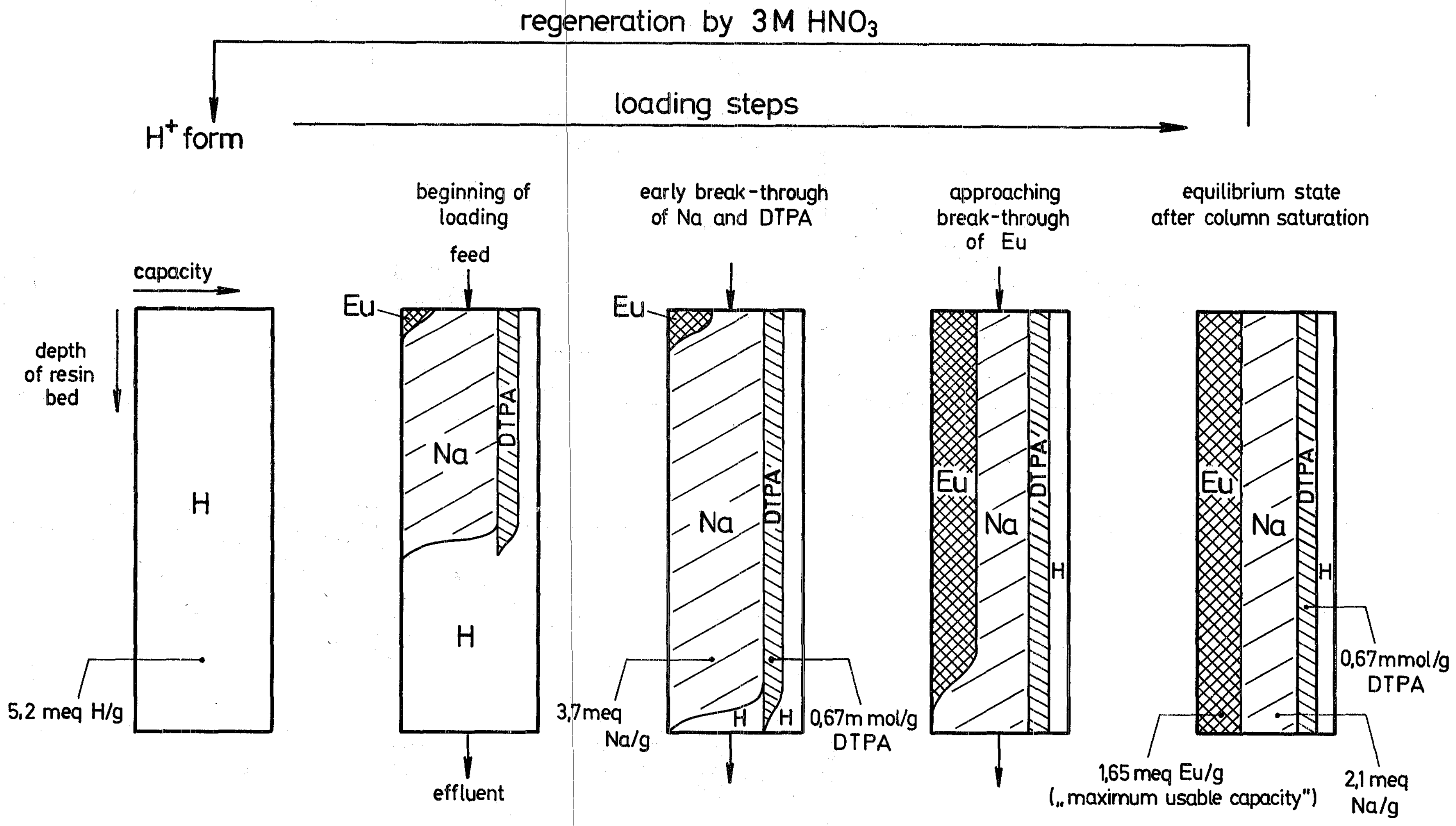

Fig. 9 State of cation exchange column at different stages of loading

