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Laser perforated porous electrodes in conjunction with interdigitated flow field for mass transfer enhancement in redox flow battery



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ARTICLE INFO ABSTRACT Keywords: To further enhance the efficiency of redox flow batteries, there is a vital need to decouple the conflict between Redox flow battery specific surface area and hydraulic permeability in porous electrodes. In this work, we propose a facile and highly Porous electrode productive method to devise porous electrode's pore structure in conjunction with the flow field topology based Pore-network model on the infrared laser perforation to reconcile the combination of large and small pores. We find experimentally Flow field that the method of perforation under the center of the rib yields a significant improvement on the overall per-Laser perforation formance, with an increase in the maximum power density by approximately 2.4 times compared to that without the perforation. We also devise a pore-network model, by using which we explore the effects of large pores formed by the laser perforation on the distributions with respect to the local velocity, active species concentration and reaction rate, as well as the effects of the perforation location and diverse operating conditions on flow cell's overall performance. The results show that under the general operating conditions of the VRFB, we recommend to produce large pores in the region under the rib close to the outlet channel to enable substantial performance gains.

1. Introduction

In recent years, the proportion of renewable energy such as solar and wind in the global electricity generation has been increasing. The intermittent and unpredictable nature of renewable energy generation makes it impossible for the electricity generated to be directly integrated into the grid. At the same time, with the transformation of the energy system toward a low-carbon and intelligent one, the importance of energy storage is increasingly prominent. Among various types of energy storage technologies, redox flow batteries (RFBs) have the advantages of safety, flexible power and energy capacity configuration, and low lifetime cost, making them increasingly competitive in the field of large-scale energy storage [1,2]. Meanwhile, the heat of research on RFBs is rising, especially on the heat and mass transfer process in RFBs to further enhance the energy efficiency and reduce the cost (e.g., [3–5]).

The porous electrode is one of the core components of RFBs, which largely determines activation and concentration polarizations during the operation of RFBs [6]. Precisely because porous electrodes are closely related to the efficiency of RFBs, research on porous electrodes has been a top priority [7,8]. The ideal porous electrode should possess

the following characteristics: i) high electrochemical activity and reaction specific surface area, thus improving reaction kinetics to reduce activation losses; ii) excellent hydraulic permeability for efficient transport of active species, thus reducing concentration losses and pumping losses; and iii) high conductivity, thus minimizing ohmic losses [9]. The most commonly used materials for RFB electrodes are carbon-based materials (e.g., graphite felt, carbon felt, carbon paper etc.), which generally have high electrical conductivity, chemical and electrochemical inertness, nevertheless, poor electrochemical kinetics [10].

Numerous researchers have proposed thermal treatment, chemical treatment or modification with carbon nanomaterials for carbon-based porous electrodes in recent years. These studies have made some progress in increasing the electrochemical kinetics and enhancing the mass transfer of active species [11,12], enhancing performance of porous electrodes based on various mechanisms [13–15], but the conflict between the hydraulic permeability and the specific surface area has not yet been fully resolved [16]. The straightforward method for increasing the specific surface area is to reduce the diameter carbon fibers of the electrode, but at the expenses of the reduction in the hydraulic

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permeability, leading to an increase in the concentration polarization and pumping losses [17]. Alternatively, electrodes with aligned fibers for enhancing permeability [18] and novel flow fields for enhancing concentration distribution uniformity [19-21] have also been proposed. Some researchers have proposed dual-scale porous electrodes that combine two different scales of pores to maximize the reaction kinetics by increasing the specific surface area and active sites through small pores, while improving the hydraulic permeability through large pores to reduce the pressure drop when the electrolyte passes through the electrode, thus harmonizing the conflict between the hydraulic permeability and the specific surface area [22]. Forner-Cuenca et al. [23] found that the fibers in the carbon cloth exhibited a bimodal pore size distribution, including small pores (5 \sim 10 μ m) formed between fibers within carbon fiber bundles and large pores (50~100 µm) formed between carbon fiber bundles. Compared with carbon paper and carbon/graphite felt, the pore structure of carbon cloth effectively enhanced the mass transfer without affecting the hydraulic permeability, and the different weaving pattern of carbon cloth impacted the pore size distribution and thus the hydraulic permeability and mass transfer characteristics [24]. Jiang et al. [25] proposed a gradient porosity electrode, which avoided the loss of the electrochemical reaction area by the membrane side and increased the electrode utilization through the superposition of electrodes with different porosities. Jing et al. [26] prepared 3D electrochemically reduced graphene oxide porous gelatum material with a gradient porous microstructure on the graphite plate by using the electrodeposition method, while facilitating electrolyte, ions, and electron transport simultaneously. Zhou et al. [27] devised a dual-scale porous electrode by activating carbon paper fibers with KOH, which possessed large pores ($\sim 10 \,\mu m$) between carbon fibers as macroscopic channels for electrolyte flow and small pores (\sim 5 nm) on the surface of carbon fibers to increase the active sites for electrochemical reaction. In addition, Sun et al. [28] prepared sponge-like microfiber carbon electrode with dual-scale structure by electrospinning, achieving a balance between the specific surface area and the permeability. Wan et al. [29] and Jacquemond et al. [30] introduced the use of the non-solvent induced phase separation to prepare electrodes with tunable dual-scale microstructures, resulting in improved battery performance. However, the practicality of these novel electrode structures still deserves consideration, as they are typically designed based on a specific flow field structure (e.g., flow through or interdigitated), which results in limiting their scope to a single flow field geometry, and typically face processing difficulties and cost dilemmas.

