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## MAKROFOL KG MICROFILTERS WITH LARGE PORES APPLIED FOR THE SEPARATION OF TWO DIFFERENT LIQUID PHASES (EMULSION)

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**Abstract**—Makrofol KG nuclear track microfilters have been successfully applied to separate two different and strongly mixed liquid phases (emulsion). The organic and the inorganic phase of the two emulsions used in this work were: (a) 48% tributyl phosphate (TBP) in kerosene and 2 N HNO<sub>3</sub> and (b) 48% TBP in kerosene and 13 N HNO<sub>3</sub>, respectively. Makrofol KG filters having pore sizes  $k$  in the interval of  $2.3 \leq k \leq 8.0 \mu\text{m}$  were used to get under some well defined conditions a complete extraction of the organic phase out of the first emulsion (a), and filters with a range of pore sizes  $k$  of  $5.5 \leq k \leq 10.5 \mu\text{m}$  were employed to obtain a complete separation of the organic part of the second emulsion (b).

### 1. INTRODUCTION

It has been shown that Mica Nuclear Track Microfilters (Sial *et al.*, 1987) and Kapton Nuclear Track Microfilters (Ganz *et al.*, 1988) are suitable for the separation of two strongly mixed different liquid phases of emulsions. Here we report on similar separation experiments using nuclear track microfilters made with a thin polycarbonate plastic named Makrofol KG. The filter is produced by irradiation of the thin film with fragments from the U-235 fission which is induced with thermal neutrons extracted in the beam-hole BH-10 of the IEA-R1, 2 MW pool type research reactor at IPEN, São Paulo.

The fission fragments produce tracks across the entire thickness of the plastic foil which may be selectively attacked (etched) by an appropriate chemical solution (6.25 N KOH) producing a fine channel hole with up to 10.5  $\mu\text{m}$  diameter in the bombarded material (Yamazaki and Geraldo, 1992). The hole diameter is thus controlled by the etching time and the density of holes is determined by the particle dose or irradiation time (Fleischer *et al.*, 1975). For illustration, Figs 1 and 2 show SEM microphotographs of a typical sample of Makrofol KG microfilter produced by the technique described above. As can be seen the pores are cylindrical and practically the same size. Also apparent is the smoothness of its surface and of the inner walls of the pores.

For some industrial processes there is the problem of emulsion constituent separation when the emulsion is a mixture of two different liquids having different densities and viscosities (Sial *et al.*, 1987). In radiochemistry processes such as in radioanalytical work as well as in nuclear fuel reprocessing it is

often necessary to extract transuranium ions out of highly concentrated uranium solutions. To perform liquid–liquid extraction, one has to bring the aqueous uranium solution (which also contains the transuranium ions) into contact with an organic solution of a suitable extracting compound. Then the organic liquid, now containing the transuranium ions, has to be separated from its emulsion with the aqueous uranium solution (Ganz *et al.*, 1988).

Makrofol KG filters have been shown to be suitable for the separation of organic liquids from emulsions they form with aqueous liquids. As examples of applications we chose two emulsions that have been employed in nuclear fuel reprocessing:

(a) emulsion A: 2 N HNO<sub>3</sub> (inorganic phase) and 48% tributyl phosphate (TBP) in kerosene (organic phase). Tri-*N*-butyl phosphate can be used to extract and separate the trivalent rare-earth nitrates moderately well and high partition coefficients can be obtained under suitable conditions. Experiments at varying TBP concentrations indicate the formation of Y(NO<sub>3</sub>)<sub>3</sub> · 3TBP and Ce(NO<sub>3</sub>)<sub>3</sub> · 3TBP. Yttrium, cerium, and other rare-earth fission products may thus be removed from fissile materials (Scargill *et al.*, 1957);

(b) emulsion B: 13 N HNO<sub>3</sub> (inorganic phase) and 48% tributyl phosphate (TBP) in kerosene (organic phase). Thorium nitrate is more readily extracted by TBP than are the rare earths, but it is less extractable than plutonium(IV) nitrate. The extraction is sufficiently good with fairly concentrated TBP for the separation of thorium from most of the fission products. It extracts as a dissolvate, Th(NO<sub>3</sub>)<sub>4</sub> · 2TBP (Hesford *et al.*, 1957).

