



**HAL**  
open science

## Analysis of radionuclides in microsystem: application to the selective recovery of $^{55}\text{Fe}$ by solvent extraction

Somasoudrame Rassou, Clarisse Mariet, Thomas Vercoouter

### ► To cite this version:

Somasoudrame Rassou, Clarisse Mariet, Thomas Vercoouter. Analysis of radionuclides in microsystem: application to the selective recovery of  $^{55}\text{Fe}$  by solvent extraction. EPJ N - Nuclear Sciences & Technologies, 2020, 6, pp.10. 10.1051/epjn/2020002 . hal-02903896

**HAL Id: hal-02903896**

**<https://hal.science/hal-02903896>**

Submitted on 21 Jul 2020

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution 4.0 International License

# Analysis of radionuclides in microsystem: application to the selective recovery of $^{55}\text{Fe}$ by solvent extraction

Somasoudrame Rassou, Clarisse Mariet, and Thomas Vercouter\*

Den-Service d'Etudes Analytiques et de Réactivité des Surfaces (SEARS), CEA, Université Paris-Saclay, 91191 Gif-sur-Yvette, France

Received: 28 October 2019 / Received in final form: 20 December 2019 / Accepted: 16 January 2020

**Abstract.** The minimization of the sample quantities required by analytical laboratories, as well as the increase of the fastness of the analytical operations are emerging axes for improved radiochemical analyses related to D&D issues. Two microsystem-based protocols were developed for the selective recovery of  $^{55}\text{Fe}$  from radioactive samples by solvent extraction. Both protocols were tested on iron solutions in two different microchips. The yields of Fe extraction were compared with macroscale batch experiments. Better performances with more than 80% of iron extracted were obtained with the second protocol, which is based on a reactive transfer of the iron cation, and more suited to the use of microchannels and very low contact times. This study already demonstrate the high potential of microfluidic technology to improve analytical operations on D&D samples. This method will further be validated with radioactive samples.

## 1 Introduction

The characterization of the sites under decommissioning or dismantling, and of the subsequent wastes is addressed by the use of validated analytical methods for radiochemical measurements with different kinds of techniques. A large variety of analytical issues and challenges exist considering the type of matrices, the nature and quantities of the radionuclides, the activity levels, etc. Destructive analysis represents a large part of the analytical methods applied to D&D samples, and large efforts are made to develop and validate the methods, particularly on heterogeneous or ill-defined materials, and to meet the requirements regarding the performance of the detection and the quantification performances. Considering the radioactivity of the samples, additional constraints have to be considered for the sampling, the shipment of the samples to the analytical laboratory, the handling of analytical operations, and finally the management of the associated wastes. Moreover, the time required for obtaining the final analytical results is also an important aspect because of the large number of samples to be analysed at the different stages of the D&D process.

The minimization of the sample quantities required by the analytical laboratory, as well as the increase of the fastness of the analytical operations are emerging axes of development of radiochemical laboratories. The conception

and development of miniaturized analytical device, the so-called lab-on-a-chip, can answer these issues by integrating and optimizing one or several analytical operations in the same object that uses only the right quantity of samples for the measurements thanks to microfluidics coupled with appropriate detection equipment. In radiochemistry, the use of analytical microsystems is still at the level of R&D projects, but is developing very fast because of the high potential of this technology to considerably reduce the hazards and constraints related to radionuclides analysis. It benefits from the progresses made in other fields like in the health sector, in bioanalysis, or in microelectronics, and needs to be adapted to the requirements of radiochemical analysis.

This study has therefore been dedicated to the evaluation of the performances of a microsystem-based method for the analysis of a chosen radionuclide in relevant samples for D&D applications [1], with the objective to further validate the method and eventually integrate it in radiochemical laboratory protocols.

The miniaturization of the analysis device is relevant when the analytical steps involve the use of hazardous reagents or require significant quantities of radioactive samples. It is particularly the case for solvent extraction steps. Among the protocols applied at CEA, we selected the analysis of  $^{55}\text{Fe}$  which is measured by liquid scintillation counting after several sample preparation steps in order to remove interfering isotopes. After a purification step by solid phase extraction, iron (III) is separated from other metals by solvent extraction. Chloroform is used as the

\* e-mail: [thomas.vercouter@cea.fr](mailto:thomas.vercouter@cea.fr)

diluent, and cupferron as the iron chelating agent. Yet, as chloroform is classified as a carcinogen, mutagen or substance toxic to reproduction (CMR), the protocol must be modified, and the use of a microsystem-based protocol is of great interest in this case.