The investigations on the pore structure of porous electrodes have been largely carried out by simulations, since the experimental preparation specific pore structures and the *in-situ* mapping of concentration at the pore scale are still challenging (note that the recently reported work by Wong et al. [31] successfully realized the *in-situ* mapping) [32]. For example, Chen et al. [33] simulated the effects of the gradient or bilayer porous electrode stacking by using COMSOL Multiphysics® and explored the effects of the parameters such as porosity, electrode thickness, and gradient of porosity change on battery's performance. Liu et al. [34] also developed a 3-D numerical model to evaluate the enhancement of battery performance by using different variable cross-section electrodes under the interdigitated flow field. Pore-scale models have also been developed to investigate the impacts of electrode's microstructure. For example, Sadeghi et al. [35,36] proposed a pore-network model that approximated the electrode pores and throats as spheres and cylinders, respectively, and used the model to investigate the trade-off between the surface area and the porosity in the hydrogen bromide RFB. In addition, the fabrication of the electrodes with bimodal pores is also a challenge, even for the variations in the woven structures such as carbon cloth. Alternatively, advanced manufacturing methods, particularly laser perforation, have been devised to control pore structure. Mayrhuber et al. [37] generated through-plane pores in carbon paper by a CO₂ laser and observed an improvement in RFB's performance. This approach allows for a controlled size of the generated pore morphology. Furthermore, the design of the pore structure also needs to consider the flow field structure [38], as the velocity and the active species distribution in the porous electrode are as the result of the combined effect of the flow field and the pore structure [39]. Dennison et al. [40] investigated the effects of the laser array perforated carbon paper for various flow field designs and found that array perforated carbon paper worked better in the flow channels with continuous paths from the inlet to the outlet (e.g., serpentine or parallel flow channels), compared to that where the flow channels with discontinuous paths (e. g., interdigitated) were applied. However, these studies have focused on the effects of the pore size and the perforation density, with less exploration related to how to design porous electrodes in conjunction with a certain flow channel structure. Thereby, the simultaneous optimization of the flow field structure and porous electrode is of great significance to further enhance the power density of RFB and to develop electrodes for the next-generation RFB [41,42]. Moreover, the effects of perforation location and varying operating conditions on the performance under different flow fields should also been further understood.

Inspired by the previous studies, in this paper we proposed a simple and highly actionable method to design porous electrode's pore structure in conjunction with the flow field structure to reconcile the combination of large and small pores. Large pores at the well-controlled size were generated by the infrared laser perforation and processed at different positions of the graphite felt considering the interdigitated flow field. The performance of the perforated graphite felts was tested in a VRFB single cell. Note that although the test was carried out for the flow cell with the interdigitated flow field, the proposed method can be easily generalized to other flow fields as well as various redox flow battery systems. In addition, a pore-network model was developed to investigate the effects of large pores formed by the laser perforation on the distributions with respect to local velocity, active species concentration, and reaction rate and explore the effects of the perforation location and diverse operating conditions on VRFB's overall performance. The rest of this paper proceeds as follows. Section 2 describes the VRFB single-cell experiments, the laser perforation method, and the pore-network model. Section 3 presents the experimental and simulation results and our discussion regarding practical application and future work, and Section 4 concludes the paper.

2. Methods

2.1. VRFB single cell experiments

In order to test the performance of the designed porous electrode, a VRFB flow cell was built (see Supplementary Material). As shown in Fig. S1(a), the experiments were conducted by using the setup including the electrochemical workstation (Bio-logic® VSP), electrolyte tanks, peristaltic pumps, and pressure sensors (Tem-tech® SE3300). The flow cell consisted of electrode frames, graphite felt electrodes, proton exchange membrane (Nafion® 117), current collector plates, and graphite bipolar plates with interdigitated flow field as shown in Fig. S1(b). The geometric parameters of the flow channel are summarized in Table S1. The electrodes used in the present study were graphite felts (SGL®, GFA5) with the thickness corresponding to approximately 500 µm, and the effective area corresponding to approximately 9 cm². A compression rate of 40 % was applied to the electrodes through the electrode frames to reduce the internal contact resistance. The electrolytes (1.6 M vanadium in 4 M sulfuric acid) were pre-charged to SOC = 0.5 before experiments and circulated by using the peristaltic pumps (Masterflex®, L/ S 07,525-40). In this work, the performance of different designed electrodes was compared at the same superficial velocity (v_e , m/s):

$$v_e = \frac{Q}{t_e w_e} \tag{1}$$

Where *Q* is the volumetric flow rate (m^3/s) , t_e is the compressed electrode thickness (m), and w_e is the electrode width. All experiments



