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# A comprehensive study of electrode compression effects in all vanadium redox flow batteries including locally resolved measurements

Purna C. Ghimire[a,b,1], Arjun Bhattarai[b,c,1], Rüdiger Schweiss[d], Günther G. Scherer[e], Nyunt Wai[b], Qingyu Yan[f,*]

[a] Interdisciplinary Graduate School, Nanyang Technological University, Singapore
[b] Energy Research Institute @Nanyang Technological University, Singapore
[c] VFLOWTECH Pte Ltd, Singapore
[d] SGL Carbon GmbH, Meitingen, Germany
[e] 5607 Hägglingen, Switzerland
[f] School of Material Science and Engineering, Nanyang Technological University, Singapore

## HIGHLIGHTS

- VRFB performance was studied by varying electrode compressions.
- Segmented cell study was performed to monitor the flow distribution.
- Best performance was achieved at electrode compression of 25%.
- A relationship between SOC conversion and flow distribution was established.

## ARTICLE INFO

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## ABSTRACT

Graphite felts are the most commonly used electrode materials in vanadium redox flow batteries. In the conventional cell design, flat sheets of graphite bipolar plates and porous graphite felts are stacked without any bonding, which requires a certain degree of compression to minimize the contact resistance. Excessive compression of the electrode, however, leads to non-uniform flow distribution and potential occurrence of zones with the retarded flow of electrolyte. This study investigates a wide range of electrode compressions and their effect on the cell performance. The results show that a compression of 25% is the optimal trade-off between contact resistance, homogeneity of flow distribution and pumping losses. Moreover, spatially resolved measurements using a segmented cell are employed to visualize the flow distribution across the electrode in real time. The open circuit voltage after the termination of the cell charge/discharge is converted to the corresponding state of charge (SOC) of the electrolyte, and the difference between the theoretical and experimental state of charge of electrolyte is used to quantify the flow distribution across the electrode. The results show that the optimum conversion of the reactant can be achieved during a single pass at 25% electrode compression. This method of segmentation is simple and scalable to any size of the battery.

## 1. Introduction

The vanadium redox flow battery (VRFB) has emerged as one of the most promising technologies for large-scale electricity storage. It offers unique characteristics, such as the ability to independently configure energy and power, absence of cross-contamination due to the use of the same metal ions in both electrolyte tanks, ambient temperature operation and long cycle life [1,2]. The inevitable crossover of active species in flow batteries is not critical in VRFB since the use of species from a single element allows for easy rebalancing of the electrolyte.

Graphite felts electrodes are used in VRFB because of their three-dimensional structure, stability in acidic condition, high porosity (> 90%), high conductivity and large surface area. Moreover, these materials show high elasticity upon compression, meaning that they do

* Corresponding author.
*E-mail address:* alexyan@ntu.edu.sg (Q. Yan).
[1] These authors contributed equally to this work.

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not require bonding to the current collector. Numerous studies have been conducted to improve the performance of VRFB by enhancing electrode kinetics, reducing the pressure drop, increasing the thermal stability of electrolytes and decreasing the cell resistance to operate at higher current density. Examples include thermal surface oxidation [3] and chemical etching [4], both of which increase the active surface area and the number of oxygen-containing functional groups. An alternative method to increase the charge transfer kinetics is to deposit catalysts onto the surface of the electrode. Noble metals, such as, Pt, Ir, Mn, Pd and Au impregnated on carbon fibre were studied, with only Ir-coated electrode exhibiting acceptable electrochemical behaviour. By contrast, hydrogen evolution was prominent for the electrodes decorated with other noble metals [5]. W. H. Wang et al. further studied the Ir-modified electrode and reported that its use in a positive electrode decreases the cell resistance by 25% compared to non-modified electrodes [6]. Electrodes containing metal oxides ($Mn_3O_4$ and $TiO_2$, e.g.) were reported to be promising candidates due to their low cost [7,8]. For instance, niobium oxide was proven to have a high catalytic activity towards both redox couples [9]. Suarez et al. reported the use of bismuth nanoparticle decorated graphite felt as a negative electrode and observed efficient inhibition on the irreversible reaction of hydrogen formation [10]. Similarly, perforation or integration of channels into the electrodes to reduce the pressure drop [11,12], additives to the electrolyte for increased thermal stability [13], and design changes aimed at lowering the cell resistance have also been attempted. Xu et al. [14] performed numerical simulations of VRFBs for various flow field configurations. Their results showed that the presence of flow fields ensures an even distribution of the electrolyte over the electrode surface and facilitates the transport of electrolyte toward the membrane. Mohamed et al. [15] used a standardized test-bed system to evaluate the performance at different operating parameters and recorded the highest energy efficiency of 82% at current density of 60 mA $cm^{-2}$.

In the conventional VRFB design, the cell frames accommodate the bipolar plates (BPs) and provide a cavity for the felt electrodes. Compression of the felt electrode improves the electronic contact to the BP at the expense of a reduction in electrode porosity. Excessive compression, however, has several negative consequences. In particular, it leads to a higher-pressure drop. This is accompanied by higher pumping losses and non-uniform flow distribution, which could ultimately lead to a reduction in electrode utilization and formation of dead zones (i.e. areas with less supply of fresh electrolyte). These effects are most critical during charging, as they could trigger elevated gas evolution and corrosive degradation of the BPs [16]. Therefore, optimization of electrode compression is a necessary task for obtaining optimal performance and longevity of flow battery stacks.

