同行专家业内评价意见书编号: \_20250858245

# 附件1 浙江工程师学院(浙江大学工程师学院) 同行专家业内评价意见书

**学号:** <u>22260455</u>

浙江工程师学院(浙江大学工程师学院)制 2025年05月10日

## 填表说明

一、本报告中相关的技术或数据如涉及知识产权保护 、军工项目保密等内容,请作脱密处理。

二、请用宋体小四字号撰写本报告,可另行附页或增 加页数,A4纸双面打印。

三、表中所涉及的签名都必须用蓝、黑色墨水笔,亲 笔签名或签字章,不可以打印代替。

四、同行专家业内评价意见书编号由工程师学院填写,编号规则为:年份4位+申报工程师职称专业类别(领域)4 位+流水号3位,共11位。 一、个人申报

(一)基本情况【围绕《浙江工程师学院(浙江大学工程师学院)工程类专业学位研究生工程师职称评审参考指标》,结合该专业类别(领域)工程师职称评审相关标准,举例说明】

1. 对本专业基础理论知识和专业技术知识掌握情况(不少于200字)

在实践中,对储能行业的背景、世界各国的储能政策布局和发展态势,以及各类储能方式的 优劣进行了系统的学习。在理论知识方面,我学习了电化学、流体力学、电路原理、优化算 法等多学科知识,深化了对这些领域的理解,并掌握运用学科交叉思维破解实际问题的能力

随着全球能源结构的转型与可再生能源的大规模接入,储能技术作为解决能源供需不匹配、 提高电力系统灵活性与稳定性的关键手段,其重要性日益凸显。这一认识让我深刻体会到, 液流电池作为储能技术的一种重要形式,其研发与应用对于推动能源绿色转型、实现可持续 发展目标具有不可估量的价值。不同国家和地区基于自身的能源结构、经济发展需求及环境 保护目标,制定了各具特色的储能政策与战略规划。中国自2021年以后,储能市场也进入了 真正意义上规模化发展。在支撑新能源体系和"双碳"战略意义方面,70%以上的储能需求 是大规模、长时间的储能,主要包括:抽水蓄能、压缩空气储能、液流电池储能。液流电池 储能技术具有能量效率高、响应速度快、不受地理条件限制等优点,是最具发展前景的长时 储能方式。另外,液流电池不会出现易燃易爆的问题,其安全性优势在大型电化学储能电站 中更加突出。这些政策的出台与实施,不仅为储能技术的发展提供了强有力的政策保障与市 场激励,也促进了储能产业的快速发展。明确了液流电池在储能领域中的独特优势与潜在应 用前景,同时也认识到其在技术成熟度、成本控制等方面面临的挑战与不足。

## 2. 工程实践的经历(不少于200字)

在实习期间参与了国家重点研发计划:水运港 —

船多能源融合技术及集成应用(示范应用)。本人负责储能系统中全钒液流电池(VRFB)储 能系统的电堆结构设计。全钒液流电池作为大规模、长期储能技术,具备安全、响应快等优 势。电堆中流道结构设计直接影响电解液流动与电池性能,合理的流道结构可提升电解液利 用率和电池能量效率,但其设计需综合考虑电解液流速、流量、压力分布等因素,是电堆结 构开发的技术难点。目标是设计出一种 kW 级高效率的钒液流电池电堆结构。

## 3. 在实际工作中综合运用所学知识解决复杂工程问题的案例(不少于1000字)

在实际工作中,全钒液流电池的性能与运营成本受多方面因素影响,其中部件材料、电池结构及运行条件均起着关键作用。因此,开展全钒液流电池单电池结构研究,对于提升其运行效率、推动大规模应用具有重要意义。鉴于现有研究多集中于实验室小型电池,本次实践着重建立了 kW

级全钒液流电池的三维多物理场仿真模型,通过仿真对比不同单电池流道结构的流场分布与放电性能,筛选出最优流道结构,并进行电堆仿真分析,旨在为 kW

级钒液流电池电堆设计提供有力参考。

在单电池结构设计方面,采用 COMSOL

多物理场仿真软件,建立全钒液流电池单电池的三维模型,针对不同的电极框导流结构和电极结构开展流场电化学耦合仿真。深入探究在不同流速、电流密度条件下,单电池电化学性能的变化情况,以及不同结构下流场对钒液流电池传质和电化学性能的影响,为优化单电池结构提供详实依据。通过仿真发现,当电极框流道的通道数较少时,多孔电极内部的速度分布相对不均匀,电解液流速较低的区域反应离子难以通过对流进行补充,进而产生较大的浓

