Use of Separator with Regular Granular Filling in Extraction Processes - 12209

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ABSTRACT

The separator with a regular granular filling has been developed to be used for the operations of the removal of carbon-black impurities from the aqueous flow; the intracycle regeneration of back-extractants; and the concentrating back-extraction of plutonium in the extraction SNF reprocessing technology. The process conditions for those operations have been experimentally tested using this separator.

KEYWORDS: separation, mass transfer, emulsion, extractant regeneration, plutonium back-extraction, salt-free reagents

INTRODUCTION

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Extraction technologies for reprocessing of spent nuclear fuel (SNF) are an important component of a closed nuclear fuel cycle.

The high degree of purification of the recycled nuclear material requires several consecutive cycles of extraction reprocessing, that besides the direct cost of holding them, rising new volumes requiring disposal of liquid radioactive waste (LRW). Among the factors limiting the possibility of treatment of the extraction cycle is very important emulsion carryover of "polluting" phase with product flows.

Upto the moment the extraction technology has developed only one approach to minimizing pollution flow of extraction cycle by emulsion carryover. This approach is to manage the types of emulsions formed in the extractor by small recirculation flow in the mixing chamber of the extractor.

However, this method roughly increasing the load on the extractor is only applicable to discrete devices and is ineffective if the system has stabilizers of undesirable emulsion type.

Thus, the development of the separation process flow of extraction technology in order to avoid contamination of emulsion and its devices design using a new type - separators with regular granular is an important task.

Any contacting of the aqueous and organic phases in the extraction process is accompanied by interphase mass transfer, hence the separator-apparatus with an effective separation phase is also mass-transfer extraction apparatus.

In the development of flow separation techniques to remove micro-emulsion carryover as a way to impact on the purified stream it was adopted forcable coalescence of microemulsion particles forming large drops with further subsequent gravitational separation of drops from the continuous cleansed phase flow. Forced coalescence produced during filtration flow through the layer with a regular granular.

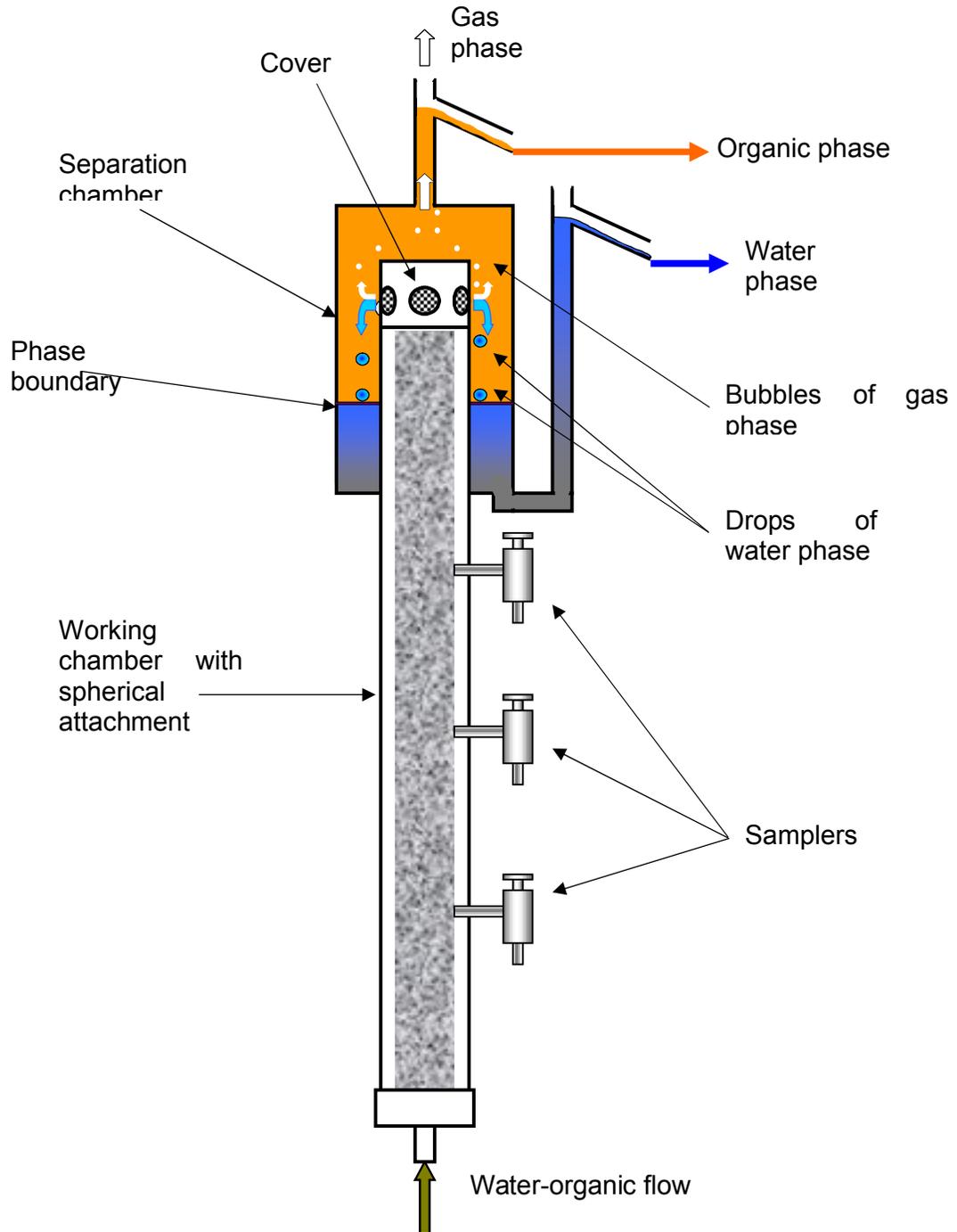Picture 1 shows a schematic diagram of the separator.

