A Novel Electrochemical Flow Reactor with Improved Mass Transfer – The *e*$^{-}$NETmix

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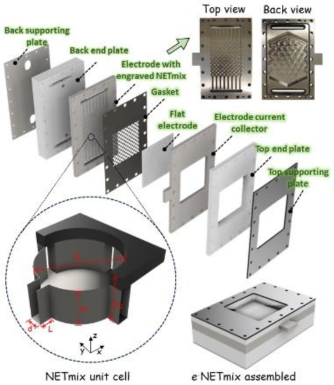

This study introduces a novel electrochemical flow reactor named *e*$^{-}$NETmix, which comprises a unique static mixer for enhanced mass transfer. This static mixer is referred to as NETmix and features cylindrical chambers connected by prismatic channels at a 45° angle. In the current reactor configuration, the NETmix static mixer is imprinted in a plate acting simultaneously as a fluid distributor and an electrode. The other electrode corresponds to a flat plate. The reactor was characterized as regards mass transfer employing the limiting current technique for the reversible ferricyanide-ferrocyanide reaction. The *e*$^{-}$NETmix exhibited volumetric mass transfer coefficients ($k_m A$) ranging from ~5.7×10^{-2} s^{-1} to ~2.5×10^{-1} s^{-1} for Reynolds numbers (*Re*) from 130 to 1400. It outperformed existing electrochemical flow reactors, in which $k_m A$ values typically do not surpass 10^{-3}. The *e*$^{-}$NETmix excels in mass transfer mainly due to the NETmix static mixer, which encourages the development of a laminar chaotic flow regime.

Introduction

Electrochemical flow reactors revolutionized electrochemical engineering by allowing for increased mass, heat, and photon transfer characteristics, energy performance, scalability, process safety, automation, and reproducibility [1, 2]. Key reactor designs include parallel plate reactors, also named filter-press cells [3, 4]. These reactors typically involve solid-phase electrodes, which rely on efficient mass transfer for substrate transport, adsorption, and subsequent desorption of products from the electrode surface [5]. The use of narrow interelectrode gaps, static mixers, and structured electrodes (especially three-dimensional) play a crucial role in optimizing mass transfer in electrochemical flow reactors [6, 7].

This study focuses on the development of a new electrochemical flow reactor incorporating a specific static mixer, the NETmix, and its characterization in terms of mass transfer properties. The new reactor is named *e*$^{-}$NETmix. A provisional patent application on this electrochemical flow reactor was recently filled [8]. The NETmix static mixer is composed of cylindrical chambers interconnected by prismatic channels arranged at 45° angles. The current *e*$^{-}$NETmix configuration is composed of a plate with the NETmix static mixer engraved, working as an electrode and a fluid distributor, and a flat plate, acting exclusively as an electrode. This reactor is optimized regarding the fluid inlet/outlet scheme, chamber and channel dimensions, electrical insulation, and fluid temperature control.

Material and Methods

In more detail, the applied *e*$^{-}$NETmix reactor comprises two electrodes: (i) a 316-grade stainless steel plate with an imprinted fluid distributor based on the NETmix static mixer on one side and a heat exchanger on the opposite side for fluid temperature control, and (ii) a 316-grade stainless steel flat plate electrode with length × width of 100 mm × 100 mm or 49.0 mm × 49.0 mm that was placed in a stainless steel current collector. The NETmix structure includes chambers with a diameter of 6.65 mm, and channels with a width and length of 1.0 mm. Additionally, there are 18 side chambers with a diameter of 4.7 mm. The fluid enters and leaves the reactor through a single inlet/outlet port connected to a trapezoid-shaped slot. The NETmix electrode served as the anode and counter electrode. The flat plate electrode acted as the cathode and working electrode. The anode and the cathode were separated by a gasket made of ethylene propylene diene monomer (EPDM) with 1.0 mm thickness to prevent short circuit and simultaneously ensuring the fluid circulation exclusively in the NETmix network. Frontal and back polyethylene terephthalate (PET) end frames are responsible for holding the components together and sealing the reactor by mechanical compression. Frontal and back stainless steel supporting sheets give resistance to these end frames. Various EPDM O-rings were employed to prevent leakages. A 86.5 mm × 86.5 mm window or a 37.8 mm × 37.8 mm window exists at the top end frame/supporting sheet to allow for backside illumination of the flat plate electrode in light-assisted processes.