Fig. 1. (a) Symmetric structure of interdigitated flow field and division of electrode regions. Red dashed lines illustrate the flow direction, and the yellow dashed lines illustrate the division of the electrode regions. Subplots (b)–(d) illustrate different perforated schemes: (b) perforation under the center of the half inlet channel, (c) perforation under the center of the rib, and (d) perforation under the center of the half outlet channel. Note that the illustration of the perforated pores is not plotted to scale.

were performed at the constant superficial velocity ($v_e = 2.97 \times 10^{-4} \text{ m/s}$, which corresponds to a volumetric flow rate of 10 mL/min) unless otherwise stated. For all the experiments, the volume of the electrolyte in the catholyte and anolyte tanks was 50 mL at a constant ambient temperature (25 °C) and under the nitrogen protection.

2.2. Laser perforation

The graphite felt electrodes were perforated in the present study by using an infrared laser perforation system (UNIVERSAL Laser System® VLS3.50), as shown in Fig. S2 (*Supplementary Material*). All graphite felts to be perforated (as well as the original graphite felts) were thermally

treated at the rate corresponding to 5 $^{\circ}$ C/min until 400 $^{\circ}$ C for 24 h to enhance the reactivity of the graphite felts.

Sun et al. has shown that different flow field designs corresponded to different typical flow paths [43], such as U-shaped flow path (interdigitated/serpentine flow fields). Besides, Messaggi et al. [39] and Wang et al. [44] found a significant difference in the flow rate and electrochemical reaction rate of the electrolyte under the flow channel and under the rib when the interdigitated flow field was applied. Considering the symmetry of the interdigitated flow field and its resulting U-shaped flow path, in the present study, we divided the electrode into three regions (under the half of the inlet channel, under the rib, and under the half of the outlet channel) as shown in Fig. 1(a). Pores with the



Fig. 2. (a) Schematic of the computational domain. (b) Comparison between the experimentally measured pore size distribution through MIP and that obtained through the pore-network modelling for the pristine graphite felt. Sim. and Exp. are short for simulative and experimental results, respectively.

diameter $d = 100 \ \mu\text{m}$ and spacing $s = 200 \ \mu\text{m}$ were generated by using the infrared laser along the centerline of three regions as shown in Fig. 1 (b)–(d).

In order to validate the effect of the perforation, we tested the pore size distribution of the three designed porous electrodes and the original graphite felt by using MIP (Mercury Intrusion Porosimetry, Micromeritics® AutoPore IV 9500), and the detailed results will be discussed in Section 3.1.

2.3. Pore-network model

In order to investigate the large pores formed by using the laser perforation on the distributions of the local velocity, active species concentration and reaction rate, and thus better understand the effects of the perforation location and diverse operating conditions on the overall performance, a pore-network model based on Sadeghi et al. [36] [45] was developed. The pore-network model was demonstrated to significantly reduce computational resources compared to direct numerical simulations (e.g., LBM) and to be more effective for pores with the regular geometry. The pore-network model is based on the following assumptions:

- (1) The electrolyte is considered to be a dilute solution;
- (2) Each pore is a well-mixed entity, while within the pores, intensive properties such as pressure and concentration vary slightly;
- (3) The reaction occurs only in the pore but not in the throat.

Computational domain. In the present study, the cathode and anode of VRFB operating under the steady-state were modeled respectively. Where the reactions in the cathode and anode are described by Eqs. (1) and (2), respectively.

$$VO_2^+ + 2H^+ + e^- \rightarrow VO^{2+} + 2H_2O, E_0^+ = 1.004V$$
 (2)

$$V^{2+} - e^- \rightarrow V^{3+}, E_0^- = -0.255V$$
 (3)

As shown in Fig. 2(a), the computational domain was chosen to be the porous electrode with the half of the inlet channel, a full rib, and the half of the outlet channel due to the symmetry of the interdigitated flow field. The rib and membrane defined the boundary conditions. Due to the interdigitated flow field, the active species concentration was considered to remain constant along the channel. The cathode and anode were structurally identical and differed in their boundary conditions and other physical parameters.