The aim of our work was to develop an extraction protocol for the selective recovery of  $^{59}\text{Fe}$ , and fulfil the criteria of green chemistry, by:

- using another diluent than chloroform;
- reducing the manipulated quantities using microsystems.

This was achieved by testing two protocols based on solvent extraction in macro and micro-scales. The results will be compared regarding the yield of extraction and the time needed to recover the analyte.

## 2 Solvent extraction

### 2.1 State-of-the art in solvent extraction miniaturization

The miniaturization of liquid–liquid extraction began in the mid-1990's. In 1996, Liu and Dasgupta described a drop-in-drop system consisting in a micro-drop of chloroform suspended in an aqueous drop of sample [2]. During the last decade, solvent microextraction (or liquid-phase microextraction) techniques (SME/LPME) have undergone notable development [3–6]. We can distinguish several methods that are included in the SME group, for example, single-drop microextraction (SDME), and dispersive liquid–liquid microextraction (DLLME). These methods differ in design, but they all have one common feature: namely, they only use micro volumes of organic solvent and thus comply with the requirements of green analytical chemistry [7,8]. They are inexpensive, rapid and simple since no special equipment is required and they can be combined with many techniques for determination of the analytes. However, SDME presents instability of the drop that implies a lesser accuracy; furthermore the volume and surface of the drops are limited, which exclude reactions with slow kinetics [8]. In the case of DLLME, three solvents are needed and there are some restrictions on the selection of extraction solvent. Otherwise, additional steps including centrifugation, freezing, and auxiliary solvent demulsifiers must be employed, which undo the benefits of the scale reduction.

The rapid development of microsystems for chemical analysis has been greatly promoted by the progress made within micro-fabrication technology [9,10]. These microsystems are known as micro-total-analysis systems ( $\mu$ -TAS) or lab-on-a-chip [11]. Yager et al. micro-fabricated a H-filter micro-device that separates particles based on their diffusion coefficients [12]. Kitamori et al. have developed a Y–Y shaped micro-device for ion-pair solvent extraction of Fe(II) with 4,7-diphenyl-1,10-phenanthrolinedisulfonic acid by tri-*n*-octylmethylammonium chloride diluted in chloroform [10]. Then, they studied solvent extraction with stratified flows in a microchannel as a separation technique in the pre-treatment step of the trace metal assay [13]. These studies showed that a micro flow channel is particularly suitable for the interfacial

reaction of solvent extraction, since it increases the ratio of the interface area (between aqueous and organic phases) to the total volume of the aqueous and organic phases.

Compared with the solvent microextraction (SME) methods, stratified flows solvent extractions in microsystems present the advantage to allow a precise control of the contact times of the two phases, especially short contact times, to lead to high speed and high performance without any mechanical stirring, mixing or shaking. Moreover, recent technological breakthroughs allow to work with automated microsystems which can be used in parallel processing to increase the throughput or in multiplexed processing of separation/purification steps coupled to a detection system [13,14]. There are few examples of microchemical systems that utilize two or more liquid streams with parallel laminar flow in a microchannel for radiochemical applications [15–18]. Examples include the extraction of uranium (VI) in nitric acid media by tributylphosphate in dodecane or in ionic liquids [19,20]. The extraction of U from hydrochloric acid media by a malonamide [21], the extraction of Y, Eu, La or Pr, Nd, Sm from nitric acid by 2-ethylhexyl phosphonic acid mono-2-ethylhexyl ester (PC-88A) diluted in toluene [22,23] or in kerosene [24]. The extraction of Am (III) by *n*-octyl (phenyl)-*N,N*-diisobutylcarbamoylmethylphosphine oxide (CMPO) was also studied [25].

In light of this state of the art, glass microsystems with Y–Y junction have been chosen to perform the liquid-liquid selective extraction of iron. A further objective will be to couple it with a detection step by liquid scintillation. Recently, a Y–Y glass microsystem was successfully used for the extraction of Pu by 30% TBP in *n*-dodecane, and the outlet Pu-enriched organic phase was mixed on-line with a scintillation cocktail and driven to a flow-through cell of an  $\alpha$ -liquid scintillator counter [26].

