Separation and Purification Technology 177 (2017) 8–20

Contents lists available at ScienceDirect

Separation and Purification Technology

journal homepage: www.elsevier.com/locate/seppur

Removal of cobalt ions from aqueous solution by forward osmosis

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ARTICLE INFO

Article history:
Received 4 May 2016
Received in revised form 27 October 2016
Accepted 15 December 2016
Available online 23 December 2016

Keywords:
Forward osmosis
Co(II) retention
Operational condition
Radioactive wastewater

ABSTRACT

The performance of Co(II) removal from aqueous solution by forward osmosis (FO) was investigated. NaCl solutions were used as the draw solutions (DS). The effects of membrane characteristics, feed solutions properties, DS concentration, cross-flow rates on FO performance (water flux, Co(II) flux and retention, and reverse NaCl flux) were determined. The separation mechanisms under the various operational conditions were discussed. Changes of the FO membrane after Co(II) separation were characterized. The results show that CTA-ES featured the highest water fluxes of 15.5 ± 0.5 L m^{-2} h^{-1} at AL-FS and 23.4 ± 2.5 L m^{-2} h^{-1} at AL-DS among the three membranes. The active layer charge of the FO membrane was a main factor affecting Co(II) retention by FO. The texture of the support layer played a great role in the water permeability and reverse NaCl flux. The optimal operational conditions for Co(II) retention was NaCl concentration of 1.0 M in DS, the cross-flow rates of 11 and 5 cm s^{-1} at the feed and DS sides respectively. After used, the active layer of the CTA-ES membrane became more rough and hydrophobic. Co might attach to the membrane and became the membrane foulants. FO process could be an alternative technology for radioactive wastewater.

1. Introduction

Along with the rapid development of nuclear industry in China, more and more radioactive wastewater will be produced from the nuclear power plants (NPPs) [1,2]. The radioactive wastewater contains a large plenty of radionuclides (Co, Cs, Sr, etc.) with 10^4–10^6 Bq L^{-1} activity concentrations, other impurities (organic complex builders, non-radioactive dissolved salts, suspended particulates, biofoulants) [3,4], which are harmful to ecological environment and human health. The treatment of radioactive wastewater has become an urgent problem needed to be solved. Radionuclides exist in the radioactive wastewater as the form of cations. Radioactive wastewater treatment aims to separate the metal cations from the radioactive wastewater. The conventional treatment technologies include traditional filtration, chemical precipitation, sedimentation, adsorption, ion exchange, thermal evaporation, biological methods and so on [5,6]. With very high quality of the produced water, the membrane separation technologies have been successfully used to treat the radioactive wastewater, including micro-filtration (MF) [7], nano-filtration (NF) [8], membrane distillation (MD) [2,6] and reverse osmosis (RO) [9,10]. RO, which features high water recovery rate and high salt retention, has been considered as one of the most efficient technologies for radioactive wastewater treatment so far [11]. However, RO consumes high power, which significantly increases the cost of radioactive wastewater treatment.

Forward osmosis (FO) is an emerging technology, in which water molecules are transported across a RO-like membrane driven by the osmotic pressure difference. Compared to RO, FO features equally high contaminant rejections [12,13], lower membrane fouling [12,14–16] and easier equipment [17,18] due to the lack of applied hydraulic pressure [19]. FO performance has been investigated for desalination [20–22], wastewater treatment and drinking water treatment [23–27]. However, the draw solution (DS) recovery consuming high energy is one of the prominent problems blocking FO wide application [28].

In some cases for some special water treatment, FO exhibits its distinctive benefits rather than other membrane technologies. In this study, we propose FO for the treatment of intermediate or low level radioactive wastewater (ILRW or LLRW) produced from the NPPs. Most of the NNPs are located by the seaside with great amount of seawater available for cooling. When FO process is applied for ILRW and LLRW treatment, plenty of seawater is available to be applied as DS for FO. Since FO retention is as high as RO, the nuclides in the radioactive wastewater could be well rejected.

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http://dx.doi.org/10.1016/j.seppur.2016.12.025
1383-5866/