Geometric modeling. The pore-network model was employed in this study with the dimension corresponding to approximately $2 \times 2 \times 0.5$ mm for the x, y, and z directions, respectively. A cubic pore network was used to represent the original graphite felt electrode. The network was assumed to be fully connected with the coordination number of pores corresponding to 6 and being equally spaced in the x, y, z directions (center-to-center spacing set as 85 µm). As illustrated in Fig. 2(b), in this work, the pore size distribution of the original graphite felt was determined by MIP tests, and fitted to a log-normal bimodal pore size distribution to reasonably represent the porous structure of the graphite felt. The pores produced by using the laser perforation ($d = 100 \ \mu m$, s =200 μ m) were represented by inserting a single-layer network with the center-to-center spacing of 100 μ m in the x and y directions and the center-to-center spacing of 125 µm in the z direction at different locations along the x-axis. By controlling the pore size and throat diameter to 100 µm at odd grid points in the y direction, we could simply represent the large pores formed by using the laser perforation as shown in the cross-sectional view in Fig. 2(a), while the pore size distribution at the remaining grid points kept consistent with that in the pristine graphite felt (Section 3.1).

Velocity distribution. The velocity distribution under steady-state conditions is solved through the Stokes flow module in OpenPNM (an

open source project built by Gostick et al. [35]), which is based on the following mass conservation:

$$-\sum_{i=1}^{n_i} \rho u_{ij} S_{ij} = 0$$
 (4)

Where, n_i denotes the number of pores neighboring pore *i*, u_{ij} denotes the flow rate from pore *i* to pore *j*, and S_{ij} denotes the cross-sectional area of the connecting throat. To accurately describe the velocity between pores, the Hagen-Poiseuille equation is defined as:

$$u_{ij} = \alpha_{ij} (p_i - p_j) \tag{5}$$

Where, p_i and p_j denote the pressure of pore *i* and pore *j*, respectively, and $\alpha_{ij} = S_{ij}/8\pi\mu l_{ij}$ denotes the hydraulic conductance of the throat with the length l_{ij} . The inlet flow rate, Q_{in} and discharge pressure $p_{out} = 0$ are adopted as the inlet and outlet boundary conditions, respectively, and the rest of the boundaries are adopted as no-flux conditions.

Concentration distribution. The concentration of the active species is solved based on mass conservation, which for pore *i* can be expressed as

$$\sum_{j=1}^{n_i} m_{ij} S_{ij} = R_i = k_i c_i$$
 (6)

Where, k_i is the apparent rate constant, R_i is the net reaction rate in pore *i*, and m_{ij} denotes the mass flux from pore *i* to pore *j*, which is expressed by the following equation [46]:

$$m_{ij} = u_{ij} \left(c_i + \frac{c_i - c_j}{\exp\left(\frac{u_{ij}l_{ij}}{D_i}\right) - 1} \right)$$
(7)

Where, c_i and c_j denote the concentrations of species in pore *i* and pore *j*, respectively, and D_i denotes the diffusivity of the active species *i*. The inlet boundary is constant active species concentration c_{in} (VO_2^+ and V^{2+} for cathode and anode, respectively), and the rest of the boundaries are no-flux conditions.

Potential distribution. The charge conservation can be expressed as Eq. (7):

$$-\sum_{j=1}^{n_i} I_{ij}A_{ij} = R_i^p \tag{8}$$

Where, I_{ij} is the charge flux from pore *i* to pore *j* and R_i^p is the net charge generation rate in pore *i*. The charge flux I_{ij} is linearly related to the potential difference between the two pores and can therefore be replaced by the following equation:

$$I_{ij} = \beta_{ij} \left(\phi_i - \phi_j \right) \tag{9}$$

Where, $\beta_{ij} = \sigma_l / l_{ij}$ is the electrical conductance of the connecting throat, σ_l is the bulk electrical conductivity of the electrolyte, ϕ_i and ϕ_j denote the electric potential of the electrolyte at pore *i* and pore *j*, respectively. R_i^p can be calculated through the Butler-Volmer kinetics, which is defined as follows:

$$R_{i}^{p} = j_{0}A_{i}\left(\frac{c_{i}}{c_{0}}\right)\left[\exp\left(\alpha_{a}\frac{zF}{RT}\eta_{c}\right) - \exp\left(-\alpha_{c}\frac{zF}{RT}\eta_{c}\right)\right]$$
(10)

Where, j_0 is the exchange current density, α_a and α_c are the transfer coefficients of the cathode and anode, respectively, and $\alpha_a + \alpha_c = 1$. A_i is the internal surface area of pore *i*, c_0 is the reference concentration of active species when measuring the open-circuit voltage V_{oc} , *z* is the number of electrons involved in the reaction, *R* is the universal gas constant, *T* is the operating temperature, *F* is the Faraday constant, and η_c is the potential difference between the solid and liquid phases.

$$\eta_c = \phi_s - \phi_l - V_{OC} \tag{11}$$

The average voltage loss across the membrane, $\Delta \phi_m$, is considered in

Table 1

Variables used in the present study.