Fig. 1 –The separator with the upper chamber dividing for mass transfer processes in gas production

Forced coalescence of emulsion particles passing the flow through the separating layer is due to inhibition of the particles as they move in the intergranular (pore) space,

increasing the concentration of particles and subsequent coalescence. Further, the motion is to be cleansed flow through a porous layer represents the motion of two solid phases, moving at different speeds. At the exit of the separator is one of the phases (purified) continues to move without breaking the flow, the flow of the second impurity, the phase is broken into large drops (fig. 2).

- water phase
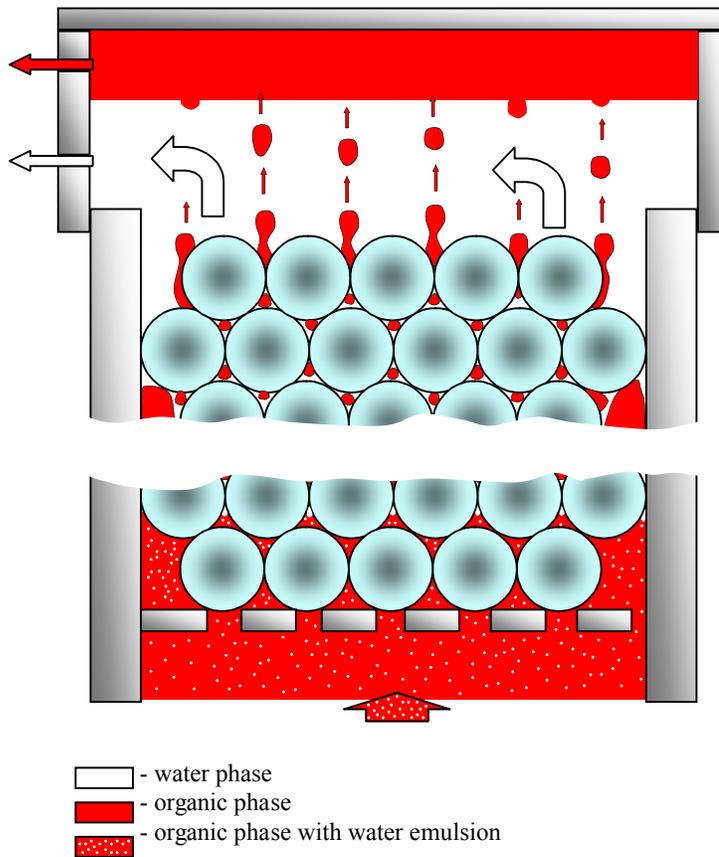- organic phase
- organic phase with water emulsion

     Fig. 2 - Schematic diagram of the coalescence of emulsion in the granular layer of the separator with spherical attachment and separation of the separation chamber.

     Next, the two phases, cleaned and impurity, pass through a separation chamber equipped with a water seal and removed from the apparatus, the separator.

