Progress in Energy and Combustion Science 49 (2015) 40–58

Contents lists available at ScienceDirect

Progress in Energy and Combustion Science

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

Review

# Fundamental models for flow batteries

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ARTICLE INFO

*Article history:*
Received 10 September 2014
Accepted 9 February 2015
Available online 13 March 2015

*Keywords:*
Flow batteries
Numerical modeling
Battery performance prediction

ABSTRACT

The flow battery is a promising technology for large-scale storage of intermittent power generated from solar and wind farms owing to its unique advantages such as location independence, scalability and versatility. The widespread commercialization of flow batteries, thus far, is still hindered by certain technical barriers. Removal of these barriers requires a fundamental understanding of the complex electrochemical and transport behaviors of flow batteries. Mathematical modeling and simulation serve important roles in the exploration of these complex phenomena and to the prediction as well as improvement of the cell performance of different system designs. In this review, a comprehensive study is performed to review and summarize state-of-the-art flow batteries and to provide an outlook on the future and potential of flow battery modeling. The review begins with a description of the physical and chemical processes of common flow batteries, followed by the detailed discussion of the governing equations for transports of mass, momentum, heat and charge as well as the electrochemical reactions for porous-medium models. The determination of key transport properties for the porous-medium models and their effects on modeling results are also analyzed. In addition, lattice Boltzmann method, molecular dynamics and density function simulations as well as stack-level network models for flow batteries are reviewed. Finally, the issues facing the future of flow battery modeling are addressed.



**Contents**

1. Introduction ..... 41
2. General description of physical and chemical processes in flow batteries ..... 41
   2.1. All-vanadium redox flow battery (VRFB) ..... 42
   2.2. Bromide/polysulfide flow battery ..... 42
   2.3. Zinc/cerium redox flow cells ..... 42
   2.4. Soluble lead-acid flow battery ..... 43
   2.5. Iron–chromium redox flow battery ..... 43
3. Porous-medium models ..... 43
   3.1. Governing equations ..... 43
      3.1.1. Simplifications and assumptions ..... 43
      3.1.2. Conservation equations for fluid flow ..... 43
      3.1.3. Conservation equations for mass transport ..... 44
      3.1.4. Conservation equations for heat transport ..... 44
      3.1.5. Electrochemical kinetics ..... 44
      3.1.6. Constituent relationships and physicochemical properties ..... 45
   3.2. Determinations of transport properties ..... 46
      3.2.1. Transport properties through porous electrodes ..... 46
      3.2.2. Ions transport properties through membranes ..... 48
      3.2.3. Transport properties of electrolytes ..... 49

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http://dx.doi.org/10.1016/j.pecs.2015.02.001
0360-1285/

4. Lattice Boltzmann method ..... 51
5. Molecular dynamics and density function simulations ..... 52
6. Stack-level network models ..... 52
7. Summary and outlook ..... 55
Acknowledgments ..... 55
Nomenclature ..... 55
References ..... 56

## 1. Introduction

An increase in the utilization of carbon-free, renewable energy sources is the ultimate solution in reducing the impact of $CO_2$ emissions associated with the use of carbon-based energy sources on the climate. Some major renewable energy sources, including wind and solar energy, can supply a significant amount of electrical energy worldwide. However, the intermittent and fluctuant nature of these renewable sources substantially limit their reliability, and are major reasons why currently only comprise a small percentage of primary energy sources [1]. The potential of renewables can only be fully exploited if efficient, safe, and reliable electrical energy storage systems are developed. Over the past few decades, a number of different energy storage technologies have emerged. However, many of them possess inherent limitations and disadvantages, categorizing them to be economically and practically fit for only a narrow range of applications. Among different energy storage technologies, electrochemical systems are superior to others [2,3], primarily because they can provide direct conversion between chemical energy and electrical energy, and offer unique advantages, including low environmental footprints (can be sited near residential areas) and short response time (millisecond timescale, such that they can be used simultaneously for both power quality and energy management applications) [4,5].