Variable	Description	Units
u _{ij}	Fluid velocity from pore <i>i</i> to pore <i>j</i>	m/s
Sij	Throat cross-section area	m ²
n _i	The number of pores neighboring pore i	-
α_{ij}	Throat hydraulic conductance	m ³ /(Pa·s)
p_i	Pressure at pore i	Pa
l _{ij}	Throat length	m
Q_{ij}	Flow rate from pore <i>i</i> to pore <i>j</i>	m ³ /s
m _{ij}	Mass flux from pore i to pore j	kg/(m ² ⋅s)
R_i	Reaction rate in pore <i>i</i>	kg/s
Ci	Concentration of active species at pore i	mol/m ³
I_{ij}	Charge flux from pore <i>i</i> to pore <i>j</i>	A/m ²
ϕ_i	Electric potential of electrolyte	V
R_i^p	Current generation at pore i	Α

Table	2
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Parameters used in the present study.

Parameter	Description	Value	Units
ρ	Electrolyte density	1000	kg/m ³
μ	Electrolyte average dynamic viscosity	$3.237e^{-3}$	Pa⋅s
Pout	Outlet pressure	0	Pa
D_{V2}	V ²⁺ diffusion coefficient	$2.4e^{-10}$	m ² /s
D_{V5}	V ⁵⁺ diffusion coefficient	$3.9e^{-10}$	m ² /s
C _{in}	Concentration of active species at inlet	800	mol/m ³
c _o	Reference concentration of active species	800	mol/m ³
α_a	anode transfer coefficient	0.5	-
α_c	Cathode transfer coefficient	0.5	-
σ_m	Electrical conductivity of membrane	8.3	S/m
δ_m	Membrane thickness	50	μm

this work to address the voltage loss across the membrane due to the relatively low electrical conductivity of the membrane. Then, this voltage loss can be compensated by setting the boundary condition at the cathode and anode membrane interface to $\phi_{l}=\Delta\phi_{m}$ and $\phi_{l}=0$, respectively. $\Delta\phi_{m}$ can be calculated based on Ohm's law as follows:

$$\Delta \phi_m = I_m R_m \tag{12}$$

Where, R_m is the ohmic resistance of the membrane and I_m is the net current through the membrane. Since the operating condition is steadystate, all protons passing through the membrane are involved in the reaction in the electrode, and I_m can be calculated by summing up the charge generation rate R_i^p for all the internal pores:

$$I_m = \sum_i R_i^p \tag{13}$$

The ohmic resistance of the membrane can be calculated according to Pouillet's law:

$$R_m = \frac{\delta_m}{\sigma_m A_m} \tag{14}$$

Where, σ_m is the electrical conductivity of membrane, A_m and δ_m is the cross-section area and the thickness of the membrane, respectively. The potential of the solid phase is assumed to be constant throughout the electrode and equal to the voltage applied to the rib $\phi_s = V_{cell}$ in this work, since the electirc conductance of the solid phase is relatively high. It should be noted that the apparent reaction rate constant k_i is not a constant: Since the reaction rate of the active species is coupled with generated current inside the electrode, it needs to be solved iteratively. The iterative scheme is described subsequently. And for other boundaries, no-flux boundary conditions are applied. The charge generation rate R_i^p is linearly proportional to the reaction rate of active species:

$$R_i = \frac{R_i^{\rho}}{zF} \tag{15}$$

The apparent reaction rate constants can be solved by combining



Fig. 3. Polarization curves and power density curves obtained through the pore-network model and experiments. Sim. and Exp. are short for simulative and experimental results, respectively.

$$k_i = \frac{1}{zF} \frac{j_0 A_i}{c_0} \left[\exp\left(\alpha_a \frac{zF}{RT} \eta_c\right) - \exp\left(-\alpha_c \frac{zF}{RT} \eta_c\right) \right]$$
(16)

Numerical solution. In this work, the solution was programmed through Python. An iterative scheme as shown in Fig. S3 was used to address the coupling between the concentration and potential fields. The overpotential η_c is initially guessed, and then continuously refined during the iterative process until the residual becomes within 1 %. It should be noted that in this work the voltage loss across the membrane is compensated by setting the boundary condition $\phi_l = \Delta \phi_m$ on the membrane interface of the cathode, so that $\Delta \phi_m$ is alsoupdated during each iteration.

The variables and parameters used in this work are summarized in Table 1 and Table 2, respectively. Values are taken from the previous studies (Qiu et al. [32] and Wang et al. [47]).

