



Review

Co-laminar flow cells for electrochemical energy conversion



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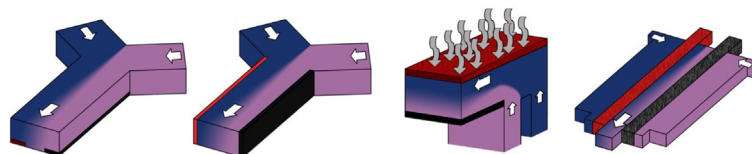
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HIGHLIGHTS

- Categorization and standardization of co-laminar flow cell technology is proposed.
- The technology is gaining importance as an analytical and educational platform.
- Practical, competitive power density levels are demonstrated.
- Emerging rechargeable 'membraneless' flow batteries are discussed.
- Recommendations for further research are provided.

GRAPHICAL ABSTRACT

This review discusses the recent trends and opportunities in the use of co-laminar flow of reactants as an alternative to membranes for power generating electrochemical cells.



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ABSTRACT

In this review, we present the major developments in the evolution of 'membraneless' microfluidic electrochemical cells which utilize co-laminar flow to minimize reactant mixing while producing electrical power in a compact form. Categorization of devices according to reactant phases is suggested, with further differentiation being subject to fabrication method and function, namely multi-layer sandwich structures for medium-power cell stacks and single-layer monolithic cells for low-power on-chip applications. Power density metrics reveal that recent co-laminar flow cells compare favourably with conventional membrane-based electrochemical cells and that further optimization of device architecture could be expedited through standardized testing. Current research trends indicate that co-laminar flow cell technology for power generation is growing rapidly and finding additional use as an analytical and education tool. Practical directions and recommendations for further research are provided, with the intention to guide scientific advances and technology development toward ultimate pairing with commercial applications.

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1. Introduction

Chemical energy storage has been the method of choice for portable power applications due its high specific energy [1]. Electrochemical energy conversion is considered galvanic when the chemical energy of two half-cell reactions is converted into electrical energy, whereas it is electrolytic when electricity is used to produce species of higher chemical energy in the reverse process

[2]. Historically, the term fuel cell has been applied to any galvanic electrochemical cell which converts the chemical energy of a fuel and oxidant combination directly into electrical energy [3]. More recently, the term redox flow battery (RFB) has become popular to describe electrochemical cells which utilize two redox couples dissolved in separate liquid electrolytes as the fuel and oxidant [4]. As thermodynamically open systems, both of these electrochemical flow cells can be instantly recharged with new reactants, a desirable property which eludes conventional closed-cell batteries and may offer a potential solution to the growing demand for compact energy supplies for portable electronics and a host of other low-power applications [1]. Occasionally, fuel cells have been used for

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the reverse electrolytic process of recreating the same fuel and oxidant combination by applying an external potential, such as the regenerative systems developed by NASA for long term space missions [5]. Since reactants often undergo phase changes during charge and discharge, these regenerative fuel cells typically suffer from low cycle efficiency due in part to the difficulty of optimizing the structure for both directions of phase change. Essentially operating as continuously recirculating regenerative fuel cells, rechargeable RFBs on the other hand do not suffer from phase change issues and can therefore be conveniently optimized to achieve high cycle efficiencies up to 80% [6].

Regardless of terminology, most modern electrochemical flow cells employ polymer electrolyte membranes to separate the reactants while maintaining the ionic conductivity necessary to complete the circuit. These specialized solid state ionomer membranes are designed to allow either hydrogen ions in acidic conditions or hydroxide ions in alkaline conditions to pass while preventing the crossover of other chemical species. Recent ionomer membrane development has contributed to significant reductions in film thickness, with 25–50 μm being a typical thickness used in hydrogen fuel cells [7] and 125 μm being used in vanadium redox batteries [8]. Although this trend has led to significant performance gains due to reduced ionic resistance, it has also led to certain technical challenges. In the case of hydrogen fuel cells, thinner membranes tend to degrade and rupture more quickly leading to cell failure [9]. In the case of direct methanol fuel cells (DMFC), typical membranes still allow considerable crossover of methanol fuel due to diffusion and electro-osmotic drag [3]. It has also been observed that rates of vanadium redox species crossover in ionomer membranes are different for each ion which eventually imbalances the concentrations of fuel and oxidant, requiring rebalancing of the entire vanadium redox battery [4,10]. In addition to durability and crossover issues, polymer electrolyte membranes considerably increase the cost of electrochemical cells, particularly for alkaline fuel cells which require more expensive hydroxide-permeable anion exchange membranes [11]. Current membrane electrode assembly (MEA) designs are also inadequate for monolithic integration into miniaturized devices [1].