差极化,影响单电池的充放电性能。当通道数较多时,流道中的流动阻力较大,引起多孔电极的入口侧横向压力不均匀,导致电解液在电极中出现对角流动,影响电解液的分布均匀性。最终通过仿真优化设计,得到了5通道的优化结构。

为确保仿真结果的可靠性与准确性,对单电池结构搭建了试验台并进行实验测试,将仿真预测结果与实验数据进行对比,验证仿真模型的精确度,为 kW

级全钒液流电池电堆的设计方法奠定坚实基础。

对于电堆的仿真分析,基于 COMSOL 软件得到的单电池仿真结果构建简化模型,在 Simulink

中搭建综合考虑旁路分流电流和物质传递的模型,对不同参数的电堆结构进行全面的效率和性能评估,进而开展电堆优化设计,通过遗传算法对10kW/40kWh电堆的结构参数进行了优化设计,提升电堆整体性能。

在实践过程中,发现全钒液流电池在系统运行时电解液温度存在一定变化,若后续能将温度 变化因素纳入多物理场模型,将进一步提高仿真的准确性。

本次实践对 kW

级钒液流电池电堆的研究意义重大。一方面,通过优化设计提高电池能量效率,提升电解液利用率,这意味着在满足同等储能要求的前提下,可减少电解液用量,进而降低企业初始投资成本;另一方面,kW

级高功率的钒液流电池能够有效增强电池的功率输出能力,使其能够迅速响应高能量需求场景,如电网调峰、大型数据中心备用电源等,且进一步缩短响应时间,在需要快速调节电力输出的应用场景中具有显著优势。此外,高功率版本的钒液流电池在设计时通常考虑到更大的容量需求,更易于实现 MWh

级别的庞大容量,同时还能减少电堆的材料和连接管路等附加成本,有力推动全钒液流电池储能的大规模商业化进程。

(二)取得的业绩(代表作)【限填3项,须提交证明原件(包括发表的论文、出版的著作、专利 证书、获奖证书、科技项目立项文件或合同、企业证明等)供核实,并提供复印件一份】

1.

公开成果代表作【论文发表、专利成果、软件著作权、标准规范与行业工法制定、著作编写、科技成果获奖、学位论文等】

| 成果名称   | 成果类别<br>[含论文、授权专利(含<br>发明专利申请)、软件著<br>作权、标准、工法、著作<br>、获奖、学位论文等] | 发表时间/<br>授权或申<br>请时间等 | 刊物名称<br>/专利授权<br>或申请号等                             | 本人<br>排名/<br>总人<br>数 | 备注          |
|--|---|-----------------------|--|----------------------|-------------|
| 一种大功率液流电池用<br>电极框  | 发明专利申请  | 2024年03<br>月06日       | 申请公布号<br>: CN<br>118099465<br>A                    | 5/12                 |             |
| Effect of flow frames<br>on performance of a<br>10 kW 40kWh vanadium<br>redox flow battery<br>based on a multi-<br>scale model | 一级期刊  | 2025年04<br>月07日       | Journal of<br>Zhejiang<br>University<br>-SCIENCE A | 1/8                  | SCI期刊<br>收录 |
|  |   |                       |  |                      |             |

2. 其他代表作【主持或参与的课题研究项目、科技成果应用转化推广、企业技术难题解决方案、自 主研发设计的产品或样机、技术报告、设计图纸、软课题研究报告、可行性研究报告、规划设计方 案、施工或调试报告、工程实验、技术培训教材、推动行业发展中发挥的作用及取得的经济社会效 益等】

| (三)在校期间课程、专业实践训练及学位论文相关情况               |                                    |  |  |  |  |  |
|---|------------------------------------|--|--|--|--|--|
| 课程成绩情况                                  | 按课程学分核算的平均成绩: 85 分                 |  |  |  |  |  |
| 专业实践训练时间及考<br>核情况(具有三年及以上<br>工作经历的不作要求) | 累计时间: 1.3 年(要求1年及以上)<br>考核成绩: 82 分 |  |  |  |  |  |
|   | 本人承诺                               |  |  |  |  |  |
| 个人声明:本人上述所填资料均为真实有效,如有虚假,愿承担一切责任,特此声明!  |                                    |  |  |  |  |  |
| 申报人签名:一杯 传流                             |                                    |  |  |  |  |  |