The extraction of the organic phase out of these emulsions was possible with Makrofol KG track

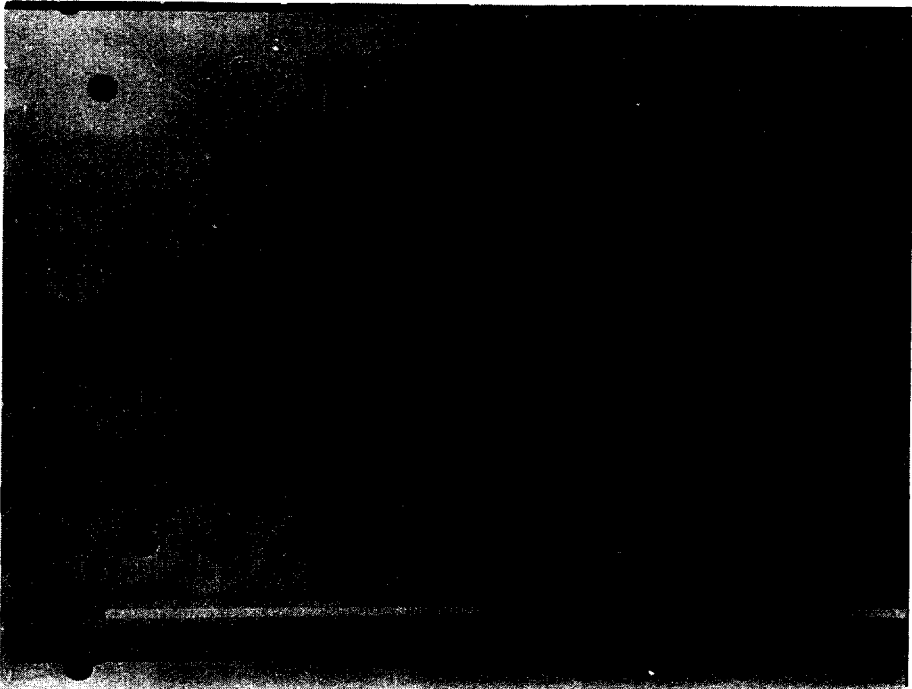


FIG. 1. A typical sample of Makrofol KG microfilter used in the present work. Pore size:  $8.0 \mu\text{m}$ . Pore density:  $10^5$  pores/cm<sup>2</sup>.

microfilters having different pore sizes. The throughput of the pure phases was also determined. It is much larger for TBP in kerosene as compared with that of  $\text{HNO}_3$ . This effect did not change, even if the two liquids were poured into the cup as an emulsion.

## 2. EXPERIMENTAL

In this experiment filters having thicknesses of  $10 \mu\text{m}$ , active areas of 3.5 cm in diameter, pore sizes from 2.3 up to  $10.5 \mu\text{m}$  and porosities from 3 to 9% were used.



FIG. 2. Lateral view of a Makrofol KG microfilter sample. Pore size:  $5.5 \mu\text{m}$ . Pore density:  $10^5$  pores/cm<sup>2</sup>.

The filtration unit consisted of two glass cups inverted over each other, a filter holder and a collection flask. The Makrofol KG filters were mechanically supported by a polycarbonate plastic Makrofol E (200  $\mu\text{m}$  thickness) grid. Figure 3 is a schematic representation of the extraction apparatus used. The filter holder is placed between the two glass cups and six rubber bands hold the arrangement which was tightened by using O-rings and the force of the rubber bands. The opening of the upper cup is equal to the filter active area, i.e. having an inner diameter of 3.5 cm. There was no pressure difference applied to the system with the exception of the gravitational pressure from the respective liquid columns.