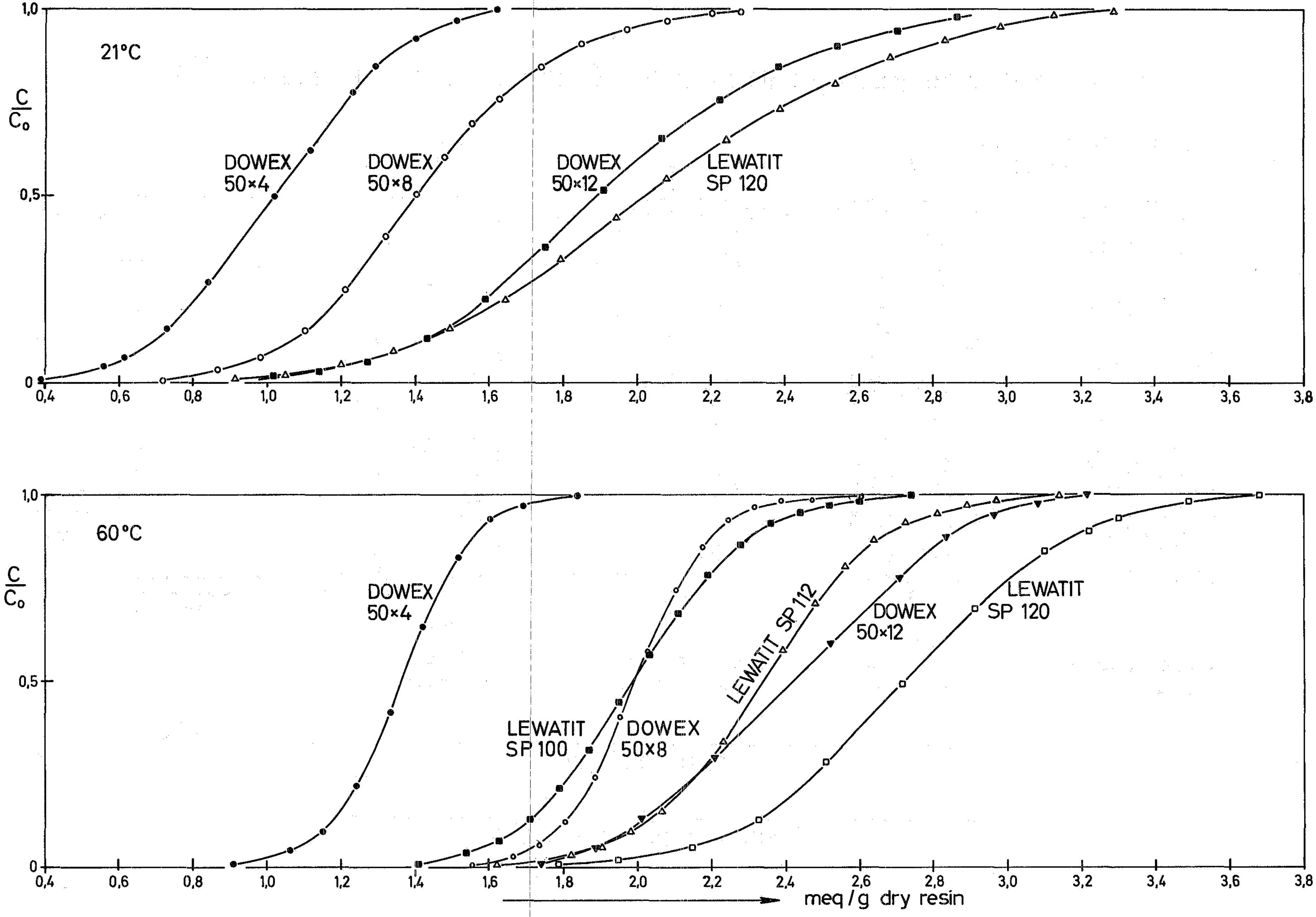

Fig. 10 Break-through curves (for conditions, see Table)

