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Separation of divalent and monovalent ions using flow-electrode capacitive deionization with nanofiltration membranes

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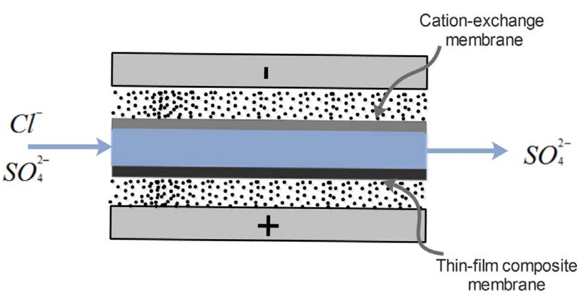
GRAPHICAL ABSTRACT



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ABSTRACT

We report on selective separation of monovalent and divalent cations (Na^+ and Mg^{2+}) and anions (Cl^- and SO_4^{2-}) from aqueous solutions using the flow electrode capacitive deionization (FCDI) process, operated with ion-exchange and nanofiltration membranes (NF). For the selective separation of cations and anions the FCDI module was operated with an NF membrane (NF270) and an anion-exchange or cation-exchange membrane, respectively, at varying applied cell potentials (0.6, 0.8 and 1.23 V) and initial mono- to di-valent ions molar concentration ratios of 1, 10 and 20. The permselectivity of the NF270 membrane, calculated as a ratio between measured ionic fluxes, was found highly dependent on the initial molar concentration ratios of the mono- to the di-valent ions. Concentration-normalized Na^+ to Mg^{2+} permselectivity was 0.69–1.04, indicating that the NF270 membrane does not pose selectivity for the separation of sodium and magnesium in the studied process. Conversely, the concentration-normalized permselectivity between Cl^- and SO_4^{2-} was found between 1.28 and 7.03 depending on the applied cell potential, indicating high potential for implementing the proposed NF-FCDI method for selective separation of anions.

1. Introduction

Development of sustainable technologies for water desalination, water reuse and recovery of valuable chemicals from water, and wastewater and process streams is a major technological challenge [1–4]. Membrane technologies, mainly pressure driven and electrochemical, play a significant role in these processes. For example, nanofiltration was shown effective for the separation of Mg^{2+}, Ca^{2+}, and SO_4^{2-} from seawater [5,6,7]. Electrodialysis with monovalent-selective ion exchange membranes has been studied for selective separation of divalent ions from seawater [8], lithium from magnesium in salt lake brines [9], phosphate from synthetic municipal wastewater [10] and others.

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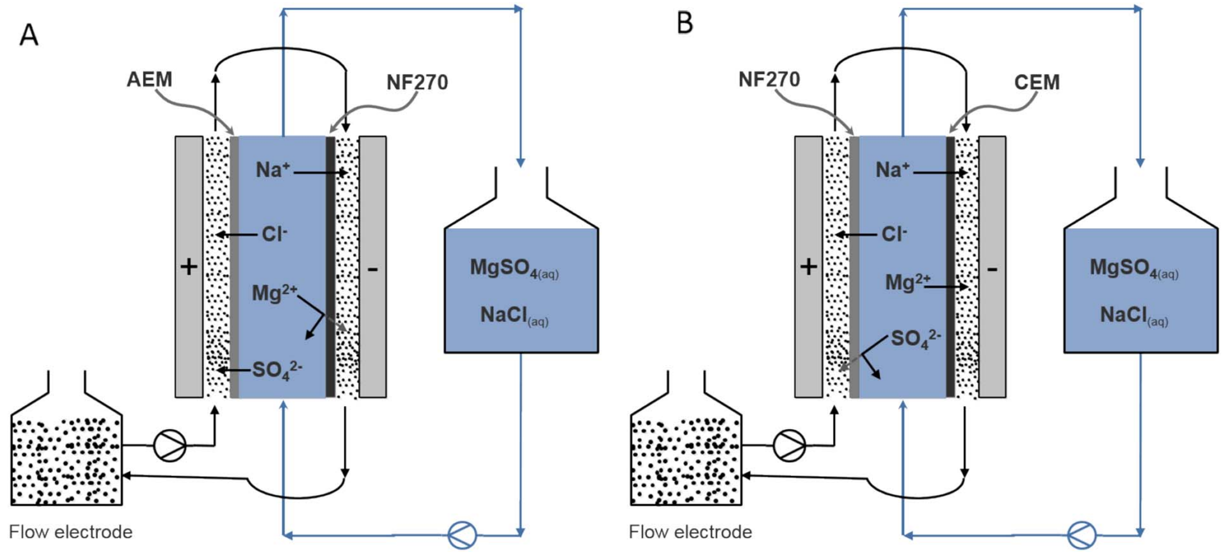

Fig. 1. Flow-electrode capacitive deionization with a nanofiltration membrane(s) and a single flow electrode for the separation of monovalent and divalent cations ((A)-AEM-NF-FCDI assembly) and anions ((B) - CEM-NF-FCDI assembly).