### 2.2 Principle of the method

The solvent extraction experiments were carried out using an acidic aqueous phase contacted with an organic phase using ethyl acetate as the diluent.

In batch experiments, the distribution coefficient ( $\mathcal{D}_M$ ) is defined by:

$$\mathcal{D}_M = \frac{[M]_{\text{org}}}{[M]_{\text{aq}}} \quad (1)$$

where  $[M]_{\text{org}}$  and  $[M]_{\text{aq}}$  are the metal concentration in the organic phase and in the aqueous phase, respectively. For equal volumes of both phases ( $V_{\text{aq}} = V_{\text{org}}$ ), we have:

$$\mathcal{D}_M = \frac{[M]_i - [M]_{\text{aq}}}{[M]_{\text{aq}}} \quad (2)$$

where  $[M]_i$  is the initial metal concentration in the aqueous phase. Then the extraction yield (%  $E_M$ ) of the metal (M) is calculated from as follows:

$$\%E_M = 100 \times \left( \frac{[M]_i - [M]_{\text{aq}}}{[M]_i} \right) = 100 \frac{\mathcal{D}_M \frac{V_{\text{org}}}{V_{\text{aq}}}}{1 + \mathcal{D}_M \frac{V_{\text{org}}}{V_{\text{aq}}}} \quad (3)$$

In microsystem, the liquid–liquid extraction reaction remains the same as in batch experiments, but the extraction takes place in stationary dynamic mode with different flow rates of the two phases [27,28]. The length of the microchannel where the phases are contacted, noted  $L$ , imposes the contact time for given flow rates.

The distribution coefficient of an analyte keeps the same definition as at the macroscopic scale but changes at each moment in the microchannel. This coefficient is defined by:

$$\mathcal{D}_{M,\text{microsystem}} = \frac{[M]_{\text{org},x}}{[M]_{\text{aq},x}} \quad (4)$$

where  $x$  determines the position in the microchannel ( $0 \leq x \leq L$ ). The value of  $\mathcal{D}_{M,\text{microsystem}}$  increases along the microchannel up to a constant value if the equilibrium is reached at the output of the microsystem. Experimentally, only the value of  $\mathcal{D}_{M,\text{microsystem}}$  at the output of the microsystem can be determined according to:

$$\mathcal{D}_{M,\text{microsystem}} = \frac{[M]_{\text{org},L}}{[M]_{\text{aq},L}} = \left( \frac{[M]_i}{[M]_{\text{aq},L}} - 1 \right) \frac{Q_{\text{aq}}}{Q_{\text{org}}} \quad (5)$$

where  $[M]_{\text{org},L}$  and  $[M]_{\text{aq},L}$  are the concentrations of the analyte  $M$  at the output of the microsystem (of length  $L$ ) in organic phase and aqueous phase, respectively;  $[M]_i$  is the initial concentration of the analyte  $M$ , and  $Q_{\text{aq}}$  and  $Q_{\text{org}}$  are the flow rates of the aqueous and organic phases, respectively.

The extraction yield is then determined by:

$$\%E_M = 100 \frac{\mathcal{D}_{M,\text{microsystem}} \frac{V_{\text{org}}}{V_{\text{aq}}}}{1 + \mathcal{D}_{M,\text{microsystem}} \frac{V_{\text{org}}}{V_{\text{aq}}}} \quad (6)$$

## 3 Experimental section

### 3.1 Materials and methods

Cupferron was supplied by Sigma Aldrich and used without purification. Cupferron solution was prepared by dissolving 2% weight amounts in deionized water (system Direct-Q UV3, Millipore). Iron nitrate solutions were prepared from a 1000 mg kg<sup>-1</sup> SPEX solution (Jobin Yvon, France). Hydrochloric acid (37% wt), nitric acid (65% wt), acetone and ethyl acetate were purchased from Sigma Aldrich (France). Aqueous and organic solutions were both pre-saturated by contact under shaking for 120 min in order to transfer water and acid from the aqueous phase to the organic phase and the small soluble quantities of solvent to the aqueous phase.

