



Numerical study of the effects of carbon felt electrode compression in all-vanadium redox flow batteries



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ABSTRACT

The porous carbon felt electrode is one of the major components of all-vanadium redox flow batteries (VRFBs). These electrodes are necessarily compressed during stack assembly to prevent liquid electrolyte leakage and diminish the interfacial contact resistance among VRFB stack components. The porous structure and properties of carbon felt electrodes have a considerable influence on the electrochemical reactions, transport features, and cell performance. Thus, a numerical study was performed herein to investigate the effects of electrode compression on the charge and discharge behavior of VRFBs. A three-dimensional, transient VRFB model developed in a previous study was employed to simulate VRFBs under two degrees of electrode compression (10% vs. 20%). The effects of electrode compression were precisely evaluated by analysis of the solid/electrolyte potential profiles, transfer current density, and vanadium concentration distributions, as well as the overall charge and discharge performance. The model predictions highlight the beneficial impact of electrode compression; the electronic conductivity of the carbon felt electrode is the main parameter improved by electrode compression, leading to reduction in ohmic loss through the electrodes. In contrast, the kinetics of the redox reactions and transport of vanadium species are not significantly altered by the degree of electrode compression (10% to 20%).

This study enhances the understanding of electrode compression effects and demonstrates that the present VRFB model is a valuable tool for determining the optimal design and compression of carbon felt electrodes in VRFBs.

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1. Introduction

For more widespread use of renewable energy resources, developing a cost effective, large-scale energy storage system (ESS) is highly demanded. Among several ESS technologies, redox flow batteries (RFBs) have received considerable attention and are expected to be applied in various renewable energy markets in the future. Particularly, all-vanadium RFBs (VRFBs) using the same vanadium species but different oxidation states show superior characteristics in terms of cyclic durability as well as capital and operating costs [1]. Particularly, a deeper understanding of the complicated electrochemical reactions and transport phenomena involved in VRFBs must be key to successful design and operation of VRFB systems.

The transport characteristics and cell performance of VRFBs during charging and discharging processes are strongly influenced

by the porous structure of the electrodes. Carbon felt has been recognized as the favored porous-electrode material for VRFBs as it offers several advantages over other electrode materials, such as low cost and high permeability of the liquid electrolyte [1–6]. During stack assembly, the carbon felt electrodes are necessarily compressed to ensure that there is no liquid electrolyte leakage and to reduce the interfacial contact resistance between individual stack components. However, if the electrodes are too strongly compressed, the porosity and permeability of the electrode are significantly reduced, leading to limited electrolyte transport through the electrode, which is an adverse effect of electrode compression.

The influence of electrode compression on the overall VRFB performance and efficiency has been evaluated in a number of studies. Park et al. [7] empirically assessed the VRFB performance for various degrees of compression of the carbon felt electrode (0%, 10%, 20%, and 30%). Based on the experimental charge/discharge curves, they estimated the cell resistance as a function of the electrode compression level, demonstrating a considerable decline in the cell resistance as the percentage electrode compression

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Nomenclature

A	cross-sectional area, m^{-2}
a	effective catalyst area per unit electrode volume, m^{-1}
C	molar concentration, mol m^{-3}
C_{KC}	Kozeny–Carman constant
D	diffusion coefficient, $\text{m}^2 \text{s}^{-1}$
E_0	thermodynamic equilibrium potential, V
F	Faraday's constant, 96487 C mol^{-1}
h	heat transfer coefficient, W K^{-1}
I	current, A
j	transfer current density, A m^{-2}
K	effective permeability, m^2
k	reaction rate/coefficient, m s^{-1} , and thermal conductivity, $\text{W m}^{-1} \text{K}^{-1}$
M	molecular weight, kg mol^{-1}
\bar{N}	species flux, $\text{mol m}^{-3} \text{s}^{-1}$
P	pressure, Pa
Q	volumetric flow rate, $\text{m}^3 \text{s}^{-1}$
R	universal gas constant, $8.314 \text{ J mol}^{-1} \text{K}^{-1}$
r_p	pore radius, m
S	source/sink term
s	entropy, $\text{J K}^{-1} \text{mol}^{-1}$
T	temperature, K
\bar{u}	fluid velocity, m s^{-1}
V	volume, m^3
v	velocity, m s^{-1}
v_{f_i}	volume fraction of species i