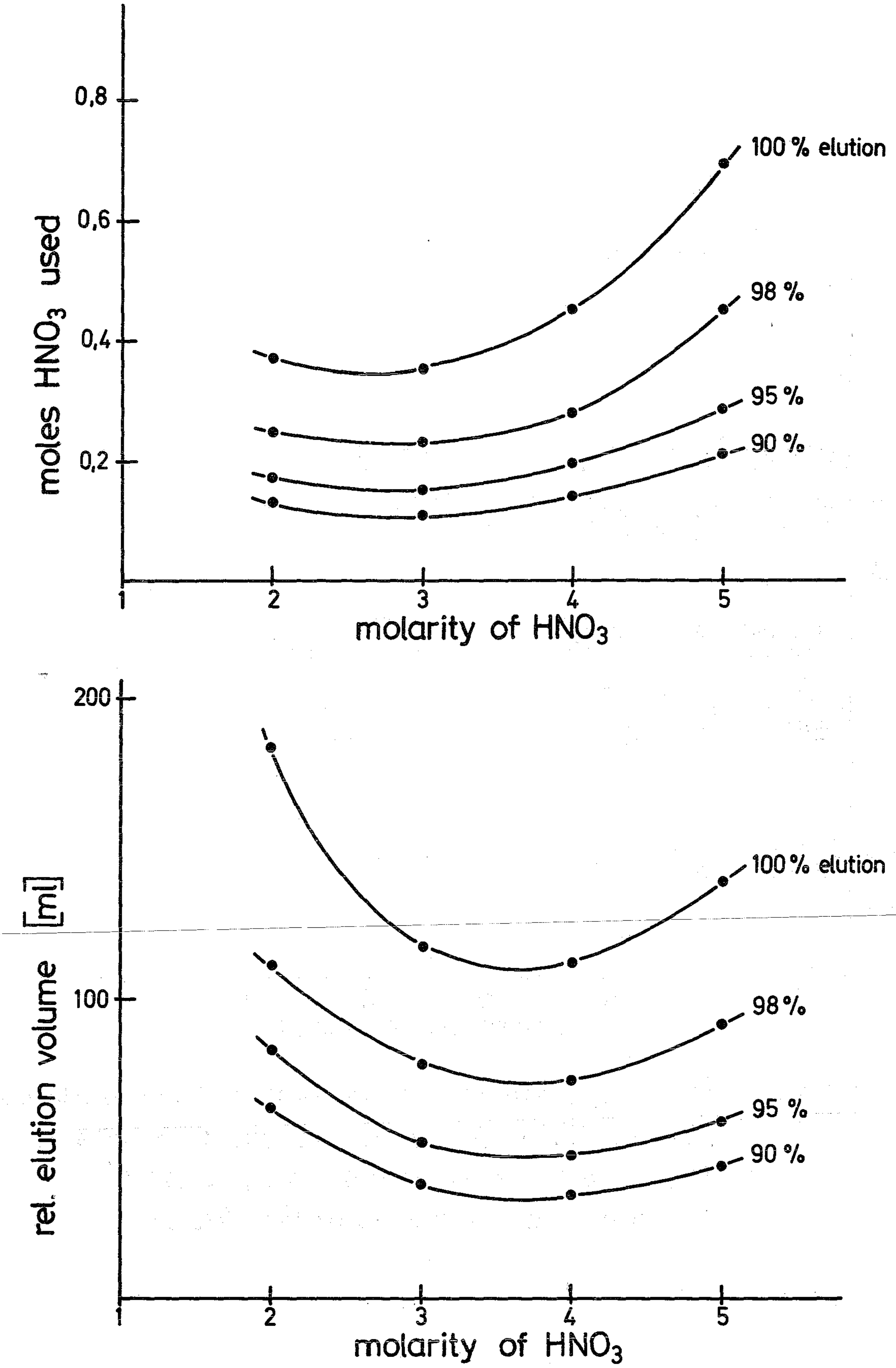

Fig. 11 Amount of HNO_3 (moles) and relative volume of solution necessary for elution of Eu^{3+} at different HNO_3 concentrations (flow rate 1.0 ml/cm min, temperature 21°C)