The solution density was measured using a DMA 4500 density-meter (Anton Paar, Austria) at a controlled temperature of 293.150 ± 0.001 K. The accuracy of the density measurement was approximately ±3 × 10<sup>-6</sup> kg dm<sup>-3</sup>. The viscosity was measured at atmospheric pressure with a

**Table 1.** Characteristics of the Pyrex<sup>®</sup> glass microsystems.

IMT reference	$L$ (cm)	Number of stages
ICC-DY15	12	Single
ICC-DY20	20	Single
DR14920	10 and 10	Double

rotational automated viscosimeter (Lovis 2000 M/ME, Anton Paar, Austria). The accuracy of the viscosity measurements was better than 0.5%.

Fe concentrations were determined by Inductively Coupled Plasma Mass Spectrometry (ICP-MS, 7700x, Agilent Technologies, France) equipped with a concentric nebulizer. Analytical calibration standards were prepared daily over the range of 0–200 ng g<sup>-1</sup> by suitable serial dilutions of the stock solution in 2% (v/v) HNO<sub>3</sub>. Germanium-72 was used as an internal standard at a concentration of 20 ng g<sup>-1</sup> from a 1000 mg kg<sup>-1</sup> standard solution. The reproducibility was determined with 3 repeats of these measurements and was within 10%.

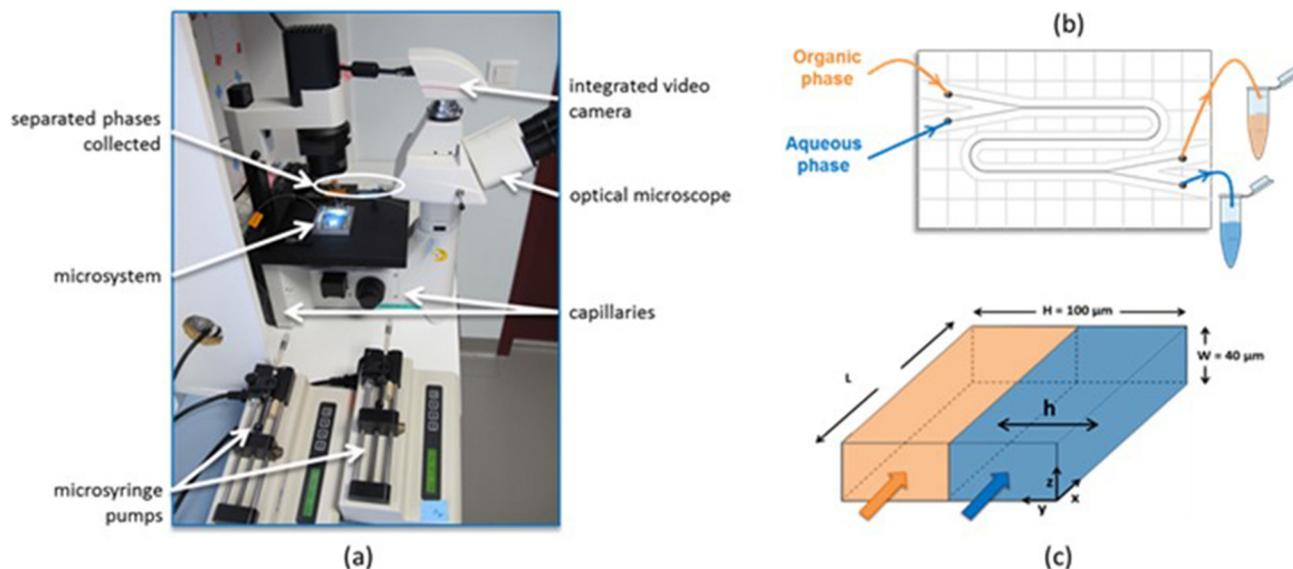
### 3.2 Solvent extraction controls in batch

A volume of 800 μl of an aqueous solution was contacted with an equal volume of an organic solution and shaken in a thermomixer apparatus under the following conditions:  $T = 293 \pm 1$  K; 1400 rpm; shaking time = 2 h. After centrifugation and phase separation, the concentrations of the Fe analyte remaining in the aqueous and organic phases were determined by ICP-MS. The distribution ratio and the extraction yield were calculated using equations (1) and (3), respectively.