The mass transfer was assessed by the limiting current technique using the ferricyanide ($Fe(CN)_6^{3-}$)-ferrocyanide ($Fe(CN)_6^{4-}$) reversible reaction [9]. For each run, a reactive solution was prepared by adding 1.0 mM or 0.60 mM $K_3Fe(CN)_6$ and 10 mM or 6.0 mM $K_4Fe(CN)_6$ to ultrapure water for the 100 mm × 100 mm or the 49.0 mm × 49.0 mm cathode, respectively, together with 0.5 M Na_2CO_3. The solution (500 mL) was recirculated in the system until reaching a temperature of 25±1 ºC. The flow rate varied from 10.3 to 128.5 L h^{-1}, resulting in Reynolds numbers (*Re*) ranging from 130 to 1400. A slow potential scan was applied with an initial cell potential of 0.1 V, gradually increased by 0.05 V or 0.1 V. The scan was terminated when the current intensity exhibited a dramatic increase.

Results and Discussion

In the e^-NETmix reactor with the 100 mm × 100 mm cathode and for *Re* from 130 to 1400, the k_m values ranged from $(7.2\pm0.1)\times10^{-5}$ m s^{-1} to $(2.9\pm0.01)\times10^{-4}$ m s^{-1}, and the k_mA values ranged from $(0.46\pm0.01)\times10^{-1}$ s^{-1} to $(1.8\pm0.1)\times10^{-1}$ s^{-1} (Figures 1 and 2). The Sherwood number (*Sh*) ranged from 170±3 to 669±2 (Figure 3).

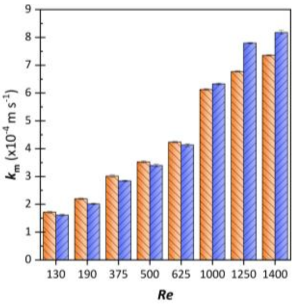

Figure 1. Mass transfer coefficient (k_m) for the optimized e^-NETmix reactor using different *Re*. Legend: ▧ cathode 100 mm × 100 mm; ▨ cathode 49.0 mm × 49.0 mm.

Quite similar k_m, k_mA, and *Sh* values were achieved for the 49.0 mm × 49.0 mm cathode.

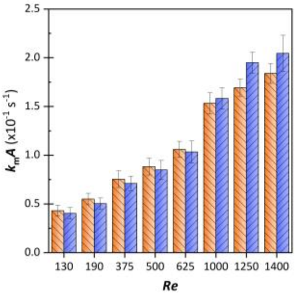

Figure 2. Volumetric mass transfer coefficient (k_mA) for the optimized e^-NETmix reactor using different *Re*. Legend: ▧ cathode 100 mm × 100 mm; ▨ cathode 49.0 mm × 49.0 mm.

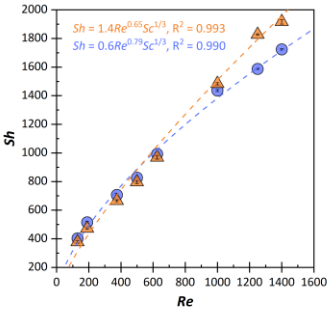

Figure 3. Sherwood number (*Sh*) (experimental and from correlation) for the optimized e^-NETmix reactor using different *Re*. Legend: ▲ cathode 100 mm × 100 mm; ● cathode 49.0 mm × 49.0 mm.

Furthermore, the e^-NETmix reactor outperformed numerous electrochemical flow reactors reported in the literature, showcasing up to ~25, ~70, and ~9-fold higher values for k_mA, k_m, and *Sh*, respectively.

Conclusions

The optimized e^-NETmix reactor showed enhanced mass transfer properties that can be attributed to the unique NETmix fluid distributor geometry, which promotes convection and a laminar chaotic flow regime.

Acknowledgments

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