     To improve the separation efficiency of fine particles in the inlet section separating layer together with the treated stream supplies flow stream with a small auxiliary of that phase, which is an impurity in the purified stream. The role of the helper flow is to increase the concentration of emulsion particles in the pore space separating layer and create a front of the entrance section of the emulsion layer of the separator with a high content of dispersed phase, like "alluvial strata" in the filtration of fine solid dispersions. But, unlike the "alluvial strata," continually revolving and moving with the flow through the treated layer separating. Both factors provide a complete coalescence of microemulsion push dirt with an emulsion formed by the auxiliary flow, followed by separation of the phases in the separation chamber of the separator (fig. 3).
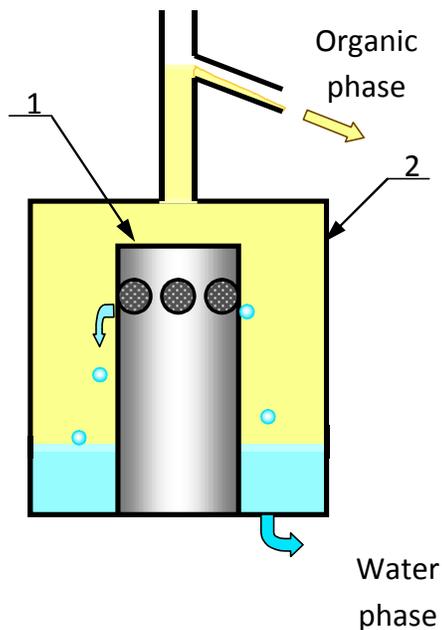
Fig. 3 - Output of organic and water phases of the working chamber (1) and phase separation chamber (2) of the separator

On the structure of the phase separator flows closest to the film extractor [1]. These devices share one important and fundamental feature - the mass transfer occurs without dispersing one phase to another and subsequent coalescence of emulsions in working chamber, i.e. without degeneration of the interface. Even when applying the auxiliary flow in the form of an emulsion in a relatively short distance separating layer in the pore channels formed by two continuous flow of liquid phases with coaxial movement along the channel [2].

The lack of degeneration of the interface allows you to operate at high flux ratios of the phases. Thus, the mass transfer in porous separator layer occurs at a highly nondegenerate and the interface.

A feature of this surface is that it moves together with the two-phase flow, as well as the velocity of the phases are very different because of the high retention of the granular layer, then a jump to the GRF flow rates (Fig. 4). This fact has important implications - separator can clean up the stream not only from the emulsion of pollution, but also from other heterophase inclusions, including suspension.

Flow pattern of organic (O) and water (W) phases in the model channel is shown in Figure 4.
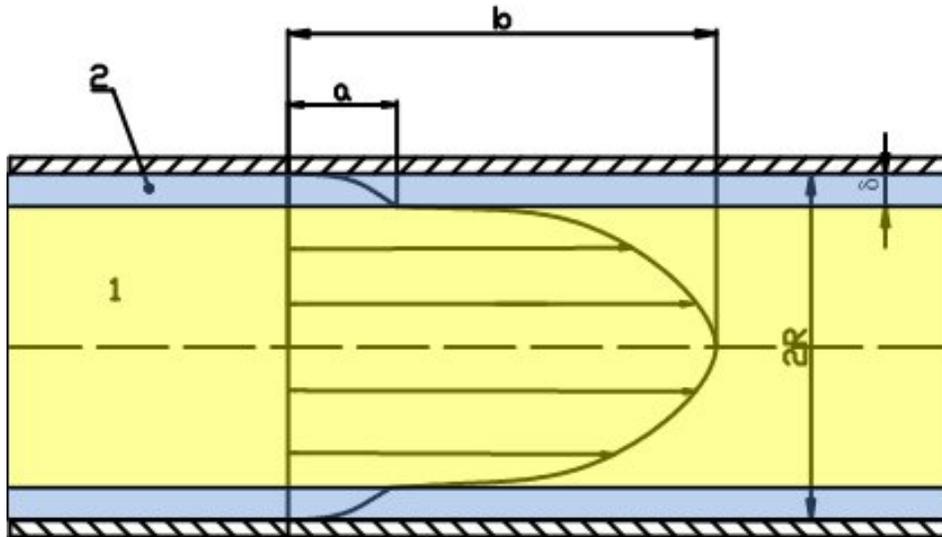
Fig. 4 The scheme of flow of immiscible fluids 1 (organic phase) and 2 (water phase) in a model channel.
R - the effective radius of the channel, δ - the thickness of the annular channel. a- velocity vector of a small-flow, b - velocity vector of a large stream.

