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Application of membrane contactor with helical flow for processing uranium ores

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ABSTRACT

The paper presents the studies of the possibility of leaching uranium ores using a membrane contactor. Membrane contactor with helical flow equipped with a tubular metallic membrane was used as an alternative to the traditional method of leaching using the stirred tank. The influence of such process parameters as: the feed flow rate and rotation frequency of the inner cylinder placed in the membrane module on leaching efficiency was investigated. High effectiveness of leaching uranium and associated metals was achieved by using the membrane contactor equipped with the rotor. Among many advantages of a new approach of conducting the leaching process, energy saving and the possibility of combining two process steps in one apparatus: leaching and separation of solid phase from post-leaching solutions, should be mentioned. Furthermore, the application of a helical membrane contactor allows the control of hydrodynamic conditions inside the apparatus and promotes turbulence. This ensured stable and efficient separation of phases after the leaching process by decreasing the intensity of the membrane blocking phenomenon by suspended particles of uranium ore.

1. Introduction

The technology of uranium recovery from its ores is essentially modern, although some of its elements have evolved from extraction technologies of other metals such as gold or vanadium (Fleming, 1986; McKetta, 1997; Pretorius, 2012; IAEA, 1993). Extraction of uranium from ores generally involves such operations as: crushing and milling, leaching, solid-liquid separation with washing, concentration of uranium liquors, precipitation of the product, its purification and finally drying or calcining (Edwards and Oliver, 2000; IAEA, 1990). Uranium can be leached from the ore using either acid or alkaline solutions, with or without heating and with or without the addition of oxidants such as atmospheric oxygen, sodium chlorate, ferric oxide, manganese dioxide and peroxides. Acid leaching is the predominant process for uranium recovery from the rocks (IAEA, 2001; Kim et al., 2014). Mostly, sulfuric acid is used due to its low cost and availability. In comparison with acid processing, alkaline leaching has the advantage of being selective for uranium. The most common alkaline leaching solution is a mixture of sodium carbonate and sodium bicarbonate (El-Nadi et al., 2005).

Leaching studies were performed in the Centre of Nuclear Chemistry and Radiochemistry at Institute of Nuclear Chemistry and Technology in order to develop a suitable method for extraction of uranium from Polish uranium ores: dictyonema shale and sandstones (Frąckiewicz et al., 2012; Gajda et al., 2015). The influence of such process parameters like temperature, pressure, particle size of solid materials (ores), type of leaching solution and its concentration on the recovery efficiency of uranium and associated metals, was studied. The efficiency of uranium leaching from dictyonema shale using sulfuric acid solution achieved 81% and from sandstones ranged 71–100% using sulfuric or hydrochloric acid. Satisfactory results were obtained also for alkaline leaching process.

The technology of uranium extraction in general is well elaborated, however, there is still a need of increasing a process efficiency due to the fact of high cost of whole venture. This can be achieved in various ways, primarily by developing innovative processes based on new materials and novel apparatus designs. It can be done by increasing the efficiency of each process step but it is usually associated with a significant increase in process parameters. Another way is to combine a few process stages in one step. There are some examples in the literature where authors present modifications of the process for uranium recovery, which are based on combination of several process steps in one stage in order to improve the efficiency and economics of the process. Abdel-Rehim (2002) has studied an innovative method for processing Egyptian monazite by alkaline leaching in ball mill autoclaves, where grinding and leaching of monazite took place simultaneously. The author concludes that the main advantages of proposed process is that grinding and leaching of monazite can be combined together in one process and preliminary grinding of monazite concentrate is not required.

Abbreviations: CTF, Couette Taylor flow; MRT, mean residence time.

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Table 1
The chemical composition of uranium ore used in the studies.

Element	Concentration [mg/kg]	Detection limit [ng/L]
U	106 ± 7	23
Th	10.7 ± 2	70
Cu	205 ± 11	165
Co	15.1 ± 1	10
Zn	7488 ± 491	140
La	40 ± 2	15
V	1431 ± 83	35
Mo	63 ± 5	1085
Ni	142 ± 4	55
Sb	6.1 ± 1	245
Fe	27,200 ± 1795	11,500

The present work, described in this paper concerns the combination of two process steps: leaching and phase separation in one apparatus. For this purpose, the membrane contactor with helical flow equipped with tubular metallic membrane was used. Application of similar construction of the membrane module for radioactive waste processing was described in the previous paper (Zakrzewska-Trznadel et al., 2009). It was found that the helical flow increased the filtration efficiency when applied for the treatment of solutions containing macromolecules and suspensions. Other advantages of the helical apparatus were also revealed such as good mixing and high mass transfer coefficients between liquid and the membrane. All these features seem to be very advantageous in uranium leaching process.