For all of the above reasons, many alternatives have been sought to replace or improve existing polymer electrolyte membranes. Recent advances in the field of microfluidics and nanofluidics enable promising opportunities to eliminate the membrane through hydrodynamic engineering. At the microscale, fluid dynamics is typically characterized by low Reynolds numbers ($Re < 10$) which indicates smooth laminar flow free of chaotic turbulence. This phenomenon permits two laminar streams to flow side by side with mixing occurring only by cross-stream diffusion. Several studies have explored the practical application of this phenomenon for DNA analysis [12], molecular positioning [13], microfluidic switches [14], and a wide range of other applications [15]. It was proposed in 2002 that stratified co-laminar flow of two electrochemical reactant streams would allow for ionic conduction while maintaining reactant purity, thereby replacing the costly membrane [16,17]. Such ‘membraneless’ cells were first demonstrated in 2002, and have been the subject of numerous other studies since.

A detailed and comprehensive review of all types of microfluidic fuel cells was published by Kjeang et al. in 2009 [18]. Since then, the research in this field has not only evolved substantially in the core technical area of microfluidic fuel cells, but has also ventured into the domain of rechargeable batteries. A critical juncture has been reached, where technology pairing with practical, commercial applications is essential for effective further development. The purpose of the present review article is to highlight notable advancements in the field since 2009 and to extract the key

historical trends in the rapid development of electrochemical cells based on co-laminar flow technology. These trends are then used to offer a forward-looking, pragmatic perspective on the advantages and disadvantages of this technology and provide practical directions for future work. The focus of this review is strictly on co-laminar flow cells and the scope does not include other types of microfluidic cells as reviewed elsewhere [18–20].

2. Design evolution

The advent of microfluidics has opened up the door for a variety of non-conventional electrochemical cell architectures that do not require a membrane. The most fundamental co-laminar flow cell design is illustrated schematically in Fig. 1. The design features stratified co-laminar flow of fuel and oxidant in supporting electrolyte through a shared microfluidic channel with electrodes positioned on the walls.

2.1. Design constraints

While the electrochemical reactions and electrode materials are generally consistent with those of conventional electrochemical cells, the membraneless function of co-laminar flow cells has certain requirements on the cell design. Firstly, the flow must be laminar to prevent turbulent mixing of the two streams, which is facilitated by low Reynolds numbers ($Re < 1000$) [21]. Secondly, the residence time t_{res} in the electrochemical chamber must be shorter than the diffusion time t_{diff} for crossover of a reactant species to the opposite electrode in order to avoid a mixed potential. In this situation, the thermodynamic potential difference between the two electrodes would follow the same Nernst relation [2] as any other electrochemical cell with identical reactants. This necessary criterion can

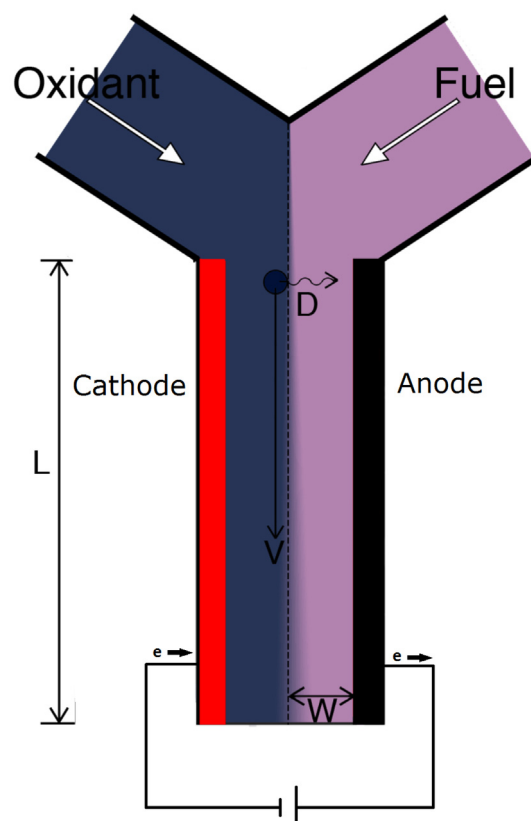


Fig. 1. Schematic of co-laminar flow cell operation.

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