In this experiment, 25 ml of nitric acid (2 N for emulsion A and 13 N for emulsion B) and 25 ml of TBP were poured into the upper part of the system and stirred vigorously with a mechanical stirrer. Immediately the organic phase began to run through the filter completely free of any visible drops of the inorganic phase. The aqueous phase was dyed with dark ink to make it distinguishable from the organic phase. Figure 4 presents a sequence of the extraction process. The system is shown on the left side filled with the emulsion at the start; on the right side the two separated phases are shown after the experiment has finished. As can be seen, at the end of the separation the aqueous phase (dark dyed liquid) stays above the filter and the organic phase (colourless liquid) is completely collected in the lower flask.

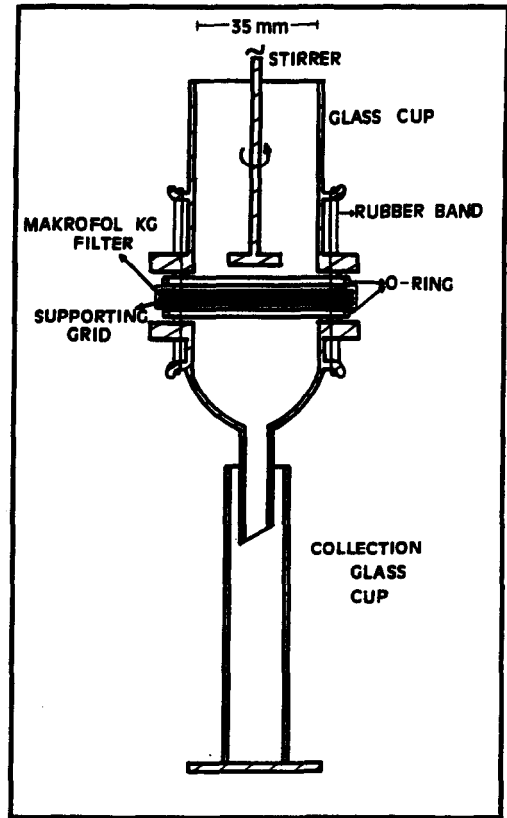


FIG. 3. Schematic representation of the extraction apparatus.

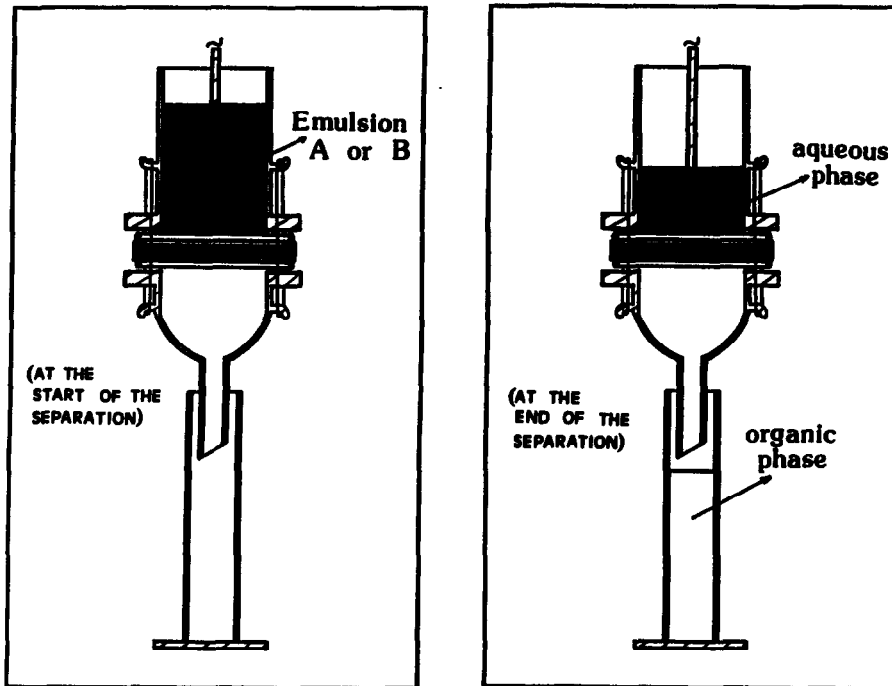


FIG. 4. Sequence of sketches showing the complete extraction process of the organic phase out of a typical emulsion by using a Makrofol KG nuclear track microfilter.