### 3.3 Microfluidic experiments

The Y–Y shaped Pyrex<sup>®</sup> glass microfluidic devices were purchased from IMT (Institute of Microchemical Technologies, Kanagawa, Japan) (Tab. 1) and used with a stainless-steel holder (ICH-04, IMT).

The microsystems were operated as described below (Fig. 1). The aqueous and organic phases were injected using two glass syringes (Hamilton, 1 mL) and the flow rates were controlled by a syringe pump connected to the microfluidic device with PEEK capillary tubing (external diameter = 510 μm and internal diameter = 125 μm) and Luer-lock Teflon<sup>®</sup> connectors (ISC-01, IMT). At the outlets of the microsystem, the same PEEK capillary tubing are used to collect both parts in Eppendorf tubes. A digital inverted microscope (DEMIL LED Leica, France) equipped with an objective lens with a 40 times magnification and a camera DFC 295 (Hamamatsu) and a binocular microscope (VWR, France) BI 100 were employed to observe the flow behaviour of the solutions in the microchannel.



**Fig. 1.** (a) Experimental setup for extraction studies in a glass microsystem; (b) scheme of the single-stage microsystem; (c) focus on a part of the microchannel: extraction length  $L = 12$  or  $20$  cm, width  $H = 100 \mu\text{m}$ ,  $h$  the position of the interface and depth  $W = 40 \mu\text{m}$ .

All the solutions were filtered before being injected into the microsystem. After equilibration of the flows using deionized water and the diluent at an equal flow rate of  $0.5 \text{ ml h}^{-1}$  for 5 min., the water was replaced by the aqueous phase containing the analyte  $M$  and the diluent is replaced by the organic phase in the two microsyringes. Then, the two phases were injected into the microsystem at an equal flow rate of  $0.5 \text{ ml h}^{-1}$  for 5 additional minutes.

The second step consists in the liquid-liquid extraction itself. For a given flow rate of the aqueous phase, the flow rate of the organic phase was imposed so as to respect the following relationship:

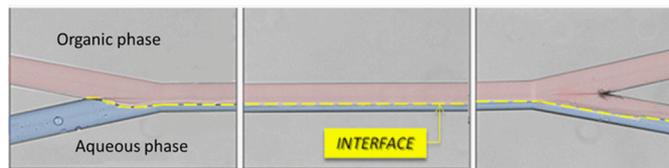
$$\frac{Q_{\text{org}}}{Q_{\text{aq}}} = \frac{\mu_{\text{aq}} h_{\text{org}}}{\mu_{\text{org}} h_{\text{aq}}}$$

where  $Q_{\text{org}}$  and  $Q_{\text{aq}}$  are the organic and aqueous flow rates, respectively, and  $\mu_{\text{org}}$  and  $\mu_{\text{aq}}$  are the organic and aqueous viscosities, respectively, and  $h_{\text{org}}$  and  $h_{\text{aq}}$  are the widths of the organic and aqueous compartments in the microchannel. For symmetric microchannel as used in the present work, this relationship becomes:

$$\frac{Q_{\text{org}}}{Q_{\text{aq}}} \sim \frac{\mu_{\text{aq}}}{\mu_{\text{org}}}$$

For each protocol and microsystem, we experimentally determined the rate flow domain where both parallel flows and good phase separation at the outlets of the microchip were obtained. The ratio of the resulting flow rates compared well with the ratio of the measured dynamic viscosities.

For other flow rate values, either the interface was not centered as illustrated in [Figure 2](#), or other types of flows (slug, droplet, wavy flows...) were observed.



**Fig. 2.** Example of the position of the interface in the ICC-DY10 microsystem (based on microscope photographs).

For each couple of flow rates a photograph of the microchannel was taken to verify the position of the interface. The contact times were calculated according to the following equations:

$$t_{\text{aq}} = \frac{hWL}{Q_{\text{org}}}$$

$$t_{\text{org}} = \frac{(H-h)WL}{Q_{\text{org}}}$$

When the steady state was achieved (i.e. laminar and parallel flows) and the interface was correctly centered (i.e.  $h = H/2$ ), usually within a few minutes at most, about  $200 \mu\text{l}$  of the two separated phases were collected at the outlets. All experiments were triplicated at  $293 \text{ K}$ .

