



# High performance zinc-bromine redox flow batteries: Role of various carbon felts and cell configurations

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## ABSTRACT

Optimization of the cell configuration utilizing various carbon felts for obtaining better performance in zinc-bromine redox flow battery (ZBRFB) system is reported. It is clearly observed that the reaction kinetics of both Zn and Br are enhanced when used in different cell configurations with carbon felts. Redox flow cells having carbon felts on both half-cells showed improved performance than the other cell configurations tested in the present study. Further, Rayon based carbon felt is evaluated as an electrode on both sides. It is noted that the Rayon carbon felt based cell exhibited good performance in terms of coulombic (96.26%), voltaic (83%) and energy efficiency (79.4%) at a current density of  $20 \text{ mA.cm}^{-2}$ . In addition to that, Rayon felt based system showed good performance even at  $40 \text{ mA.cm}^{-2}$  ( $3250 \text{ mW h. l}^{-1}$ ) which indicates the significance of the flow cell configuration on the overall cell performance.

## 1. Introduction

In recent years, demand for electricity usage has rapidly increased worldwide. To fulfill the requirements, electrical energy is retrieved from various sources like hydro, nuclear, fossil fuels, and renewable energy resources (solar and wind). Among these, renewable energy-related resources are obviously dominant because of eco-friendliness, abundant availability, low-cost and importantly zero emission of  $\text{CO}_2$  which has a significant impact on global warming. As the renewable energy resources are intermittent; an integrated grid level energy storage system is important to fulfill the peak power requirements. Redox flow batteries are considered as the most promising candidate for such storage applications, particularly in the large-scale due to their advantages such as the independent relation of power and energy, long cycle life, low capital and maintenance cost [1,2]. So far, a number of RFBs have been studied including Fe/Cr [3], Fe/V [4] all-vanadium [5–10], Zn/ $\text{Br}_2$  (ZBB) [11–17], polysulfide/ $\text{Br}_2$  [18] and Iron-Chloride [19]. Among these, the all-vanadium redox flow battery (VRFB) has been installed at many places worldwide by different manufacturers namely, Prudent energy ([www.pdenergy.com](http://www.pdenergy.com)), Glidemeister (<http://energy.glidemeister.com/en/>), American Vanadium (<http://www.americanvanadium.com>), Cellinium ([www.vanadiumbattery.com](http://www.vanadiumbattery.com)) etc. The major advantage of VRFB system is due to the presence of the same element in both the electrolytes, which avoids the cross-contamination

of electrolytes between the half-cells. However, low energy density ( $20\text{--}30 \text{ W h. kg}^{-1}$ ), use of acidic electrolyte, high-cost of ion exchange membrane and vanadium in global markets are the major worrying factors which restrict their widespread deployment [20].

ZBRFB is an alternate choice because of the added advantages such as low - cost, high cell voltage, high theoretical specific energy ( $429 \text{ Wh. kg}^{-1}$ ) [21], which in practice is  $60\text{--}70 \text{ W h. kg}^{-1}$  [22] with the use of the normal porous separator. However, the development of Zn- $\text{Br}_2$  is slow compared to VRFB due to the issues related to such as zinc dendrites formation and self-discharge due to bromine diffusion during charging process. The former one may cause a short circuit which in turn damages the cell while the latter, namely, bromine transport and bromine vapour formation ultimately imbalance the reaction kinetics in the positive side. This can be minimized by using bromine complexing agent and has been demonstrated successfully [23]. However, the addition of complex agents may not necessarily help the long-term stability; hence, the maximum cycle life of the Zn- $\text{Br}_2$  system is limited to less than 2000 cycles. The improvement in the performance of the flow system is also highly dependent on the cell configurations. Based on the electrode material, active area and the cell design, the reaction kinetics of the flow system is varied. Nonetheless, no reports are available so far dealing with the cell configuration aspect. Hence, in the present work, it is proposed to evaluate the performance of the flow system at different cell configurations with different carbon felts. Zn- $\text{Br}_2$  redox flow cells

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have a similar architecture as that of a conventional flow cell. The positive and negative electrolytes are stored in the separate tank and the electrolyte flow through the half-cells with the help of circulating pumps. Since the redox reaction occurs on an active electrode surface area, the cell components, particularly, active area should be designed according to the redox reactions. In the present investigation, different types of carbon felt with different cell configurations are evaluated at various current densities (from 4–40 mA.cm<sup>-2</sup>) for obtaining better performance of the hybrid flow system.

## 2. Experimental section

### 2.1. Materials

Graphite composite plates (6 mm thickness) were used as supporting electrodes in the present investigation. Polyacrylonitrile (PAN) and Rayon based carbon felts (3 mm thickness) were purchased and used as active electrode materials. ZnBr<sub>2</sub> and ZnCl<sub>2</sub>, were purchased from Alfa Aesar, UK and used as received for the electrolyte preparation. *N*-Methyl-*N*-Ethyl Morpholinium bromide (MEM) and *N*-Methyl-*N*-Ethyl Pyrrolidinium bromide (MEP) were procured from Sigma Aldrich, USA. A microporous polyethylene membrane (Daramic) with the thickness of 1 mm was used as a separator. Peristaltic pumps were used for electrolyte circulation through the half-cell.

