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*Short Communication*

# A Novel Bipolar Plate Design for Vanadium Redox Flow Battery Application

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A novel design of bipolar plate (BP) was proposed for vanadium redox flow battery (VFB). The BP was prepared by injecting molten polyethylene into micropores of carbon fibers (CF) *via* molding method (simplified as MBP), which behaved high conductivity and great mechanical strength due to its special morphologies of conductive network structure uniform distribution made the fibers connecting with each other compared to the BP prepared by extrusion method (simplified as EBP). The electrochemical tests also showed the VFB with the composite MBP behaved better battery performance with higher voltage efficiency which could be attributed to the lower resistance of MBP consisting of conductive CF networks.

**Keywords:** Carbon materials; Electrical properties; Energy storage and conversion;

## 1. INTRODUCTION

Vanadium redox flow batteries (VFBs) are electrochemical devices that store energy utilizing active liquid electrolyte in the external separate tanks and pump through the battery stack during operation[1]. Unlike conventional batteries, VFB is more suitable for large scale energy storage due to its long cycle life, easy scale-up and 100% discharge capability[2-6], which make it widely used in load shifting, large-scale emergency power systems. However, VFB is still confronted with great challenge of higher prices, lower energy efficiency in the scale-up course[7-8].

The electrode as an electrochemical reaction place, which is generally carbon-based materials[9-13], another core component is bipolar plate (BP) as a current collector, and high resistance of BP will lead to lower voltage efficiency[14-15]. In general, BP can be categorized into three types, such as metallic, graphitic, and composite conductive plastic material. The metallic BP has a critical disadvantage of highly corroded in an acid-based solution[16]. Similarly, the graphitic BP also has some drawbacks, such as lack of mechanical strength and low processability[17]. Compared with these two types, the composite conductive plastic BP is considered as a promising alternative in terms of good electrical conductivity, great mechanical strength and easy formability[18]. In addition, the composite conductive plastic BP is manufactured by molding process or extrusion process[19]. Compared with BP via extrusion process (EBP), the BP via molding process (MBP) showed lower electrical resistivity and better mechanical modulus/strength due to its special morphology.

In this study, MBP has been developed by integrating the carbon fiber of CF to the polyethylene (PE) matrix *via* molding method. To evaluate the performances of the MBP assembly, the mechanical strength and volume resistivity were measured. Finally, the electrochemical performance of the MBP assembly were measured and compared with those of EBP.

## 2. EXPERIMENTAL

### *2.1. Preparation of MBP*

Fig.1 shows the process of MBP. Firstly, in the experiment PE (with a thickness of 1 mm, Sinopec Shell, HDPE, 5121B) plate was prepared by molding method, then placed the PE plate on the carbon felt (with a thickness of 2 mm, Runsheng Graphite Felt Co. Ltd.) together with the die into a flat vulcanizing machine, the process was as follows: 1) preheating process, the die for closed but no pressure, heated to 240 °C for 90s; 2) pressure and temperature holding process, temperature at 240 °C, pressure for 80 kgcm$^{-2}$ for 3 min; 3) Cooling process: water cooling for 2min. Secondly, in order to prepare MBP, the prepared carbon felt and PE sheet were put on the mold into the flat vulcanizing machine. The process was as follows: 1) preheating process, the die for closed but no pressure, heated to 260 °C for 120s; 2) pressure and temperature holding process, temperature at 260 °C, pressure for 20 kg/cm$^2$ pressure for 3min; 3) Cooling process: water cooling for 2min.