Nomenclature

A	effective membrane surface area (m^2)
$C_{f,DS}$	final concentration of Co(II) in draw solution (mg L^{-1})
$C_{i,DS}$	initial concentration of Co(II) in draw solution (mg L^{-1})
$C_{fNa,FS}$	final NaCl concentration of feed solution (mg L^{-1})
$C_{iNa,FS}$	initial NaCl concentration of feed solution (mg L^{-1})
$C_{i,FS}$	initial concentration of Co(II) in feed solution (mg L^{-1})
C	solute concentration (mg L^{-1} or Moles or M)
$J_{Co(II)}$	Co(II) flux (mg m^{-2} h^{-1})
J_{NaCl}	reverse NaCl flux (g m^{-2} h^{-1})
J_w	water flux (L m^{-2} h^{-1})
n	Van't Hoff factor
R[%]	Co(II) retention (%)
R_a	mean surface roughness (nm)
R_{max}	maximum vertical distance between the highest and lowest data points (nm)
R_q	mean-square surface roughness (nm)
R_g	universal gas constant (0.0821 L atm mol^{-1} K^{-1})
T	absolute temperature (K)
$V_{f,DS}$	final volume of draw solution (L)
$V_{f,FS}$	final volume of feed solution (L)
$V_{i,FS}$	initial volume of feed solution (L)
V_P	volume of permeated water (L)
v_{FS}	feed solution flowrate (cm s^{-1})
v_{DS}	draw solution flowrate (cm s^{-1})
Δm	permeated water weight (g)
Δt	measuring time interval (h)
ρ	density of water (g cm^{-3})
π	osmotic pressure (atm)

Abbreviations

AFM	atomic force microscope
AL	active layer
AL-FS	membrane orientation of active layer facing feed solution
AL-DS	membrane orientation of active layer facing draw solution
CTA-ES	cellulose triacetate with embedded polyester screen support
CTA-NW	cellulose triacetate on heat-or RF-weldable nonwoven support
DS	draw solution
ECP	external concentration polarization
FO	forward osmosis
FS	feed solution
ICP	internal concentration polarization
ILRW	intermediate-level radioactive wastewater
LLRW	low-level radioactive wastewater
MD	membrane distillation
MF	micro-filtration
NF	nano-filtration
NPP	nuclear power plant
RO	reverse osmosis
SEM	scanning electronic microscope
SL	support layer
TFC-ES	thin-film composite with embedded polyester screen support

The used DS might be discharged to the deep sea without any high energy consuming post-processing. Brine from desalination plants is an alternative DS, which might achieve high flux than the seawater. In addition, the used DS (diluted brine) achieved beneficial environmental impact rather than direct discharge of the brine from desalination plants with high salinity.

Besides, low membrane fouling of FO lessens the membrane replacement frequency and reduces the radioactive nuclide adhesion on the membrane surface, which produces less radioactive solid wastes for final radio waste disposal.

FO performance is currently investigated mostly on micropollutants, pharmaceuticals or other organic contaminants removal [13,29–32]. The investigations on inorganics removal is concentrated in desalination [12,20–22,25,33,34]. Other investigations on inorganics removal are relatively limited to several anion and cation species, such as boron [35–37] and arsenic [35,38]. Operational conditions have been found to play an important role in FO performance. Membrane material has a major impact on the pollutant rejection and water flux [21,39]. DS concentration and crossflow velocity influence the process performance as well [37,39]. However, limited experience could be provided for ILRW or LLRW treatment by FO. It is necessary to widen the target pollutant species for FO application.

When FO is applied for ILRW or LLRW treatment, it may face some challenges regarding environmental concerns of the used DS. Nuclides and other pollutants possibly transfer to the DS due to incomplete rejection by membrane, which would cause radioactive or environmental risk when the used DS was discharged to the sea. Gross α^- and β^- radioactivity in the discharged wastewater should be lower than 1 and 10 Bq L^{-1} according to Integrated Wastewater Discharge Standard [40]. Hence, FO performance in rejecting nuclides and pollutants has to be investigated before FO applied for LLRW and ILRW treatment. If the used DS could not meet the Standard by only one-stage FO, post-treatment processes e.g. RO, two-stage FO might be required. So far, LLRW and ILRW are treatment by multi-stage RO in many NPPs [41].

Cobalt(II) ion is a common heavy metal pollutant, which is harmful to humans and animals. ^{60}Co(II) ion is a frequent radionuclide in the radioactive wastewater. In this study, the non-radioactive isotope ^{59}Co(II) was used to simulate the radionuclide. FO performance for Co(II) ion separation from aqueous solution was investigated. Three commercial FO membranes were applied. NaCl solutions (simulated seawater) were used as DS. The factors affecting FO performance were focused on, including: membrane characteristics, feed solutions (FS) (simulated radioactive wastewater) properties, DS concentration, cross-flow rates. Changes of the FO membrane after Co(II) ion separation were characterized. FO performance for Co(II) ion separation was finally compared with other separation processes.

2. Methods and materials

2.1. FO set-up and membrane

The scheme of the FO set-up is given in Fig. 1. The heights of the flow channels for DS and FS were both 2 mm and the effective area of the FO membrane was 40.5 cm^2. For the experiments, FS and DS flowed countercurrently in each channel on both side of FO membrane. The flows were circulated along the system by using two gear pumps (BT600-2J, China).

Three commercial FO membranes (HTI, US) were applied, namely Thin-Film Composite with embedded polyester screen support (TFC-ES), Cellulose Triacetate with embedded polyester screen support (CTA-ES), Cellulose Triacetate on heat-or RF-weldable non-