## 4 Results

### 4.1 Partitioning protocol (A)

Iron extraction was investigated with the protocol A in which  $\text{Fe}(\text{Cupferrate})_3$  partitioned into ethyl acetate. The composition of the aqueous phase was

$[\text{Fe(III)}] = 5.7 \times 10^{-4} \text{ mol l}^{-1}$ ,  $[\text{Cupferron}] = 4.2 \times 10^{-2} \text{ mol l}^{-1}$  in a  $3.2 \text{ mol l}^{-1}$  HCl solution. The best extraction yields were about  $(37 \pm 3)\%$  in the 12-cm long ICC-DY15 microsystem for  $t_{\text{aq}}$  between 1.0 and 1.72 s. Similar experiments were carried out with the 20-cm long ICC-DY200 microsystem, and the best extraction yield was  $(45 \pm 6)\%$  for  $t_{\text{aq}} = 2.40$  s and  $t_{\text{org}} = 1.04$  s.

These values are both much lower than the reference value of  $(93.0 \pm 2.3)\%$  obtained from batch experiments with  $V_{\text{org}}/V_{\text{aq}} \approx 2$ , chosen to be close to the ratio of the flow rates used in microsystems. Increasing time of contact in the microsystem by lengthening the microchannel did not significantly improve the extraction. The protocol A was considered inappropriate using a single-stage microsystem.

Protocol A was applied to the DR14920 double-stage microsystem. The optimal extraction yield was  $(60 \pm 5)\%$  with  $t_{\text{aq,total}} = 2.27$  s, and  $t_{\text{org,total}} = 1.82$  s for  $Q_{\text{aq}} = 0.65 \text{ ml h}^{-1}$ . As expected the extraction yield was increased, but remained much lower than that obtained from batch experiments.

## 4.2 Reactive transfer protocol (B)

In protocol B iron (III) is extracted by complexation with cupferron in ethyl acetate. Experiments with a 12-cm long ICC-DY15 microsystem have shown that the time of pre-equilibration of the solvent phase prior to extraction has a great influence on the extraction yield. Cupferron must be diluted into ethyl acetate and equilibrated for at least 2 h. The maximum extraction yield was  $(83.1 \pm 5.2)\%$  for  $t_{\text{aq}} = 1.37$  s and  $t_{\text{org}} = 0.72$  s. This value is much closer to the reference value  $(96.4 \pm 1.4)\%$  for  $V_{\text{org}}/V_{\text{aq}} \approx 2$  at equilibrium in batch.

This protocol used the DR14920 double-stage microsystem, and the following flow rates were selected:  $Q_{\text{org}(1)} = 1.60 \text{ ml h}^{-1}$ ,  $Q_{\text{aq}} = 0.96 \text{ ml h}^{-1}$ ,  $Q_{\text{org}(2)} = 0.54 \text{ ml h}^{-1}$ . The best extraction yield obtained was  $(81.7 \pm 2.0)\%$  with  $t_{\text{aq,total}} = 1.75$  s and  $t_{\text{org,total}} = 1.25$  s. No gain was obtained compared with the single-stage ICC-DY15 microsystem.

## 4.3 Selectivity of $^{55}\text{Fe}$ extraction

The objective of the liquid–liquid extraction of iron from HCl solution is to remove potential interfering isotopes that may bias the measurement. In radioactive wastes, the  $^{55}\text{Fe}$  radionuclide is often present with other radioactive isotopes such as cobalt ( $^{60}\text{Co}$ ) and caesium ( $^{137}\text{Cs}$ ). These isotopes are potential emitters that can interfere with the  $^{55}\text{Fe}$  emission in liquid scintillation measurement.

Test experiments were conducted with an aqueous solution containing Fe, Co and Cs. The iron concentration was chosen one hundred times lower than the ones of Co and Cs because it is the most unfavourable case as encountered in samples from D&D sites. Extraction experiments were carried out with the ICC-DY15 microsystem for  $Q_{\text{aq}} = 0.65 \text{ ml h}^{-1}$  and  $Q_{\text{org}} = 1.67 \text{ ml h}^{-1}$  with respect to the measured viscosity ratio of 2.6. The

**Table 2.** Selectivity of extraction of Fe, Co and Cs in batch experiment and in a ICC-DY15 microsystem.

	$E_{\text{Fe}}$ (%)	$E_{\text{Co}}$ (%)	$E_{\text{Cs}}$ (%)
Batch	$94.0 \pm 0.8$	$23.3 \pm 7.7$	$34.3 \pm 2.0$
ICC-DY15	$75.3 \pm 1.4$	$16.0 \pm 9.9$	$17.6 \pm 6.5$

extraction yields were slightly lower than in batch experiments, but the selectivity regarding the Co and Cs cations was conserved (Tab. 2).