2.4. Model validation

The experimental polarization curves and power density curves under the pristine graphite felt at the superficial velocity $v_e = 2.97 \times 10^{-4}$ m/s (corresponds to a volumetric flow rate of 10 mL/min) and the corresponding results obtained by using the pore-network model are compared in Fig. 3. It can be seen that the experimental and simulated data fit better at relatively high current densities, but the discrepancy is approximately 5.7 % at relatively low current densities. The reason for the large deviation of the simulation results may be attributable to that the pore-network model simplifies the pore structure to spheres and cylinders. Recognizing that at low current densities, voltage losses mainly consist of activation polarization, this approach leads to loss of information at the pore scale, which underestimates the specific surface area of the electrode, and then the overall reaction area decreases. We also compared modeled and experimentally obtained in-plane and through-plane permeability coefficients for pristine graphite felts and electrodes with different perforation methods. The corresponding results are provided in Supplementary Material as Fig. S4.

3. Results and discussion

3.1. Characteristics of pore size distribution

Before conducting single-cell experiments, the pore size distributions of the three designed porous electrodes were measured and compared against the original graphite felt by using MIP. As shown in Fig. 4, the



Fig. 4. Pore size distribution obtained by MIP. Subplots (a)–(c) show the differences in the number of large pores for three perforation methods in the experiments: (a) perforation under the center of the half inlet channel, (b) perforation under the center of the rib, and (c) perforation under the center of the half outlet channel.

MIP results show that all three different perforation methods enhance the percentage of large pores with the diameter corresponding to approximately 100 µm, and that two perforation methods, under the center of half inlet channel and under the center of half outlet channel, provide more enhancement. This is due to the fact that although the number of large pores is the same for all three methods within the computational domain, the number of large pores produced by these two methods is more within the whole electrode range (details see Fig. 4). Besides, there is an increase in some of the pores larger than 100 µm, which may be due to the pores produced by infrared laser perforation are through pores and not ideal pores with an actual diameter of 100 µm, and the infrared laser perforation process may caused the carbon fibers around the perforated area to break and fall off, which increases the number of pores with diameter beyond 100 µm during the measurement process. Also by perforation, some of the "shielded" pores in the electrodes become accessible, which may be one of the reasons.

In addition, the porosity of the pristine graphite felt obtained by MIP test is 90 %, and the porosity of the three laser perforation schemes

under the center of half inlet channel, under the center of half outlet channel and under the center of the rib are 94 %, 94 %, 92.5 %, respectively. The slight increase in porosity is as expected, and as mentioned before, although the number of large pores is the same for all three methods within the computational domain, the number of large pores produced by the two methods, i.e., under the center of half inlet channel and under the center of half outlet channel, is more within the whole electrode range. Cho et al. [48] showed that laser perforation had slight effects on carbon fiber's surface functional groups, but minor effects on flow cell's overall performance. Therefore, the surface functional groups are not investigated in this work.

3.2. Experimental results

The polarization curves and power density curves under the laser perforated graphite felts are compared against those under the pristine felt as shown in Fig. 5. It can be observed that the perforation under the center of the rib significantly improves the overall performance with the



Fig. 5. Polarization and power density curves obtained from single-cell tests. Inlet, Outlet and Rib refer to different perforation methods.

maximum current density reaching 340 mA/cm² at the superficial velocity $v_e = 2.97 \times 10^{-4}$ m/s (corresponds to a volumetric flow rate of 10 mL/min), and the peak power density improving approximately 2.4 times compared to that with the pristine graphite felt. The two perforation schemes under the center of half inlet channel and under the center of half outlet channel improve the maximum power density slightly by 5 % and approximately 10 %, respectively. Further explanation for the underlying mechanism can be found in Section 3.4. In addition, the reasons for the differences in performance enhancement between experiments and simulations are analyzed and relevant discussion can be found in *Supplementary Material*.

3.3. Effects of laser perforation on local distributions

Fig. 6 further illustrates the effects of the perforated large pores on the distributions of the local velocity, active species and reaction rate by using the pore-network model. As shown in Fig. 6(b)–(d), all three different laser perforation schemes construct low flow resistance pathways in the through-plane direction and change the flow resistance distribution in the in-plane direction, resulting in increased velocities in the porous electrode near the membrane side, especially in the region under the rib. This results in an enhanced convective mass transfer by the membrane side, faster replenishment of active species, and thereby a significant increase in the high reaction rate region compared to that with the pristine graphite felt (Fig. 6(a)). It can be seen that compared to the other two perforation schemes, the perforation under the center of the rib enhances larger high reaction rate area (see red circle in the figure).

3.4. Mechanism of laser perforation on performance enhancement

In order to further understand the mechanism of the produced large pores on the flow-cell's performance, we further investigated the peak power density enhancement when perforation was applied at different locations and under different superficial velocities. Note that each point in Fig. 7 represents the results obtained by comparing the polarization curves that obtained for the electrodes with different perforation locations to that with the pristine graphite felt under specific velocities. Detailed polarization curves can be found in *Supplementary Material* (see Fig. S5).