Results from uranium leaching conducted in the membrane contactor were compared with those obtained in experiments carried out using mixer-settler system. Leaching process using mixer-settler system was described in detail in the previous paper (Frąckiewicz et al., 2012). This process was conducted in the stirred tank at 80 °C for 8 h, using 10% sulfuric acid.

2. Material and methods

2.1. Chemicals

All chemicals used in experiments were of analytical grade. Sulfuric acid with min. concentration of 95% (POCH BASIC) was used as a leaching agent. As an oxidizing agent manganese dioxide (Centrofarm) was used. Uranium ore (dictyonema shale samples) used in the studies originated from Bransk wellbore. The chemical composition of the ore determined by using inductively coupled plasma mass spectrometry (ICP-MS) is shown in Table 1. The ICP-MS instrument ELAN DRC II (Perkin Elmer) with a cross-flow nebulizer and with a Scott double-pass spray chamber and Ni cones was used.

Table 2
Membrane characteristics.

Membrane geometry	Single tube
Diameter, mm	30/34
Length, mm	450
Filtration area, m^2	0.04
Diameter of the rotor, mm	20
Membrane pore size, μm	0.1

2.2. Membrane contactor

The contactor used in the studies of uranium leaching was a custom made membrane module with tubular membrane. The system and its purpose were described in European Patent (Zakrzewska-Trznadel et al., 2015).

In the inner space of the membrane the rotating shaft creating helical flow was placed in the acentric manner. The scheme of the membrane contactor is shown in Fig. 1. In the annular space between the membrane and the rotor the Couette Taylor flow (CTF) is generated. CTF is a combination of the axial Poiseuille flow and rotating Couette flow with axisymmetric Taylor vortices.

In the membrane contactor used in experiments, the SIKA-R 0.1 AS (GKN Sinter Metals) metallic tubular membrane with 0.1 mm medium pore size was inserted. Chemical composition of the metallic membrane was as follows: 16–18% Cr, 10–14% Ni, 2–3% Mo, and the rest 65–72% - Fe. Tubular membrane SIKA-R 0.1 made of sintered alloys shows chemical stability against aggressive media, acids and alkaline solutions (SIKA-R…IS/AS, n.d., Brochure). The membrane characteristics are presented in Table 2.

2.3. Experimental methods

2.3.1. Leaching in the membrane contactor

The membrane contactor applied in uranium leaching experiments was a part of the experimental set-up showed schematically in Fig. 2. The set-up was equipped with the measurement and control devices such as flowmeters and manometers as well as with the system of rotation control composed of the rotor drive (4) and the inverter (Hitachi) (3) that allows the rotation frequency adjustment during the leaching and filtration process.

The procedure of leaching of uranium ores was as follows: portion of 3 g of uranium ore after crushing using ball mill (MM400, Retsch) was calcinated in the muffle furnace for 4 h at 550 °C. The previous studies with simple stirred cell showed that the most effective leaching was achieved with a highly shredded ore when particle size <0.2 mm (Frąckiewicz et al., 2012; Gajda et al., 2015). Such a fraction of the ground ore was used in present experiments.

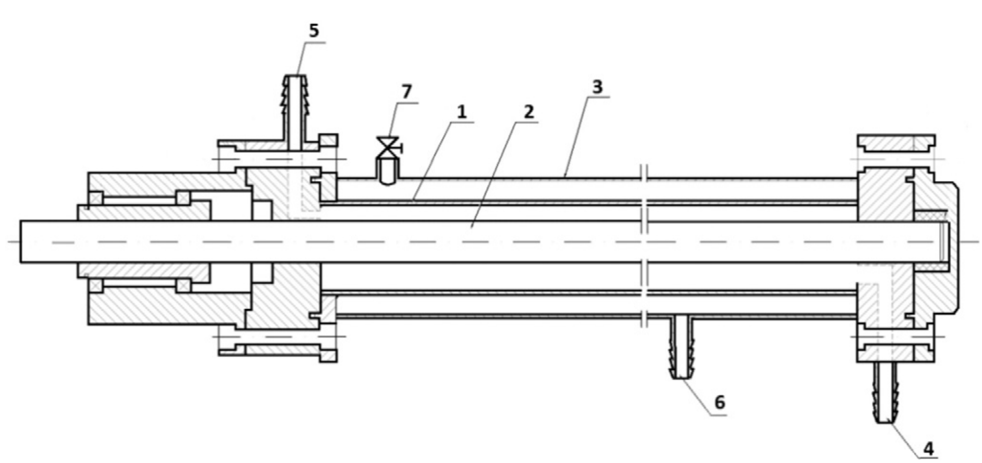

Fig. 1. Schematic diagram of the membrane contactor; 1 – tubular membrane, 2 – rotor, 3 – housing, 4 – feed inlet, 5 – feed outlet, 6 – permeate outlet, 7 – vent.