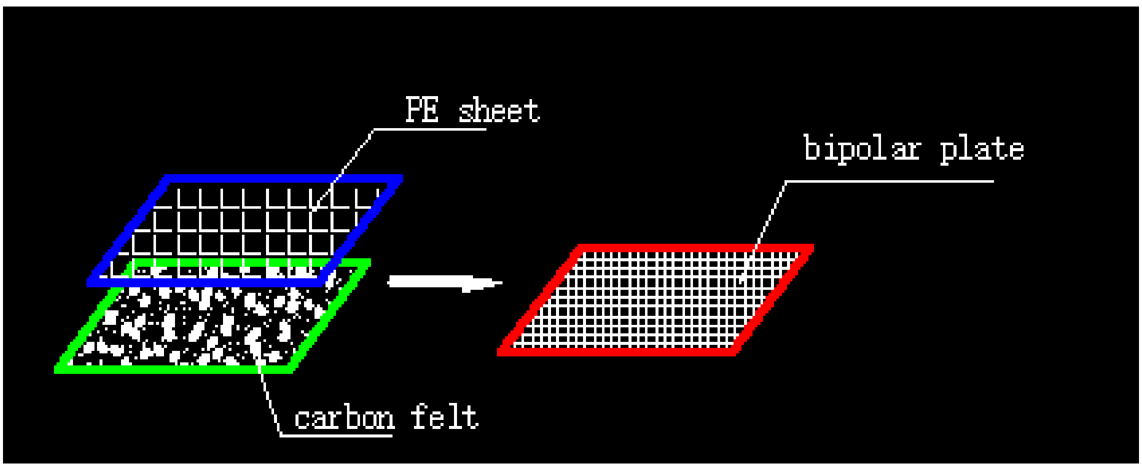


**Figure 1.** Molding process of the conductive plastic MBP

### 2.2. Physical Characterization

The sample was characterized by field-emission scanning electron microscopy (FE-SEM, Hitachi S-4800A). The mechanical strength was measured by electronic universal material testing machine (Instron5569). The volume resistivity was tested by four-electrode volume resistance meter (Suzhou Jingge Electronic Co., LTD).

### 2.3. Battery Assembly and Electrochemical Measurement

Fig.2 showed the structure of the battery, the components were end plate, the plate for import and export, bipolar plate, liquid flow frame, the electrode, ion-exchange membrane in sequence. Moreover, the MBP and EBP were prepared for the BP of the battery, respectively. The electrode was CF (SGL, German), the membrane was Nafion 212 (Dupont, USA). The charge-discharge test was performed by a flow battery on BPChecker2000.V3 System (Kikusui Electronics Corp). The peristaltic pump (Baoding Longer Precision Pump Co., Ltd.) used was BT100-1L pump. The battery was charged to 1.7 V and discharged to 1 V at the constant current density of 80 mAcm$^{-2}$, 120 mAcm$^{-2}$, 160 mAcm$^{-2}$, respectively.

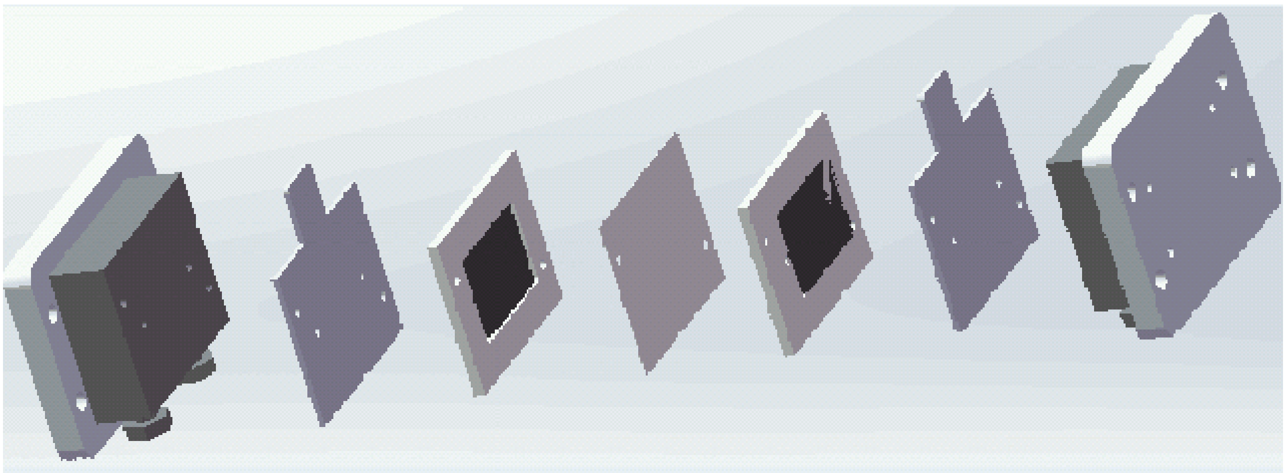

**Figure 2.** Structure of the battery

## 3. RESULTS AND DISCUSSION

### 3.1. Micro Structural Morphology

Fig. 3 showed the SEM images of MBP. Fig.3a showed a plurality of CF dispersed in the PE matrix, and the square shape was the fiber, which was the end of CF, the SEM showed the distribution of CF end was a little not uniform, this can be explained by the shift of part CF under the hot pressing condition. From Fig.3b a magnification of 20,000 times of a single fiber end, it can be clearly observed that CF and PE substrate were integrated tightly, and the connection was smooth, indicating both materials had good compatibility. In order to observe the binding extent of CF with PE matrix, the cross section images were characterized. From Fig.3c we could observe the fracture of CF felt was neat in the middle area, also there was gap between the end part of CF and PE matrix in marginal area, otherwise CF was uniformly distributed in PE matrix. Fig. 3d was the pore between the fiber end and the matrix in the marginal area, which may be partly because the microporous of CF was not sealed completely in the flow pressing process of melt PE.

