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ScienceDirect

Procedia Chemistry 21 (2016) 487 – 494

Procedia Chemistry

5th International ATALANTE Conference on Nuclear Chemistry for Sustainable Fuel Cycles

Design, development and testing of miniature Liquid-Liquid Extraction contactors for R&D studies in nuclear environment

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Abstract

Testing liquid-liquid extraction flowsheets on radioactive solutions at laboratory scale requires specific contactors, that must be as small as possible (to minimize the quantities of reagents) and suitable for use in glove boxes or hot cells. As such contactors are not commercially available, CEA designs dedicated apparatus for its experiments. Two examples of such contactors are highlighted: a stage-wise one (a miniature mixer-settler) and a differential one (the Taylor-Couette column). Both combine similarities with larger contactors that make possible flowsheet extrapolation to industrial scale, and specific features, mainly because of the major role of materials wettability when volumes are reduced.

Peer-review under responsibility of the organizing committee of ATALANTE 2016

Keywords: liquid-liquid extraction, laboratory-scale contactors, miniature mixer-settler, Taylor-Couette extraction column

1. Introduction

Spent nuclear fuel contains re-usable products (uranium and plutonium) and numerous other elements including radionuclides with short to very long half-lives. Nuclear fuel reprocessing aims at separating these components, in order to reuse or store them adequately. In the classical industrial PUREX process, implemented for instance in the Areva La Hague plant, these separations are carried out by liquid-liquid extraction (LLE). Several other processes under development at CEA Marcoule, intended to complement the PUREX process or to propose an alternative to it, are also based on liquid-liquid extraction.

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Peer-review under responsibility of the organizing committee of ATALANTE 2016
doi:10.1016/j.proche.2016.10.068

At industrial scale, LLE of nuclear elements is operated with flowrates typically ranging from 100 to 1000 L/h (and even much more in other fields such as metals refining). LLE operations are implemented in different kinds of apparatus sorted into stage-wise contactors (e.g. mixer-settlers, centrifugal contactors) or differential ones (e.g. pulsed or agitated columns). The development of such solvent extraction processes starts with the selection of the adequate extracting molecule and the measurement of relevant data regarding thermodynamics (distribution ratios) and kinetics (mass transfer rate). Then, a key step consists in testing the whole extraction flowsheet at laboratory scale on radioactive solutions (first on laboratory-made representative solutions, then on solutions produced from actual used fuel). Because handling these radioactive solutions implies several issues (effluent storage and reprocessing, operators protection against radiations), these tests should be conducted at a very small scale, involving as few reagents as possible. Therefore, the laboratory liquid-liquid contactors used for these pilot trials must fulfil the following requirements:

- to be as small as possible,
- to be similar to the industrial contactors, in order to make easier a later scaling-up of the process,
- to be operable in nuclear environment (glove boxes or hot cells).

According to our knowledge, at present the smallest commercially available laboratory contactors are:

- regarding differential contactors, agitated columns with 30 mm inner diameter (SULZER Kühni-type column),
- regarding stage-wise contactors, mixer-settlers whose mixer's volume is 25 mL (MEAB MSU 0,1).

Both are appropriate for flowrates of several liters per hour, and thus are too large for experiments in high activity conditions. Furthermore, they were not especially designed in order to be used in hot cells. Thus, CEA has been developing for several decades more appropriate liquid-liquid contactors.

For many reasons, it is not possible to design a small contactor by simple scale reduction of a larger model. Obviously some mechanical parts (motors, walls) cannot be reduced proportionally to the mixer and settler inner volumes. More important, dominating phenomena in hydrodynamics are changing while reducing the size: at lower size, capillary effects grow in importance and, for instance, getting smooth low flows through small openings becomes challenging. The development and optimization of miniature contactors usually begins with the design of a first prototype and ends up with the testing in pilot plant. This challenging task will be introduced hereafter by two examples: a stage-wise contactor and a differential one.

2. Example of a stage-wise contactor: the « Next Generation » mixer-settler

Mixer-settlers (MS) are well-known and widely used liquid-liquid contactors. In MS, each stage consists of an agitated compartment (the mixer, where mass transfer occurs) and a non-agitated one (the settler, where both phases separate). To reach the required overall efficiency of each operation of a liquid-liquid separation process (extraction, stripping…) several stages are interconnected together to form batteries where both phases circulate in opposite directions (countercurrent flow). Classical features among the numerous models of MS are:

- the head to tail positioning of adjacent stages (the mixer of a stage is on the same side as the settlers of the adjacent stages), thus eliminating connection pipes between stages,
- the agitating mobiles (called "pump-mixer") that ensure both pumping of the liquids from adjacent stages into the mixer and mixing by shearing the fluids inside of the mixer.

The following paragraphs deal with the transposition of these features to tiny MS and highlight the performances achieved so far regarding hydrodynamics and extraction.

2.1. Design

A very compact and quite simple mixer-settler model had been designed by CEA three decades ago. The mixer and settler volumes (respectively 6 mL and 20 mL) are adequate for total flowrates (aqueous + organic) in the range 200 - 500 mL/h. Although it was gradually optimized, the alternative separation processes currently under study triggered the need for a new MS model, more convenient to extracting systems with low transfer kinetics (e.g. enabling appropriate control of the interfacial area and of the residence time), and that could handle larger variations of the liquids properties (e.g. higher viscosity) or of the operating conditions (e.g. when aqueous and organic flowrates are very different). The newly developed apparatus, named mixer-settler "Next Generation", is shown on