In the extraction technology of reprocessing of irradiated nuclear fuel one of the key operations is stripping of plutonium from the uranium-plutonium organic extract in water solution.

EXPERIMENTAL METHOD

Concentrating Reextraction of Plutonium

Features extraction apparatus for concentration of plutonium due to compression reextracted flow limited. Usually the flow ratio for plutonium stripping operation is less than O:W = 10:1. Increasing the concentration of multiplicity in the transition to O:W>(15-20):1 can lead (even in a multi-extractor) to unbalance the process due to entrainment of the water phase emulsion (strong plutonium solution) with the uranium extract plutonium and the incompleteness of stripping, disruption of stability extractors work at a small flow of water solution. Consequently, stable holding of concentrating process reextraction of plutonium in the device makes it possible to operate at flow ratios of the phases is greater than 20:1 is very promising.

An experimental verification of the possibility of concentrating reextraction of plutonium in the separator with the ratio of 30:1 flow phases was made. The following was considered as reextracted systems:
  a) $[N_2H_4]$=0,5 mol/l, [Diethylene triamine pentaacetic acid (DTPA)]=3 g/l, $[HNO_3]$= 0,2 mol/l
  b) [U(IV)]=74,5 g/l, $[N_2H_4]$=0,53 mol/l, $[HNO_3]$= 0,3 mol/l.

The both case feed flow was an organic extract of the: [Pu]=1 g/l, [U(VI)]=102 g/l, [Tc]=55 mg/l, $[HNO_3]$<0,1 mol/l. The parameters of the process flow of the concentrating reextraction

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concentrating reextraction were – 700 ml/h, O:W=30:1, t=40 °C.

The results are shown in Tables I and II.

Table I - Concentrations of Pu and Tc in the Product Solutions of Stripping Pu Process with Hydrazine - DTPA

т, h	O:W in fact	Plutonium Reextract		After Separator Extract		
		Pu, g/l	Tc, mg/l	Pu, g/l	Tc, mg/l	U, g/l
2	29	19,5	630	0,096	13,2	-
4	27	22,4	820	0,064	11,4	-
6	30	21,0	905	0,038	7,0	-
8	32	22,9	640	0,062	21,0	-
10	33	29,5	620	0,109	17,3	-
12	33	28,8	770	0,228	19,0	-
14	32	27,8	770	0,315	18,3	-
Average		28,1	770	0,192	19,0	-

In the second experiment evaluated the effect on the amount of load flow 2 $m^3 / (m^2 \cdot h)$ and 3 $m^3 / (m^2 \cdot h)$ at a temperature of 30 ° C separator with a depth of reextraction of plutonium and process stability.

Table - II Concentrations of Pu and Tc in the Product Solutions of Stripping Pu Process with U (IV) - N_2H_4

Load Flow $m^3/(m^2 \cdot h)$	т, h	Plutonium Reextract		After Separator Extract	
		Pu, g/l	Tc, mg/l	Pu, g/l	Tc, mg/l
1,9	2	23,6	561,0	0,048	3,6
	4	29,1	528,0	0,038	-
	6	26,1	651,0	0,038	-
	8	23,6	525,3	0,032	5,0
	10	26,0	495,0	0,033	4,0
	12	26,5	725,0	0,048	-
2,7	2	20,4	624,8	0,040	9,2
	4	23,4	560,0	0,050	-
	6	23,2	960,0	0,045	10,2
	8	23,1	802,0	0,043	11,0

The stripping plutonium process of uranium (IV) solution showed a number of advantages over the reextraction solution hydrazine-DTPA, the main ones being the completeness of extraction of plutonium and technetium in reextract, the phase stability of plutonium reextract and process stability.

Regeneration of the Extractant

Another promising technological operation that is sensitive to the emulsion carryover in extraction technology and requires a high ratio of flows is the operation phase of the extractant regeneration.

The initial stage of work was to study the influence of heigh separating layer to the cleaning efficiency of extractant from dibuty phosphoric acid. An aqueous solution of soda was used as a regenerating system. The results are shown in Table III.