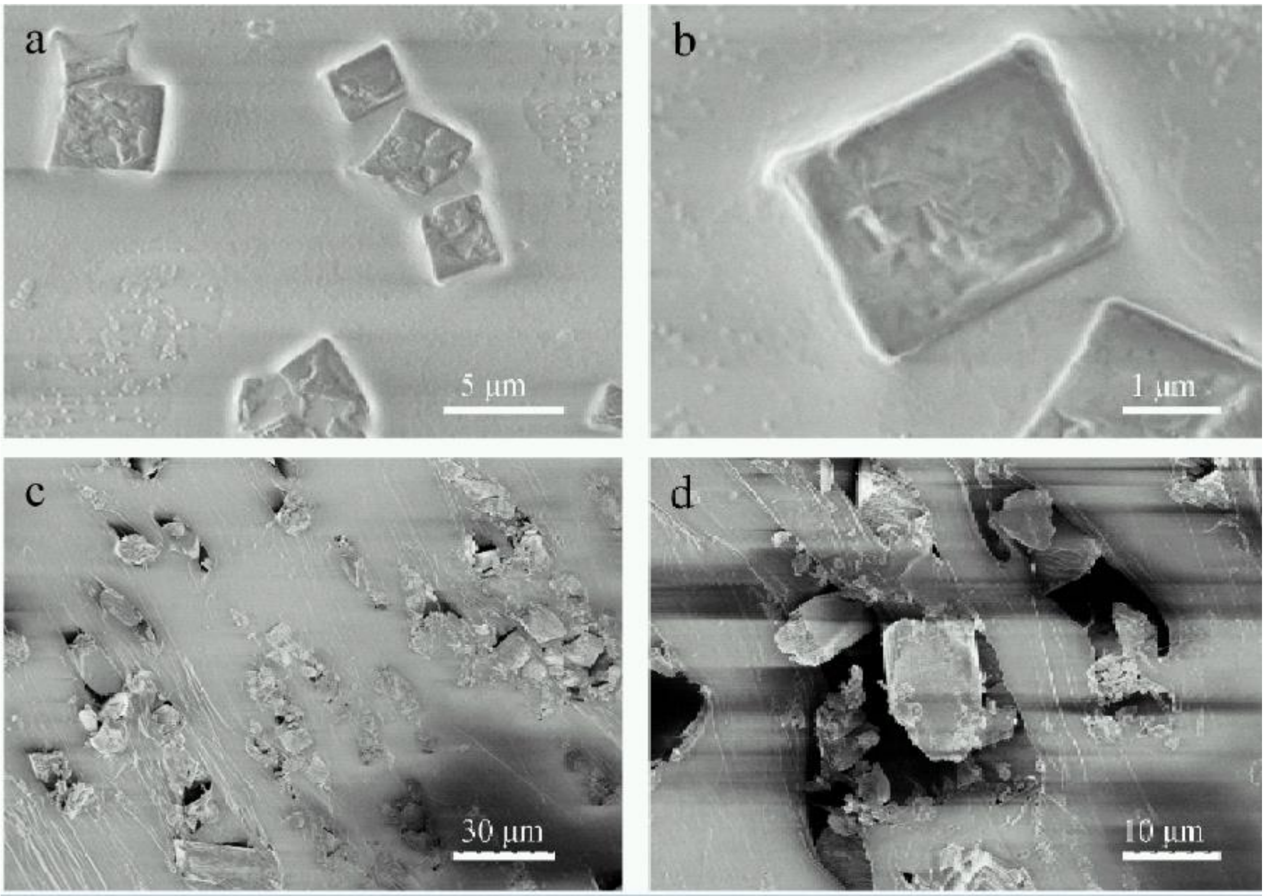


**Figure 3.** SEM images of MBP (a) & (b) the surface, (c) & (d) the fractography

### *3.2. Mechanical Properties*

The tensile test and analysis was used to evaluate the mechanical properties of MBP, and compared with EBP acquired from Suzhou Deskai Plastics Technology CO., LTD. Table.1 showed the elongation and tensile strength of both MBP and EBP samples, from which we could observe the elongation was almost the same, but the tensile strength of MBP was significantly better than EBP. The average tensile strength of MBP was 18.56 Mpa, about 1.8 times as large as EBP samples, also the data dispersion of MBP is smaller than EBP, indicating that the strength and the uniformity extent of MBP is better than EBP, this was due to the reason that the melt PE sealed into the microporous structure of CF which reinforced PE base material.