The position ratio θ is defined to characterize the variation of the perforated location as the ratio of the perforated position to the length of

the porous electrode in the x-direction. Where $\theta < 0.25$ represents the perforation position under the inlet channel region, $0.25 < \theta < 0.75$ means the perforation position under the rib, and $\theta > 0.75$ means the perforation under the outlet channel. The variation of the superficial velocity is controlled by changing the total electrolyte pressure drop ΔP from the inlet to the outlet of the pore-network model. Electrode performances are evaluated at different superficial velocities, 5.09×10^{-5} , 2.55×10^{-4} , 5.09×10^{-4} , 1.02×10^{-3} and 2.55×10^{-3} m/s, which correspond to total pressure drops of 2, 10, 20, 40, 100 Pa, respectively. While the range of values for the velocity is close to the VRFB general operating range and is discussed in detail in Section 3.4.

As shown in Fig. 7, the performance improvement is most pronounced at low superficial velocities, with an average performance improvement by over 52 % at $v_e = 5.09 \times 10^{-5}$ m/s. At such a velocity, perforation at θ =0.261 and θ =0.739 both lead to a better effect on the performance improvement (approximately 60 %), corresponding to the area under the rib near the inlet channel and under the rib near the outlet channel, respectively. When the applied superficial velocities increase, the trends with varying perforation locations change as only one performance improvement peak is observed in each peak power density enhancement curve. To be specific, the performance is at its best when the perforation is applied under the rib near the outlet channel. To further explain the above observations, the concentration distributions inside the cathode for different superficial velocities and perforation positions are exhibited in Fig. 8.

Based on the simulative results, the large pores produced by the infrared laser function in two distinct ways for enhancing mass transfer in the through-plane and the in-plane directions, respectively. To be specific, one is to change the flow resistance in the through-plane direction, and the other is to change the flow resistance distribution in the in-plane direction within the porous electrode, while the performance enhancement through these two mechanisms varies with different perforation locations and different velocities. As a result, the behavior of data points in Fig. 7 is not uniformly monotonic across velocities.

At the relatively low velocity $v_e = 5.09 \times 10^{-5}$ m/s (corresponding to $\Delta P = 2$ Pa), perforation at both $\theta = 0.261$ and $\theta = 0.739$ gives high performance gains, corresponding to the area under the rib near the inlet channel and under the rib near the outlet channel, respectively. This is due to that at a relatively low velocity, both above mechanisms achieve better effects due to the extremely non-uniform distribution of the active species in the porous electrode. As shown in Fig. 8(a), the perforation in the area under the rib near the inlet channel ($\theta = 0.261$) constructs low-



Fig. 6. Reaction rate distribution and velocity distribution (cathode, $v_e = 2.97 \times 10^{-4}$ m/s). The red throats represent the region in the porous electrode where the local velocity is relatively high (> 0.003 m/s). (a) represents the case with the pristine graphite felt, (b)–(d) represent electrodes perforated under the center of half inlet channel, under the center of rib, and under the center of half outlet channel, respectively.

flow resistance pathways in the through-plane direction, resulting in a significant enhancement in the mass transfer in the through-plane direction. While the perforation under the rib near the outlet channel (θ =0.739) changes the flow resistance distribution of the porous electrode, resulting in enhanced mass transfer in the in-plane direction and uniform concentration distribution of the active species, which also

enhances the performance.

With increasing superficial velocity (e.g., $v_e=1.02\times10^{-3}$ m/s), as shown in Fig. 8(b), the distribution uniformity of active species in the area under the inlet channel remarkably improves. Therefore, the effectiveness of the former mechanism diminishes, the perforation near the inlet channel (θ =0.261) has minor effects on the concentration



Fig. 7. Variation of peak power density improvement as compared to that with the pristine graphite felt at different perforation positions and under varying superficial velocity setups. The superficial velocities of 5.09×10^{-5} , 2.55×10^{-4} , 5.09×10^{-4} , 1.02×10^{-3} and 2.55×10^{-3} m/s can be converted to pressure drops corresponding to 2, 10, 20, 40, 100 Pa, respectively. The results are simulated by using the developed pore network model for the computational domain as illustrated in Fig. 2.

distribution uniformity in the through-plane direction. However, the latter mechanism still achieves better effects: that is the perforation under the rib near the outlet channel effectively improves the distribution uniformity in the in-plane direction. Thus, the jump at position $\theta \approx 0.25$ disappears as the velocity increases.

Although, the performance gain obtained through the perforation is decreasing with increasing velocity, this trend is reversed for certain velocity intervals (v_e ranging from 2.55×10^{-4} to 1.02×10^{-3} m/s). This observation is due to that with increasing superficial velocity, the location where the concentration gradient changes sharply in the through-plane direction shifts toward the outlet channel gradually, as shown in Fig. 8(c). When the large concentration gradient is near the outlet channel, perforation under the rib near the outlet channel ($\theta \approx 0.75$) gives better effects with enhanced mass transfer in both through-plane and in-plane directions.