Greek symbols

α	transfer coefficient
ε	porosity
η	overpotential, V
κ	ionic conductivity, S cm^{-1}
μ	dynamic viscosity, $\text{kg m}^{-1} \text{s}^{-1}$
ρ	density, kg m^{-3}
σ	electronic conductivity, S cm^{-1}
τ	viscous shear stress, N m^{-2}
Φ	potential, V
δ	electrode thickness

Subscript

0	equilibrium or initial value
amb	ambient
cc	current collector domain
ch	charged species
dis	discharged species
e	electrolyte domain
elec	electrode
i	species index
in	channel inlet
mem	membrane domain
NE	negative electrode
out	channel outlet
PE	positive electrode
s	solid electrode domain

Superscript

0	equilibrium or initial value
e	bulk electrolyte
eff	effective value
s	electrode surface

increased up to 10%, after which the rate of decline became notably gradual for compression in the range of 10–30%. Using a two-dimensional (2-D), isothermal VRFB model, Shah et al. [8] numerically investigated the effect of the electrode porosity on the charge and discharge behavior of VRFBs. Their simulation results demonstrated that the use of a denser electrode (lower porosity) leads to a lower bulk reaction rate in the electrode and thus results in a higher coulombic efficiency and longer charging time to reach an equivalent state of charge (SOC). However, that study did not discuss why the charging time was affected by the magnitude of the electrode porosity even at fixed charging current (galvanostatic mode). You et al. [9] conducted 2-D isothermal VRFB simulations, from which it appeared that higher transfer current density and more severe vanadium transport limitation were operative at higher degrees of electrode compression. However, the effect of electrode compression on the overall charging and discharging performance was not discussed in their numerical analysis. Qiu et al. [10] developed a 3-D isothermal pore-scale model for precise examination of the key transport mechanisms through a realistic VRFB electrode structure in which porous carbon felt geometries were constructed by using X-ray computed tomography, and the lattice Boltzmann method was adopted to predict electrolyte flow through the complex electrode geometries. They applied the model to a positive half-cell and found that lower overpotential and resultant higher half-cell potential were achieved with denser electrodes (lower porosity) due to the relatively larger active surface area for redox reactions. In their simulations, however, the electrode thickness was considered constant regardless of changes in the porosity, which is not the actual case during electrode compression.

The desired porous structure and properties of carbon felt electrodes can be achieved by optimizing the assembly clamping pressure. Chang et al. [11] experimentally showed that as the clamping pressure increases, the thickness and porosity of the carbon felt electrode (F1-75P4) decrease, whereas its electronic conductivity is improved, which would significantly affect electrolyte and electron transport through the electrodes and the overall cell performance. However, as mentioned above, the effects of electrode compression on the electrode transport properties have not been accurately taken into account in the majority of reported VRFB modeling studies. In this report, we present a numerical evaluation of the effects of the porous properties of carbon felt electrodes on the operation of VRFBs using a three-dimensional (3-D), transient VRFB model developed in a previous study [12]. Based on the empirical data documented by Chang et al. [11], we precisely account for the relation between the electronic conductivity, porosity, and thickness of the electrode as a function of the electrode compression level (see Table 1). The model employing the modified electrode correlations is then applied to a simple VRFB geometry (shown in Fig. 1), and charging and discharging simulations are carried out under different levels of electrode compression. The findings of the present study should contribute to enhancing the fundamental understanding of key complex and interacting phenomena due to electrode compression and identifying the optimal design and compression of carbon felt electrodes in VRFBs.

2. Numerical Model

A 3-D, transient VRFB model used for this study was successfully validated against the experimental data [12]. The detailed description of the 3-D transient VRFB model was already provided in our previous papers [12]; therefore, only a brief summary of the model equations and assumptions have been repeated herein. The governing equations and relevant source terms are summarized in

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