**Table 1.** The parameters of mechanical properties

| Specimen | Thickness (mm) | Width (mm) | Elongation at break（%）（ASTM D638） | Tensile strength（Mpa）（ASTM D638） |
|---|---|---|---|---|
| MBP-1 | 1.07 | 6.21 | 15.8 | 17.51 |
| MBP-2 | 1.10 | 6.20 | 16.1 | 19.84 |
| MBP-3 | 1.02 | 6.17 | 16.2 | 18.35 |
| EBP-1 | 1.17 | 6.41 | 15.1 | 9.92 |
| EBP-2 | 1.12 | 6.35 | 15.7 | 12.15 |
| EBP-3 | 1.15 | 6.37 | 16.3 | 8.39 |

### *3.3. Conductive Properties*

Resistivity is used to indicate resistance of various material. We prepared the samples in dimension of 5mm x 11mm according to the four-electrode volume resistance meter. Both MBP and EBP were tested for three samples, and the average value of three samples was taken as test parameters, which showed the resistivities of MBP and EBP were 3.13 mΩ·cm, 48.9 mΩ·cm, respectively, indicating MBP behaving better electrical conductivity than EBP. The reason was related to the morphologies between compression molded MBP and injection molded EBP samples. As shown in Fig. 4(a), the conductive carbon fiber were randomly oriented in three-dimensional direction for carbon fibers were remained in the PE substrate, thus making the connection between conductive carbon fiber maintain continuity. However, in Fig. 4(b), most of the particles were aligned , but not connecting with each other in the through-plane direction, preventing the formation of a continuous conductive filler phase and resulting in higher resistivity[20-21].

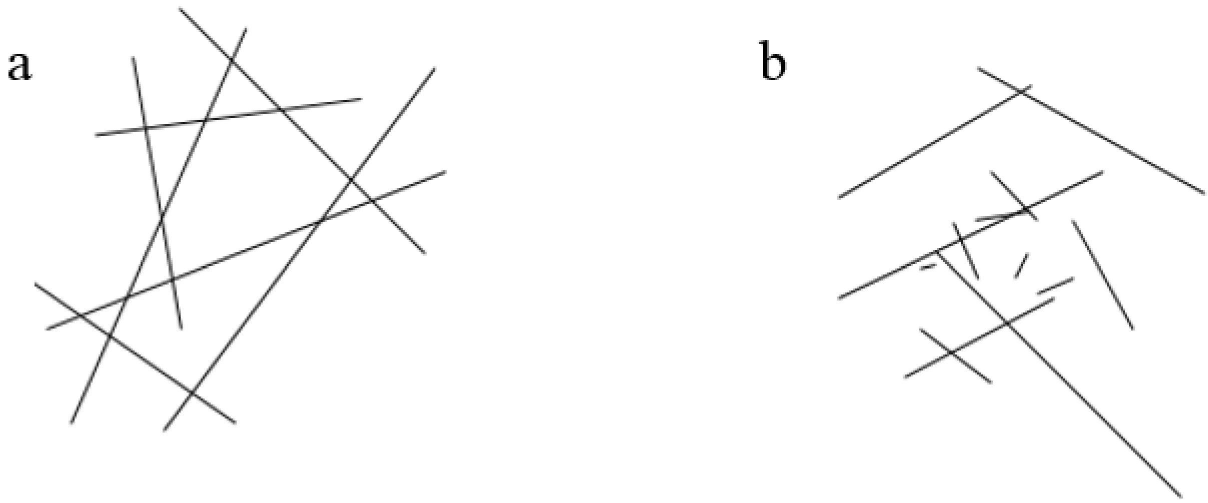


**Figure 4.** Schematics of alignment situation of conductive filler in PE matrix: (a) random orientation after compression molding, (b) aligned orientation after injection molding.

### *3.4. Electrochemical Performance*

In order to investigate the electrochemical performance of MBP, cyclic charge/discharge tests were performed using the above mentioned cell of VFB. Fig.5(a) showed the average values of coulomb efficiency(CE), voltage efficiency(VE) and energy efficiency(EE) averaged from the data of 18 cycles of charge/discharge with respect to the MBP and EBP as the bipolar sheets, respectively. It can be seen from Fig.5 that the EE, CE and VE were decreased with the increasing of current density ,which could be related to the ohmic polarization and electrochemistry polarization aggravated with the increasing of current density [22].