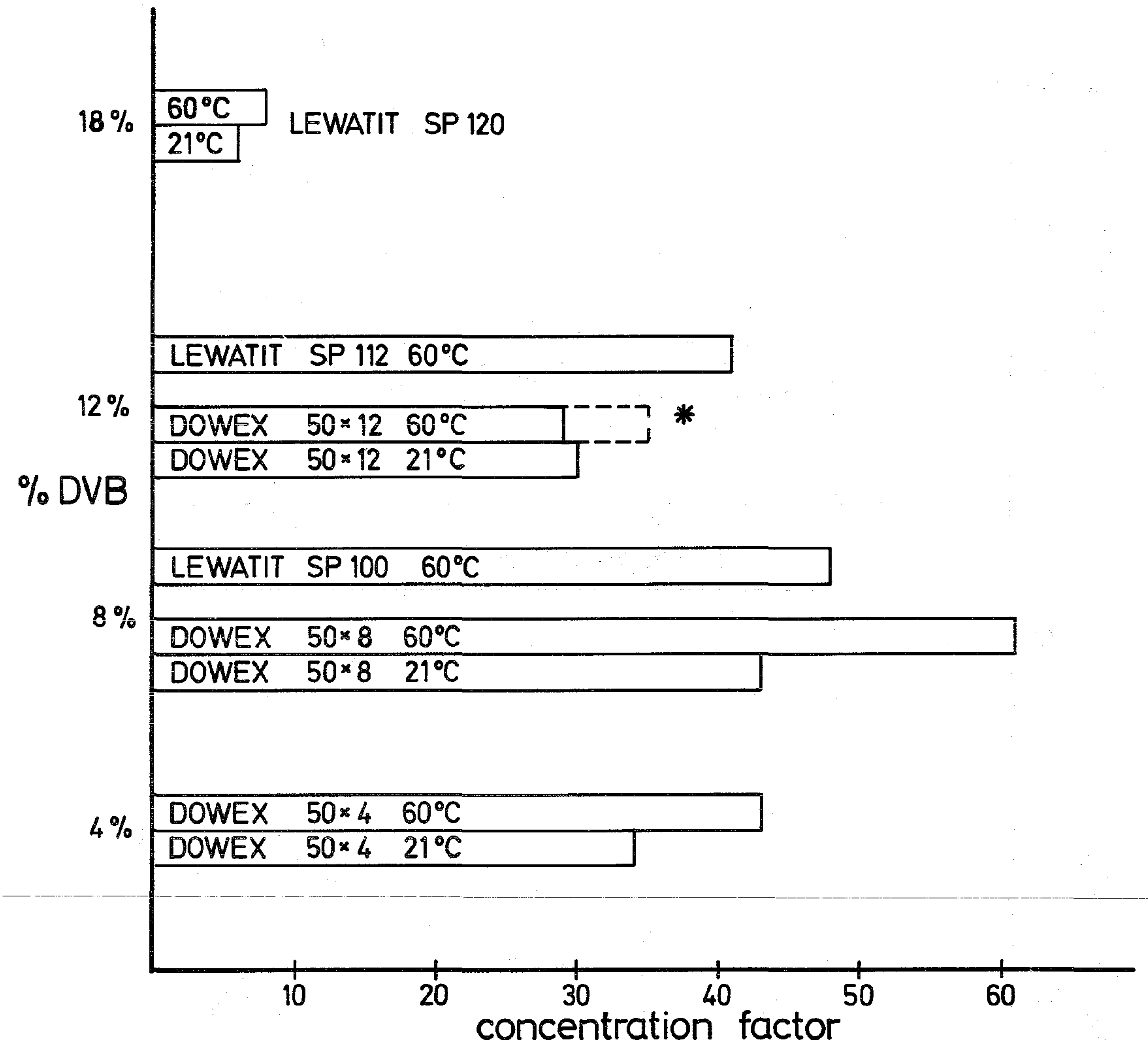

Fig. 12 Concentration factors in relation to cross-linkage. The LEWATIT SP resins are macroporous.
(* Disturbance of the elution step caused too low value of concentration factor)