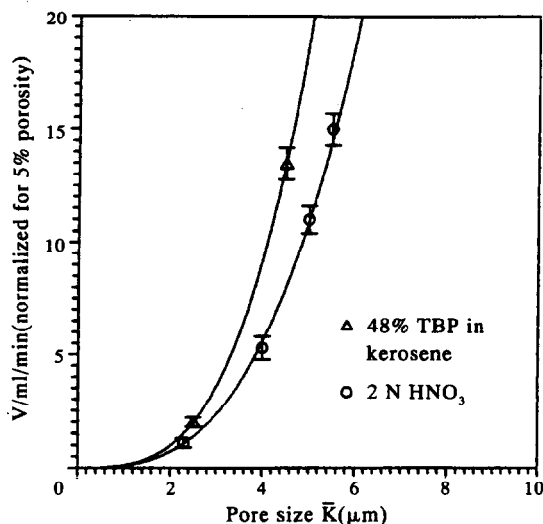


FIG. 5. Throughput  $\bar{V}$  for each pure phase of the emulsion consisting of 48% TBP in kerosene and 2 N  $\text{HNO}_3$  vs pore size of the microfilter.

### 3. RESULTS AND DISCUSSION

#### 3.1. Volumetric throughput of pure phases

The volumetric throughput of each pure phase of emulsion A through Makrofol KG filters was investigated. The time taken to pass 50 ml of each pure phase through filters with pore sizes ranging from  $(2.3 \pm 0.2)$  up to  $(5.5 \pm 0.3)$   $\mu\text{m}$  was measured. The result is shown in Fig. 5 where the throughput (in ml/min, normalized for 5% filter porosity) is plotted against the average pore size  $\bar{k}$  ( $\mu\text{m}$ ) of the Makrofol KG filters. As can be seen, for a given pore size  $\bar{k}$ , the time taken for the organic phase to pass through the filter is significantly shorter than the time taken for the aqueous phase (nitric acid). In each case, the filter had to be carefully wetted with its own organic phase

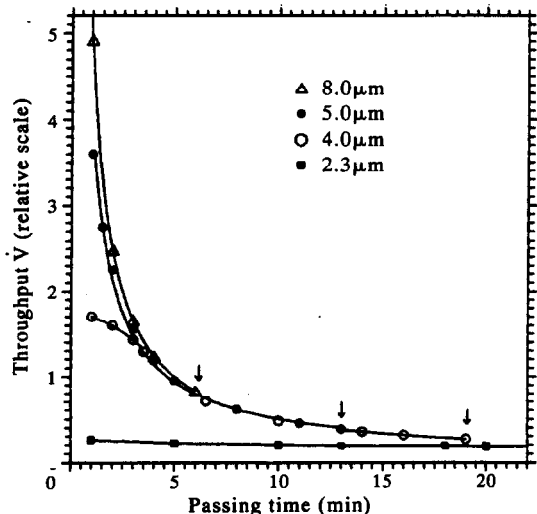


FIG. 6. Throughput  $\bar{V}$  of the organic phase extracted from the emulsion formed with 2 N  $\text{HNO}_3$  vs passing time.

Table 1. Characteristics of organic phase (48% TBP in kerosene) extraction from the emulsion formed with 2 N  $\text{HNO}_3$

Pore size ( $\mu\text{m}$ )	Filtering time (90% of volume) (min)	Filtering time (100% of volume) (min)
$(8.0 \pm 0.4)$	0.5	6.0
$(6.5 \pm 0.3)$	1.0	9.0
$(5.0 \pm 0.3)$	2.0	13.0
$(4.0 \pm 0.3)$	3.5	19.0
$(2.3 \pm 0.2)$	Very long	Very long

before any reproducible throughput measurement could be taken.