Table III. Recovery of Circulating Extractant Containing [DBFK]= 340 mg / l, [U] = 15 mg / l, [HNO3] = 0,005 mol / l, the separator with 1.02 mol / l Na_2CO_3 solution at about O:W = 30:1

Height Separating Layer, mm	[DBFK]organic, mg / l	[DBFK]water, mg / l	"Distribution Factor" DBFK
390	25,8±2	8940	$2{,}9\cdot10^{-3}$
650	18,1±2	9155	$2\cdot10^{-3}$
900	15±2	9325	$1{,}6\cdot10^{-3}$

It is noted that the height separating layer has little effect on the cleanup of DBFK, but affects the value of the residual ash emulsion (Fig. 5).

a)   b)
Fig. 5 Separation chamber phase of the separator with a working height of 390 mm separating layer (a) and 900 mm (b)

As an alternative to soda solution there were considered so-called salt-free reagents, the thermal decomposition of nitrate salts, which produce only gaseous products of decomposition. As the regenerating agent was chosen ethylenediamine (EDA) in the form of carbonate mud. Carbonate solution is prepared by treatment of an aqueous solution of EDA certain amount of carbon dioxide in the absorber.

To select the composition of the regenerating solution it was carried out several

experiments in static conditions, in which were installed:

– Depending on the pH solutions of the concentration of the EDA, as well as on the concentration of carbonate ion in solutions of 0.5 and 1.0 mol /l EDA;

– influence the degree of EDA carbonation solutions to a depth of extraction dibutilfosfornoy acid from extractant;

- the specific load on the separating layer.

Table IV. Cleaning of the Extractant from DBFK by Carbonized Solution of 0.5 and 1 mol /l EDA in the Separator at t = 45 C° and [DBFK] = 500 mg / l.

O:W	Load, $m^3/m^2 \cdot h$	EDA, mol / l (pH solution)	Y_{dbfk}, mg/l	α_{dbfk} in separator	Residual Emulsion, %
30:1	1,9	0,5	4,6	$3,9 \cdot 10^{-4}$	0,006
	2,5	(9,7)	5,8	$6,4 \cdot 10^{-4}$	0,006
	3,1		6,9	$4,9 \cdot 10^{-4}$	0,008
	4,3		7,5	$6,9 \cdot 10^{-4}$	0,014
30:1	1,9	1	4,6	$4,2 \cdot 10^{-4}$	≤0,005
	2,5	(11,0)	5,3	$4,0 \cdot 10^{-4}$	≤0,005
	3,1		5,5	$3,9 \cdot 10^{-4}$	0,006
	4,3		6,0	$4,2 \cdot 10^{-4}$	0,010
20:1	2,5	5	5,5	$5,6 \cdot 10^{-4}$	0,006

Received data show that the regeneration of the extractant by carbonate solutions of ethylenediamine in the separator is comparable to the regeneration of soda solutions. The distribution coefficients between DBFK inferred from the separator is the organic and water flows varied from $3,9 \cdot 10^{-4}$ to $6,9 \cdot 10^{-4}$. The residual concentration of uranium in organic phase after regeneration with at O:W=(20÷30) was ≤ 0,5 mg / l in water regenerate - up to 0.9-1.3 g /l.

Working with a load of 1,9-2,5 $m^3/(m^2 \cdot h)$ full phase separation reach in the separator. With the increase of the specific load up to 4-4,3 $m^3 / (m^2 \cdot h)$ appears insignificant entrainment of the water phase emulsion of 0.01% vol.

CONCLUSION

Thus, the separator for operations of concentrating reextraction of plutonium and regeneration recycling extractant allows to:

– derive plutonium from the organic stream into an extremely concentrated form, reducing the amount of liquid radioactive waste (raffinate, decantate) plutonium branches to a minimum;

– controlling the processes of concentration of uranium and plutonium, get uranium-plutonium product with required concentration and the amount of metal with required ratio;

– eliminate contamination of the emulsion flows;

– simplify the purification of uranium from the extract plutonium;

- test results showed that when working on a solution "hydrazine-DTPA" in the separator it is achieved the extraction of plutonium reextract - 82% TC - at 56-65%. With the transition to the reextraction by solution «U (IV)-hydrazine," of plutonium into reextract

increases to 95%, technetium - up to 83%;

- regenerate the extractant recycling minimizing the amount of liquid radioactive waste generated as a technology used as well as alternative "salt-free" systems;

- it is found that the regeneration of the extractant solution of soda and EDA in the separator at a load of 3.2 m3 / (m2 • h) it can be achieved almost complete phase separation. Residual carryover of 0,005-0,006% emulsion is on the verge of analytical detection.

- It is shown that in the depth of separation phase a separation of the extractant regeneration of quality superior is more than 5-7 times to the extraction.

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