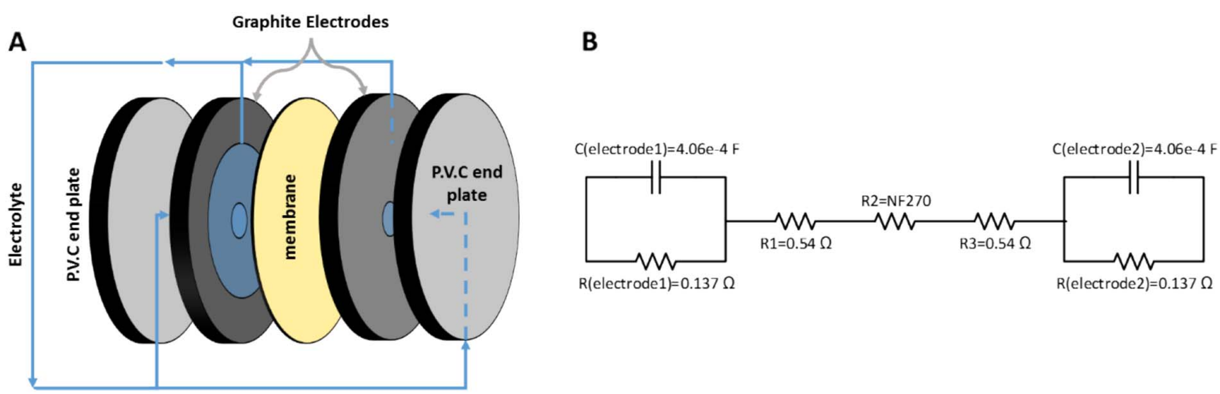

Fig. 2. Schematic structure of the electrochemical cell used for the EIS measurements (A), and the derived equivalent circuit (B). [NaCl] = 100 g/L, frequencies range – 0.1–10^5 Hz, effective membrane and electrodes' area – 5.76 cm^2.

Table 1
Experimental parameters applied in the NF-FCDI studies and obtained permselectivities. $(M^{\pm 1}/M^{\pm 2})_0$ – initial molar ratio of monovalent and divalent ions in the electrolyte solution; $[M^{\pm 1}]_0$, $[M^{\pm 2}]_0$ - initial molar concentrations of monovalent and divalent ions, respectively; $V^0_{electrolyte}$, $V^0_{electrode}$ - initial volume of the electrolyte solution and of the flow electrode, respectively. P_{di}^{mono} – permselectivity (calculated using Eq. (3)); $P_{di}^{mono}/\left(\frac{M^{\pm 1}}{M^{\pm 2}}\right)_0$ – selectivity normalized to the initial ratio of mono- and divalent ions.

	Exp#	$(M^{\pm 1}/M^{\pm 2})_0$ (mol/mol)	$[M^{\pm 1}]_0$ (mM)	$[M^{\pm 2}]_0$ (mM)	Cell potential (V)	$V^0_{electrolyte}$ (mL)	$V^0_{electrode}$ (mL)	P_{di}^{mono}	$\frac{P_{di}^{mono}}{\left(\frac{M^{\pm 1}}{M^{\pm 2}}\right)_0}$	C.E.
NF-AEM-FCDI	1	1	120	6	0.6	200	100	0.69	0.69	84%
	2				0.8			0.72	0.72	74%
	3				1.23			0.72	0.72	69%
	4	10	60		0.6			8.8	0.88	58%
	5				0.8			8.44	0.84	56%
	6				1.23			9.1	0.91	56%
	7	20	6		0.6			20.7	1.04	94%
	8				0.8			19.4	0.97	75%
	9				1.23			19.1	0.96	53%
NF-CEM-FCDI	10	1	120	6	0.6	100	250	6.9	6.90	27%
	11				0.8			3.2	3.19	36%
	12				1.23			5.9	5.92	25%
	13	10	60		0.6			34.1	3.41	37%
	14				0.8			43.5	4.35	35%
	15				1.23			80.7	8.07	31%
	16	20	6		0.6			25.6	1.28	29%
	17				0.8			82.9	4.15	40%
	18				1.23			138.3	6.92	58%