Pertraction of Lu(III) in a hollow fibre contactor

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Due to the favorable radionuclide ^{177}Lu radiophysical characteristics, a great number of radiochemistry and radiopharmacy investigations is orientated toward the potential ^{177}Lu-radiopharmaceutical production and its application in endoradiotherapy of malignant tumors. One of the main phases in the production process of ^{177}Lu-radiopharmaceutical applicable for human *in vivo* use is its separation from the unbound radionuclide harmful to the patient's healthy tissue. The most common technique for purification of radiopharmaceuticals is cation-exchange chromatography. Supported liquid membrane (SLM) extraction, also named pertraction, offers the possibility for the separation of the unbound radionuclide from the labeled radiopharmaceutical, after the labeling procedure is completed.

Pertraction is an extraction technique that involves simultaneous extraction and re-extraction. The organic phase (extractant) is filled into the pores of a microporous hydrophobic membrane, while the feed (donor) and the stripping (acceptor) solutions are placed at the opposite sides of the membrane. In our previous articles[1,2], we proposed the application of miniaturised flat-sheet and hollow fibre SLM microextraction system for the removal of the free ^{177}Lu(III) from the ^{177}Lu(III)-labelled compound. Acceptor volume in both microextraction systems was 10 to 15 μl. Both systems showed, under the optimal extraction conditions (donor flow rate, pH of the donor and acceptor phase, content of the organic phase, membrane length, etc.), high efficiency for the removal of Lu(III) from the aqueous to organic phase (which is of primarily importance for purifying of labelled compounds) and less efficient recovery of Lu(III). The recovery (re-extraction) of Lu(III) is less efficient due to the either high resistance in the membrane or in the acceptor.

The purpose of this study was to investigate the pertraction of Lu(III) with DEHPA as a carrier in a continuous hollow fibre contactor operated in a recirculated mode, with the aim of improving the efficiency of re-extraction of Lu(III) from the organic to the acceptor phase.

The pertraction of Lu(III) was performed in the hollow fibre membrane contactor consisting of the U-shaped glass tube, and one 12 cm long polypropylene hollow fibre membrane ACCUREL PP 50/280, with wall thickness of 50 μm and porosity of 60% (Membrana GmbH, Wuppertal, Germany). The membrane was impregnated with 5% DEHPA in di-n-hexyl ether (DHE) for 30 s. The donor solution (11 μM Lu(III) in the 0.2 M sodium acetate buffer, pH 3.5) was fed along the shell side (at different flow rates, 0.4 – 5.3 $cm^3\ min^{-1}$) in recirculated mode of operation by a peristaltic pump. The acceptor (2 M HCl) solution was recirculated (at 0.4 $cm^3\ min^{-1}$ flow rate) along the lumen of the hollow fibre. Samples were periodically collected from both the donor and the acceptor reservoir. The withdrawn volume is sufficiently small to keep the total volume constant. The Lu(III) concentration in the samples was determined by the indirect voltammetric method[3].

The efficiency of Lu(III) transfer through the liquid membrane is evaluated by three parameters: pertraction efficiency (*P*), that represents a fraction of Lu(III) initially present in the donor phase that was found in the acceptor after extraction, removal efficiency (*E*), that represents the fraction of Lu(III) removed from the donor to the organic phase and memory effect (*M*), that represents the fraction of Lu(III) captured in the organic phase after

extraction. According to the data found in the literature[4,5] and our previous results[1,2], transport of Lu(III) across SLM depends on pH of the donor solution, i.e., the species of Lu-DEHPA complex. In spite of the fact that the Lu-DEHPA complex, permeable across membrane and responsible for the transport of Lu(III) from the donor to the acceptor, is formed on the donor pH 2.5 – 3.5 the value of pertraction efficiency (20%) pointed out that Lu-DEHPA complex was accumulated in the membrane. However, the distribution coefficient of Lu(III) between acceptor and organic phase, which was determined by static liquid-liquid extraction with equal volumes of the phases, was 0.1, meaning that almost all of the Lu(III) was re-extracted. With regards to these facts, it is assumed that, except for the hindered diffusion, a small acceptor volume could be the reason for low re-extraction, i.e., pertraction of Lu(III). The influence of the acceptor volume (Fig. 1) and the donor flow rate (Fig. 2) on the pertraction of Lu(III) was observed.

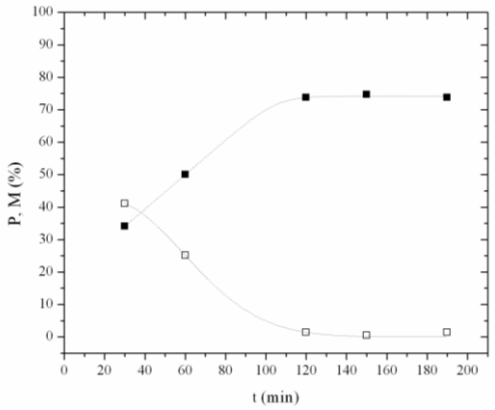

Figure 1. Time variation of the parameters P (■) and M (□) in a recirculated mode of operation

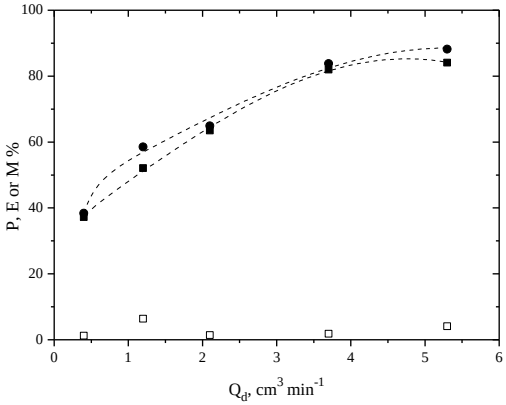

Figure 2. Effect of the donor flow rate (Q_d) on the parameters P (■), E (●) and M (□)

Experimental results show that the acceptor volume considerably influences the transport of Lu(III) from the organic to the acceptor phase, and thus influences the pertraction. It is obviously from Fig. 1 that a part of the extracted Lu(III) that is left in the organic phase is continuously decreasing with the extraction time. After 120 min of the pertraction process less than 1% of the extracted Lu(III) remained in the organic phase.

Figure 2 presents the influence of the donor flow rate on the pertraction parameters. Results show that the extraction and pertraction efficiency increase with the increasing of the donor flow rate. Time variation of the extraction and pertraction efficiency, i.e., Lu(III) concentration in the donor and acceptor phase at different donor flow rates enables determination of the overall mass transfer coefficient for the applied SLM system.

The continuous extraction system with the acceptor recirculation enables almost complete re-extraction of Lu(III) into the acceptor. Thus, the application of the continuous hollow fibre contactor operated in the recirculation mode could be efficiently used for Lu(III) removal from the feed solution, but also for its recovery in order to manage its reusage.

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