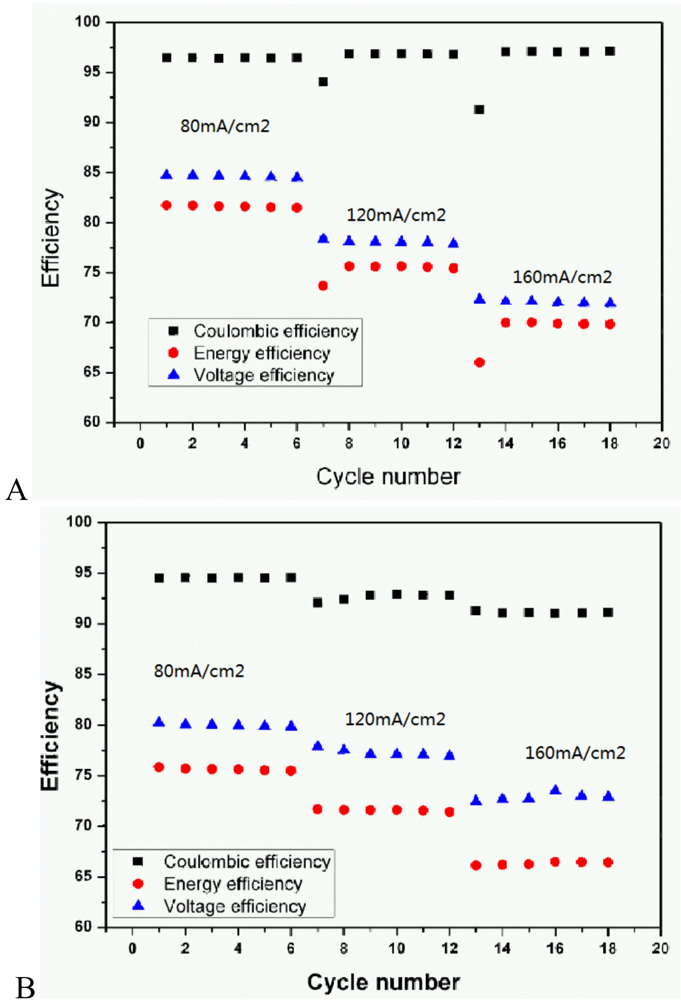


**Figure 5.** Electrochemical performance of VFB in single battery: (a) cycling performance of employing MBP electrode at 80 mAcm$^{-2}$ /120 mAcm$^{-2}$/160 mAcm$^{-2}$/(b) cycling performance of employing EBP electrode at 80 mAcm$^{-2}$/120 mAcm$^{-2}$ /160 mAcm$^{-2}$

The average VE, CE and EE of MBP at the current density of 80 mAcm$^{-2}$ were higher than those of MBP, and the VE, CE and EE of MBP of were 84.61%, 96.45% and 81.61%, respectively, while the average VE, CE and EE of EBP were 79.99%, 94.54% and 79.99%, respectively. And compared with the performance of Skyllas-Kazacos.M[23] reported, the electrodes prepared by graphite felt bonded onto carbon plastic sheets as current collectors, and the study demonstrated the coulombic efficiency of 90%, voltage efficiency of 81%, and an overall energy efficiency of 73% in a vanadium redox cell. Thus, it was suggested that MBP behaved better electrochemical performance, even at a current density of $80mAcm^{-2}$, a voltage efficiency of 85% and an energy efficiency of 82% were still obtained, The reason[24-25] could be due to the deterioration of conductive filler in the electrode substrate material of EBP, making the conductivity of the electrode decrease and maybe leading to side reactions during charge and discharge. While MBP was molded by carbon fiber into PE as substrate material having conductive network structure uniform distribution, this made MBP perform higher conductivity and more durable than the conventional designs.

## 4. CONCLUSION

A novel conductive plastic composite bipolar (MBP) of molding polyethylene into carbon felt was prepared for VFB. The MBP showed significant improvements in mechanical properties and conductivity compared with that of the bipolar *via* blend extrusion method (EBP). The novel electrode MBP was evaluated in the vanadium redox battery and the results showed better electrochemical performance than conventional EBP for exhibiting higher voltage efficiency in single cell .Therefore, the MBP bipolar will be great potential for VFB application.

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