In comparison to different electrochemical energy storage technologies such as capacitors or supercapacitors, lead-acid batteries, Ni-metal batteries, and Li-ion batteries, redox flow batteries are the most suitable for large-scale stationary energy storage [6–9]. They offer unique features, including but not limited to: i) low maintenance, ii) tolerance to deep discharge without risk of damage, iii) long lifetime (generally thousands of charge–discharge cycles) compared to that of galvanic batteries and, iv) simplicity in building large systems based on module design [10–17]. In addition, unlike other battery systems, energy is stored in the electrolyte solutions and the capacity of the system is determined by the concentration of the active redox couple species and the electrolyte tank volume of the redox flow battery, while the power rating of the system is determined by the number of cells in the stack and the electrode area. As such, the power and energy capacity of an RFB system can be designed separately.

The rapidly approaching commercialization of redox flow batteries sets a series of new challenges in developing the technology: notably, scale-up and optimization (with respect to electrode geometries and operating conditions), improvement in electrolyte stability, development of electrode materials more resistant to oxidation, and mitigation of membrane fouling [18–28]. Many of these challenges are not particularly well-suited to laboratory analysis alone, by virtue of the associated financial costs and lengthy timescales. Moreover, as the intrinsically coupled physicochemical processes occur simultaneously in the redox flow batteries including heat and mass transfer, electrochemical reactions, as well as ionic and electronic transfer, it is difficult to experimentally quantify the interrelated parameters that govern the flow batteries. Therefore, numerical modeling that incorporates coupled heat/mass transport and electrochemical kinetics becomes essential to gain a better understanding of flow batteries and to shorten the design and optimization cycles.

To obtain a general overview of modeling treatments on flow batteries, this paper summarizes the various issues associated with flow batteries, and presents a critical review on the numerical investigations of each issue.

The remainder of this paper is organized as follows: i) Section 2 introduces the general principles of the five kinds of flow batteries and the physical/chemical processes during operating the flow batteries; ii) Section 3 shows the governing equations and the derivations of key transport properties for porous-medium models; iii) Section 4 reviews the applications of the lattice Boltzmann method; iv) Section 5 reviews the molecular dynamics and density function simulations for flow batteries; v) Section 6 reviews stack-level network modeling works, and vi) Section 7 summarizes the review and presents a brief discussion about the future investigation directions of flow battery modeling.

## 2. General description of physical and chemical processes in flow batteries

In a typical redox flow battery system shown in Fig. 1, the reactions occur at the two electrodes can be expressed as:

Positive electrode:

$$C^n - ye^- \underset{\text{Discharge}}{\overset{\text{Charge}}{\rightleftharpoons}} C^{n+y} \tag{1}$$

Negative electrode:

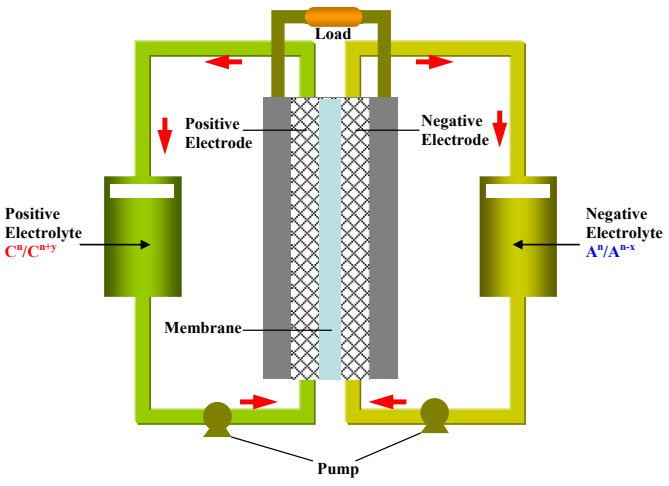


**Fig. 1.** Schematic of a flow battery system.