## 5 Conclusion

The recovery of iron(III) by solvent extraction is effective using glass microsystem. The transposition of the chemical protocol (i.e. protocol A) used in batch to the microsystem is not appropriate, certainly as a result of kinetic limitations. The yield of extraction is much higher when using a new protocol in which the cupferron extractant is first added to the ethyl acetate solvent. In this case the extraction yield is very close to the one obtained in batch experiments. Quantitative extraction of iron is achieved with a single-stage microsystem, and was not improved with a double-stage microsystem.

The extraction of Fe by cupferron was still selective regarding Co and Cs that are elements with potential interfering isotopes in liquid scintillation measurements of  $^{55}\text{Fe}$ .

The performances of the microsystem-based method will further be validated in the course of the INSIDER EU project with analyses of radioactive samples containing  $^{55}\text{Fe}$ . The effective gain will be evaluated considering the reduction of volumes of chemicals and samples, and the reduction of the time of the extraction step. It is already demonstrated that once the flow rates conditions are set, based on viscosity measurements, extraction and phase separation are achieved in a few seconds only. Automation and parallelization of microchannels may also be of great interest to improve the statistics of the measurements or to handle higher number of samples.

The research leading to these results has received funding from the Euratom research and training programme 2014-2018 under grant agreement No 755554. The authors thank Dr. R. Brennetot and Dr. D. Roudil (CEA, France), and Dr. B. Russel (National Physical Laboratory, UK), for their interest and fruitful comments.

## Author contribution statement

Dr. Somasoudrame Rassou carried out the extraction experiments with the microsystems and analyses as a post-doctoral fellowship, under the supervision by Dr. Clarisse Mariet who also participated in the experimental work. Dr. Clarisse Mariet and Dr. Thomas Vercouter

contributed by providing support and expert viewpoints. This article was written by the three co-authors, and is associated to the presentation given by Dr. Thomas Vercouter at the mid-term workshop of the INSIDER EU project in May, 2019.