#### 3.2. Extraction of the organic phase out of the emulsion

The Makrofol KG filters used for the separation of the organic phase from emulsion A had different pore sizes and porosity varying between 3 and 6%. After the filter had been wetted with the organic phase, 50 ml of emulsion was poured into the cup and stirred vigorously with a mechanical stirrer. The organic phase started immediately to pass through the filter and was completely free of inorganic component up to the end of the separation process. None of the aqueous phase passed through the filter even after a long filtration. This means that the whole process is self controlling. At the end of the process the liquid in the bottom cup was completely transparent and its volume was half its original value, showing that the organic phase became completely separated from its emulsion with the inorganic phase.

Figure 6 shows a general feature of Makrofol KG filters, that is, their ability in separating the organic and the inorganic phase for emulsion A. The throughput of organic phase out of the emulsion is plotted (on a relative scale) against time which passes during the separation experiment for four Makrofol KG filters with different pore sizes. It can be seen that the initial throughput is higher for the larger pore size filter and the larger the initial throughput, the steeper is the decrease in throughput. As passing times increase, the amount of organic phase in contact with the filter reduces; therefore, throughput decreases with increasing passing time. The results of the separation experiment are presented in Table 1. As can be seen, filters with pore sizes smaller than 4.0  $\mu\text{m}$  have longer separation times and insufficient extraction results.

Table 2. Characteristics of organic phase (48% TBP in kerosene) extraction from the emulsion formed with 13 N  $\text{HNO}_3$

Pore size ( $\mu\text{m}$ )	Filtering time (90% of volume) (min)	Filtering time (100% of volume) (min)
$(10.5 \pm 0.6)$	1.5	11.0
$(8.8 \pm 0.5)$	3.5	19.0
$(7.7 \pm 0.4)$	7.0	29.0
$(5.5 \pm 0.3)$	Very long	Very long

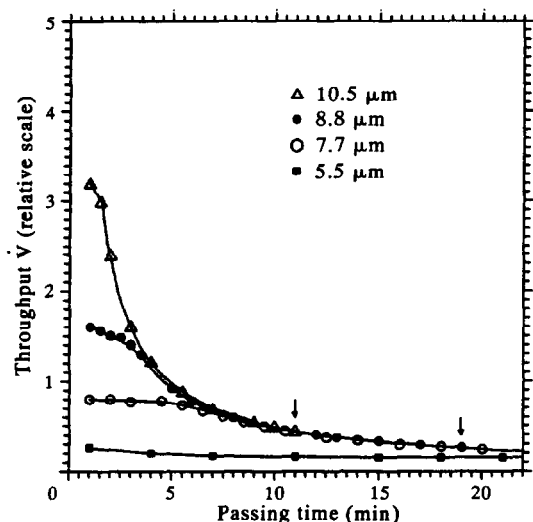


Fig. 7. Throughput  $\bar{V}$  of the organic phase extracted from the emulsion formed with 13 N  $\text{HNO}_3$  vs passing time.

In order to extract the organic phase from emulsion B, filters with porosity between 4 and 9% and different pore sizes were used. The results obtained are presented in Table 2 and Fig. 7 shows the general features of the separation of the organic phase out of the emulsion. In this case, filters with pore sizes smaller than  $8.8 \mu\text{m}$  gave poor extraction results.

For practical purposes, we can consider a Makrofol KG filter having a pore size of around  $8.0 \mu\text{m}$  and porosity of about 5% as a good option for the extraction of the organic phase out of emulsion A (Table 1), and a pore size of around  $10.5 \mu\text{m}$  and porosity around 5% in the case of emulsion B (Table 2). However, if a shorter filtration time is needed, filters with pore size greater than  $8.0$

and  $10.5 \mu\text{m}$  can be used for emulsions A and B, respectively.

#### 4. SUMMARY

In the present work, it was shown that Makrofol KG filters are well suited for the separation of organic liquids out of the emulsion they form with aqueous liquids. Under well defined conditions a complete extraction of the organic phase was achieved for two types of emulsions which have been used in radioanalytical work.

It is our aim that separation processes for other types of emulsions may also be successfully carried out using Makrofol KG microfilters.

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