### 2.2. Cell assembly and electrochemical characterization

As mentioned earlier, the cell was assembled at different configurations in the following manner: i) without carbon felt on both sides; ii) carbon felt on bromine side(positive half cell only); iii) carbon felt on both positive and negative side in which PAN and Rayon based as purchased carbon felts were studied, independently. Photograph of the flow cell setup and its exploited view of the single flow cell used in this study are shown in Fig. 1a and Fig. 1b, respectively. The electrolyte mixture of 3 M ZnBr<sub>2</sub> + 1 M ZnCl<sub>2</sub> + 1:1 MEP:MEM in DI water was prepared and used for the flow cell testing. The given electrode materials (Graphitecomposite plate or Carbon felt) Platinum (Pt) and Ag/AgBr were used as working, counter and reference electrodes, respectively in three electrode system. Cyclic Voltammogram (CV) of the half cell was recorded in the potential range from -1.2 V to 1.5 V for each electrode, separately. Autolab Model PGSTAT30 was used to record both the CV and impedance of the electrode materials. Conductivity of the electrolyte medium was measured directly using Proflin WTW-Multi3320 portable conductivity measuring unit.

The volume of electrolyte used for testing in the flow cell was about 80 ml each side and the electrolyte flow were maintained at 20 ml.min<sup>-1</sup> on both sides throughout the experiment. The flow cell was tested at different current rates typically in the range of 4, 8, 12, 16, 20, 30 and 40 mA.cm<sup>-2</sup>. Galvanostatic charge-discharge (GCD) performance was carried out using ARBIN battery tester. The charge-discharge studies were carried out for 30 min. each for the given flow cell system. Finally, the volumetric capacity and energy density were calculated based on the volume of the anolyte. The results obtained from different conditions of the flow cells are discussed here in detail.

## 3. Results and discussion

### 3.1. Cyclic voltammetric studies

With an aim to understand the electrocatalytic effect, CV curves (cyclic voltammograms) were recorded for graphite composite plate (supporting electrode), PAN and Rayon based carbon felt electrodes in an electrolyte composed of 3 M ZnBr<sub>2</sub> + 1 M ZnCl<sub>2</sub> + MEM:MEP (1:1 mol ratio) solution at a scan rate of 20 mV.s<sup>-1</sup>. The obtained CV curves of the samples are shown in (Fig. 2a). It is well documented that Zn<sup>2+</sup> ion present in the electrolyte gets reduced to Zn in the negative

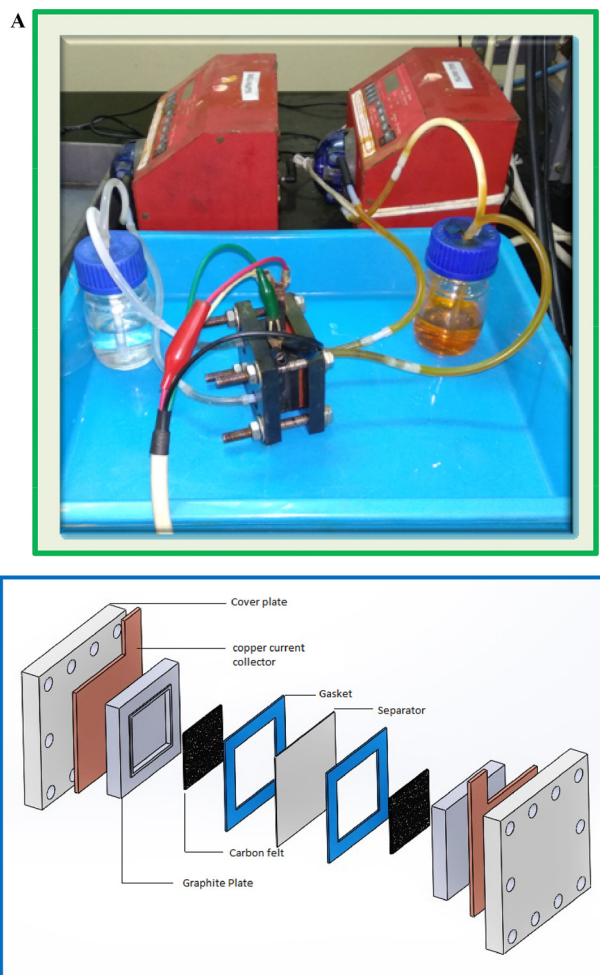


Fig. 1. a) Photograph of the flow cell setup used in the present study. b) Exploited view of the single cell setup used in the present study.

sweep potential [24]; the onset potentials at -0.89 V, -0.86 V and -0.85 V related to the Zn deposition were obtained corresponding to the graphite composite plate, PAN and Rayon felt, respectively. On the other hand, the anodic peak current densities of graphite composite plate, PAN and Rayon felt are found to be 75, 47 and 135 mA.cm<sup>-2</sup>, respectively. The obtained values can be increased further by activating the electrode materials [25]. This implies that the reduction of zinc nuclei highly depends on the surface properties, particularly, roughness factor of the electrode; thus, the Rayon based carbon felt electrode showed better activity than the other studied electrode materials. During the anodic sweep, peaks observed at 0.92, 0.90 and 0.88 V corresponding to the oxidation of bromide ions (Br<sup>-</sup>) to bromine (Br<sub>2</sub>) on graphite composite bipolar plate, PAN and Rayon felt, respectively. On reversal of the potential scan at 20 mV.s<sup>-1</sup>, Br<sub>2</sub> was reduced back to Br<sup>-</sup> ions, giving rise to a cathodic peak in the potential range 0.35–0.90 V vs. Ag/AgBr. The cathodic peak current density of Rayon felt, PAN, and graphite composite plates are found to be 119, 80 and 87 mA.cm<sup>-2</sup>, respectively. The enhanced peak current density of Rayon felt is mainly attributed to the larger number of active sites and good electrical properties. The electrochemical active sites for all the samples were calculated from the cathodic reaction. Here, it is considered only the active area under the bromine reduction peak for calculating the active sites of the electrode materials. The selective area which was considered for the active site calculation is given in the Fig. S1. It was noted that the Rayon carbon felt showed higher magnitude in the electrochemical active sites (0.038VA) when compared with the other samples (0.022 VA-PAN; 0.023 VA-graphite). The lower peak current

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