## References

1. S. Rassou, T. Vercouter, C. Mariet, Sustainable Solvent Extraction Process for Fe Analysis in Radioactive Samples Based on Microfluidic Tools, *Solvent Extr. Ion Exch.* **38**, 236 (2020)
2. H. Liu, P.K. Dasgupta, Analytical chemistry in a drop. Solvent extraction in a microdrop, *Anal. Chem.* **68**, 1817 (1996)
3. R. Burakham et al., Exploiting sequential injection analysis with lab-at-valve (LAV) approach for on-line liquid-liquid micro-extraction spectrophotometry, *Talanta* **68**, 416 (2005)
4. M.A. Jeannot, F.F. Cantwell, Mass transfer characteristics of solvent extraction into a single drop at the tip of a syringe needle, *Anal. Chem.* **69**, 235 (1997)
5. M. Ma, F.F. Cantwell, Solvent microextraction with simultaneous back-extraction for sample cleanup and preconcentration: preconcentration into a single microdrop, *Anal. Chem.* **71**, 388 (1999)
6. S. Pedersen-Bjergaard, K.E. Rasmussen, Liquid-liquid-liquid microextraction for sample preparation of biological fluids prior to capillary electrophoresis, *Anal. Chem.* **71**, 2650 (1999)
7. L. Kocúrová, I.S. Balogh, V. Andruch, Solvent micro-extraction: A review of recent efforts at automation, *Microchem. J.* **110**, 599 (2013)
8. A. Spietelun et al., Green aspects, developments and perspectives of liquid phase microextraction techniques, *Talanta* **119**, 34 (2014)
9. A. Manz, J.C. Eijkel, Miniaturization and chip technology. What can we expect? *Pure Appl. Chem.* **73**, 1555 (2001)
10. M. Tokeshi, T. Minagawa, T. Kitamori, Integration of a microextraction system on a glass chip: ion-pair solvent extraction of Fe (II) with 4, 7-diphenyl-1, 10-phenanthroline-disulfonic acid and tri-n-octylmethylammonium chloride, *Anal. Chem.* **72**, 1711 (2000)
11. H. Becker, W. Dietz, P. Dannberg, *Microfluidic manifolds by polymer hot embossing for  $\mu$ -TAS applications in Micro Total Analysis Systems* (Springer, Berlin, 1998)
12. J.P. Brody, P. Yager, Diffusion-based extraction in a microfabricated device, *Sens. Actuators A Phys.* **58**, 13 (1997)
13. M. Tokeshi, T. Kitamori, Continuous flow chemical processing on a microchip using microunit operations and a multiphase flow network, *Prog. Nucl. Energy* **47**, 434 (2005)
14. T. Thorsen, S.J. Maerkl, S.R. Quake, Microfluidic large-scale integration, *Science* **298**, 580 (2002)
15. D. Ciceri, J.M. Perera, G.W. Stevens, The use of microfluidic devices in solvent extraction, *J. Chem. Technol. Biotechnol.* **89**, 771 (2014)
16. G. Hellé, C. Mariet, G. Cote, Microfluidic tools for the liquid-liquid extraction of radionuclides in analytical procedures, *Procedia Chem.* **7**, 679 (2012)
17. G. Hellé, C. Mariet, G. Cote, Liquid-liquid microflow patterns and mass transfer of radionuclides in the systems Eu (III)/HNO<sub>3</sub>/DMDBDTMA and U (VI)/HCl/Aliquat<sup>®</sup> 336, *Microfluid. Nanofluidics* **17**, 1113 (2014)
18. G. Hellé, C. Mariet, G. Cote, Liquid-liquid two-phase microflow patterns and mass transfer of radionuclides, in *2014 9th IEEE International Conference on Nano/Micro Engineered and Molecular Systems (NEMS), 2014*
19. H. Hotokezaka et al., System for high-level radioactive waste using microchannel chip extraction behavior of metal ions from aqueous phase to organic phase in microchannel, *Prog. Nucl. Energy* **47**, 439 (2005)
20. D. Tsaoulidis et al., Extraction of dioxouranium (VI) in small channels using ionic liquids, *Chem. Eng. Res. Des.* **91**, 681 (2013)
21. G. Hellé, C. Mariet, G. Cote, Liquid-liquid extraction of uranium (VI) with Aliquat<sup>®</sup> 336 from HCl media in microfluidic devices: Combination of micro-unit operations and online ICP-MS determination, *Talanta* **139**, 123 (2015)
22. F. Kubota, J.I. Uchida, M. Goto, Extraction and separation of rare earth metals by a microreactor, *Solvent Extr. Res. Dev.* **10**, 93 (2003)
23. T. Maruyama et al., Liquid membrane operations in a microfluidic device for selective separation of metal ions, *Anal. Chem.* **76**, 4495 (2004)
24. S. Nishihama, Y. Tajiri, K. Yoshizuka, Separation of lanthanides using micro solvent extraction system, *Ars Separatoria Acta* **4**, 18 (2006)
25. Y. Ban et al., Extraction of Am (III) at the interface of organic-aqueous two-layer flow in a microchannel, *J. Nucl. Sci. Technol.* **48**, 1313 (2011)
26. M. Yamamoto et al., Development of an online measurement system using an alpha liquid scintillation counter and a glass-based microfluidic solvent extraction device for plutonium analysis, *Appl. Radiat. Isot.* **152**, 37 (2019)
27. B. Malengier, S. Pushpavanam, S. D'haeyer, Optimizing performance of liquid-liquid extraction in stratified flow in micro-channels, *J. Micromech. Microeng.* **21**, 115030 (2011)
28. L.R. Mason et al., Modelling of interfacial mass transfer in microfluidic solvent extraction: part I. Heterogenous transport, *Microfluid. Nanofluidics* **14**, 197 (2013)

**Cite this article as:** Somasoudrame Rassou, Clarisse Mariet, Thomas Vercouter, Analysis of radionuclides in microsystem: application to the selective recovery of <sup>55</sup>Fe by solvent extraction, *EPJ Nuclear Sci. Technol.* **6**, 10 (2020)