The pressure drop is another important parameter that must be considered during the selection of the appropriate electrode compression. Energy consumed by pumps used in circulating the electrolyte varies linearly with the pressure drop. The pressure drop (ΔP) occurs between the flow inlet and outlet, due to the viscous resistance to flow, which is dependent on the viscosity of electrolyte and permeability of the porous electrode. For laminar flow, the pressure drop is given by Darcy's law

$$\Delta p = \frac{\eta_e}{K_e(\varepsilon)} \cdot Q \cdot \frac{L}{\varepsilon A} \quad (1)$$

where $\eta_e$, $Q$, $L$, $A$, $K_e$, represent the viscosity of the electrolyte, the flow rate, the flow path length, the cross-sectional area of the felt and the permeability of the electrode (which is dependent on the porosity $\varepsilon$), respectively. During the operation of a VRFB, the pump consumes 1–3% of the total energy [2,11], mostly due to the pressure drop across the porous electrodes. To reduce ΔP, several approaches, such as flow channels in the porous electrodes or in the BPs, have been proposed [11,17].

To date, only a handful of studies have been performed with the aim of understanding the influence of electrode compression on the overall performance of a VRFB. Park et al. [18] observed a sharp decrease in specific resistance up to 10% of electrode compression and a flattening decline of resistance for higher levels of compression. Chang et al. [19] studied the change in electrical, morphological and mechanical properties up to a felt compression of 40%, using BPs containing flow channels. They showed that the increase in clamping force leads to a decrease in porosity underneath the rib and channel, which is attributed to the deformation of the carbon felt accompanied by a loss in void volume. A high compression pressure of 7.5 bar and a relatively thin membrane (Nafion NR-212) has been used to record the area specific resistance of 0.374 Ohm $cm^2$, lowest value recorded till date [20]. More recently, Wang et al. [21] showed an improvement in energy efficiency of 19.4% when the compression ratio was varied from 0.3% to 41.8%. In addition to those experimental studies, several numerical approaches are available. For instance, Brown et al. [22] modelled the pressure drop in 3D microstructures using computational fluid dynamics. Likewise, the effect of electrode porosity on the charge/discharge behaviour [23,24] and the pressure drop through the felt have also been simulated [25].

Overall, the previous studies on electrode compression have focused on the electronic resistance and the efficiency. However, less attention is paid to the effect of compression on flow distribution and electrolyte utilization. The discharge energy, which is lacking in most of the published reports, is a more meaningful figure of merit for the characterization of the effects brought by electrode compression. Moreover, most of the experimental studies have been performed using either a fuel cell design (with thin electrodes and BP with flow fields) or a cell with a small active area. The effect of non-uniformity and change in contact resistance is more prominent in cells with larger active area. Similarly, visualization of flow distribution and electrolyte utilization along the cross section of the electrode at different levels of electrode compression is also lacking in the literature.

Hence, this study investigates the effect of carbon felt compression on electronic conductivity, pressure drop in both half-cells and overall cell performance during charging and discharging in a 100 $cm^2$ VRFB cell. In addition, a simply designed conventional segmented cell, which is scalable to any size, is presented. For the first time, to minimize the lateral flow of current, local OCV/SOC mapping is performed to study the effect of the electrode compression on the electrolyte utilization at different electrode compression levels. A relationship between the experimental and theoretical ΔSOC is established to acquire the information about electrolyte utilization across the electrode.

## 2. Experimental

An in-house developed flow cell with an active area of 100 $cm^2$ (10 cm × 10 cm) was employed. The arrangement of the different components is provided in supplementary information (Fig. S1). The PVC flow frames were equipped with flow guides to facilitate the uniform flow of electrolyte through the porous electrode. Stainless steel plates (10 mm thick) were used as end plates to press the components into contact. Copper plates were selected as the current collectors. Battery-grade polyacrylonitrile (PAN)-based carbon felts (Sigracell® GFD, 6 mm thick, SGL Carbon GmbH, Meitingen, Germany) were employed as the electrodes. Felts were thermally activated in air at 600 °C for 5 h. The cell was assembled with Sigracell® TF6 graphite BPs (SGL Technic LLV, Valencia, CA, USA) and an anion exchange membrane (FAP 450, Fumatech GmbH, Bietigheim-Bissingen, Germany).

Six different compression levels of the porous electrode (15%, 20%, 25%, 30% 35% and 40%) were investigated. The electrode compression was varied by using combinations of silicone and Teflon™ gaskets of varying thickness (0.3, 0.5, 1 and 2 mm). A torque wrench (9 N·m) was employed to ensure uniform compression of the electrode. After each assembly, the thickness between the two end plates was measured to ensure the exact compression of the electrode. The volume of the electrolyte in each tank was 150 mL and the concentration of the

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