As the velocity increases further to extremely high values such as 2.55×10^{-3} m/s, the whole electrode tends to a high concentration state,

at which point perforation does not significantly change the concentration distribution in the through-plane and in-plane directions, so both mechanisms yield low gains, the improvement in overall performance stabilizes, and the jump at position $\theta \approx 0.75$ disappears.

However, such large velocities are not used in the general operating conditions of the VRFB, which can cause irreversible damage to the system's hermeticity and porous electrode.

3.5. Practical application

For the general working conditions of VRFB, the flow rate range in 0.4–1.23 cm³/(min·cm²·N) [49], for the electrode size investigated in this paper this flow rate range can be converted to the superficial velocity v_e range from 1.07×10^{-4} to 3.29×10^{-4} m/s. Too high velocity will not only damage the flow cell, but also increase the pump loss. For interdigitated fluid field, in such a velocity range, the perforation under the rib near the outlet channel maximizes the performance gain. Considering the manufacturing accuracy, in practical applications, regions between the center of the rib and the outlet channel is also appropriate choice for laser perforation to produce large pores. In addition, die cutting [50] can be used in practical production to produce pores similar to laser perforation, which can significantly reduce the cost.

Furthermore, the porous electrode design scheme presented in the present paper can be easily extended to other systems of redox flow batteries. In general, perforation between the center of the rib to the outlet channel is the more widely applicable option. For electrolytes with particularly high viscosity (e.g., aqueous organic systems), it is expected that perforation under the center of the rib to the inlet channel will yield better performance. Moreover, for systems with large differences in electrolytes' viscosity between cathode and anode, such as zincbromine flow batteries, it is possible to design different perforation schemes at cathode and anode electrodes based on the physical properties of the catholyte and anolyte, respectively.

3.6. Future work

The present method can also be easily extended to other flow field structures (e.g., parallel and serpentine, etc.). For a parallel flow field, we believe that perforation under the rib is a better choice because this type of perforation is expected to have a better enhancement of the active species uniformity perpendicular to the direction of flow. In the electrodes of the RFB with a serpentine flow field, the local electrolyte



Fig. 8. Concentration distributions of V^{5+} in the cathode under different perforation locations and varying superficial velocities. The yellow dashed lines indicate the location where the concentration of the V^{5+} is 50 % of its value at the inlet.

infiltration is mainly driven by the pressure difference between neighboring flow channels, so the distribution of electrolyte penetration is consistent with the trend of the distribution of the pressure difference, and there is an obvious non-uniformity of the penetration distribution. Therefore, in addition to adopting the similar perforation scheme as that applied for the interdigitated flow field, different perforation methods can be considered to optimize the distribution of flow resistance between neighboring channels of the serpentine flow field to improve the electrolyte penetration uniformity. We provide some design guidance for perforation schemes specifically for a flow cell with a serpentine flow field in *Supplementary Material*. Future work can also adjust the perforation pore size and spacing to achieve a better tradeoff between specific surface area and hydraulic permeability to further improve performance.

4. Conclusions

In this paper, three porous electrodes were crafted through laser perforation in conjunction with typical interdigitated flow field, and VRFB single-cell tests were performed. To enhance our understanding, a pore-network model was constructed for the comprehensive analysis of the effects of different perforation methods on the velocity, concentration and reaction rate distributions within the electrode. Some major findings and contributions of this research are outlined as follows:

- (1) We introduced a novel method to design porous electrode's pore structure in conjunction with the flow field based on the infrared laser perforation to reconcile the combination of large and small pores. Experimental results revealed a substantial enhancement in overall performance when perforating under the center of the rib, with a maximum power density increase corresponding to approximately 2.4 times compared to that with the pristine graphite felt.
- (2) To elucidate the underlying mechanisms behind the performance enhancement in flow cells through laser perforation, we developed a pore-network model. This model was employed to investigate the impact of perforation location and diverse operating conditions. Under general operating conditions of the VRFB, perforating in the region under the rib near the outlet channel was expected to provide more performance gains.

CRediT authorship contribution statement

Wenrui Lv: Conceptualization, Methodology, Software, Investigation, Validation, Visualization, Writing – original draft. Yansong Luo: Data curation, Investigation, Validation, Visualization, Writing – original draft. Yuhong Xu: Formal analysis, Methodology, Investigation. Kaichen Xu: Investigation, Methodology, Supervision. Menglian Zheng: Writing – review & editing, Investigation, Funding acquisition, Supervision, Project administration.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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Data availability

Data will be made available on request.

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Supplementary materials

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