| 日常実知 | 非定向生由德育导师考核评价、定向生由所在工作单位考核评价:                                   |
|------|---|
| 考核评价 | ☑优秀 □良好 □合格 □不合格  |
|      | 德育导师/定向生所在工作单位分管领导签字(公章): WP74年9月日                              |
|      | 根据评审条件,工程师学院已对申报人员进行材料审核(学位课程成绩 专)                              |
| 申报材料 | 实践训练时间及考核、学位论文、代表作等情况),并将符合要求的申报材;<br>在学院网站公示不少于5个工作日,具体公示结果如下: |
| 甲核公小 | □通过 □不通过(具体原因: )  |
|      | 工程师学院教学管理办公室审核签字(公章): 年月日                                       |

# 浙江大学研究生院

|                 |                 |        |       |          |    | · · · · · · · · · · · · · · · · · · · | 11/00001/04/24/24 |                   |    |     |          |       |
|-----------------|-----------------|--------|-------|----------|----|---------------------------------------|-------------------|-------------------|----|-----|----------|-------|
| 学号: 22260455    | 姓名:林俊宏          | 性别: 男  |       | 学院:工程师学院 |    |                                       |                   | 专业: 能源动力          |    |     | 学制: 2.5年 |       |
| 毕业时最低应获: 29.    | . 0学分           | 已获得: 3 | 31.0学 | 分        |    |                                       |                   | 入学年月: 2022-09 毕业年 |    | (年月 | <br>月:   |       |
| 学位证书号: 毕业证书号:   |                 |        |       |          |    |                                       |                   |                   |    |     |          |       |
| 学习时间            | 课程名称            |        | 备注    | 学分       | 成绩 | 课程性质                                  | 学习时间              | 课程名称              | 备注 | 学分  | 成绩       | 课程性质  |
| 2022-2023学年秋季学期 | 创新设计方法          |        |       | 2.0      | 通过 | 专业选修课                                 | 2022-2023学年春季学期   | 研究生论文写作指导         |    | 1.0 | 87       | 专业学位课 |
| 2022-2023学年秋季学期 | 工程技术创新前沿        |        |       | 1.5      | 90 | 专业学位课                                 | 2022-2023学年夏季学期   | 自然辩证法概论           |    | 1.0 | 92       | 公共学位课 |
| 2022-2023学年秋季学期 | 工程数值分析          |        |       | 2.0      | 85 | 专业选修课                                 | 2022-2023学年夏季学期   | 研究生英语基础技能         |    | 1.0 | 免修       | 公共学位课 |
| 2022-2023学年秋季学期 | 工程伦理            |        |       | 2.0      | 84 | 公共学位课                                 | 2022-2023学年夏季学期   | 现代分析测试实验          |    | 2.0 | 85       | 专业选修课 |
| 2022-2023学年冬季学期 | 智慧能源系统工程        |        |       | 2.0      | 88 | 专业学位课                                 | 2022-2023学年春夏学期   | 高阶工程认知实践          |    | 3.0 | 81       | 专业学位课 |
| 2022-2023学年冬季学期 | 产业技术发展前沿        |        |       | 1.5      | 90 | 专业学位课                                 | 2022-2023学年夏季学期   | 材料工程产业与发展         |    | 2.0 | 86       | 专业选修课 |
| 2022-2023学年春季学期 | 绿色化工与生物催化前沿     |        |       | 2.0      | 91 | 专业学位课                                 | 2022-2023学年夏季学期   | 研究生英语             |    | 2.0 | 免修       | 公共学位课 |
| 2022-2023学年春季学期 | 材料加工技术          |        |       | 2.0      | 81 | 专业学位课                                 |                   | 硕士生读书报告           |    | 2.0 | 通过       |       |
| 2022-2023学年春季学期 | 新时代中国特色社会主义理论与经 | 实践     |       | 2.0      | 83 | 公共学位课                                 |                   |                   |    |     |          |       |
|                 |                 |        |       |          |    |                                       |                   | · La 132          |    |     |          |       |
|                 |                 |        |       |          |    |                                       | A                 |                   | ~  |     |          |       |

说明: 1. 研究生课程按三种方法计分: 百分制,两级制(通过、不通过),五级制(优、良、中、

及格、不及格)。

2. 备注中"\*"表示重修课程。

学院成绩校核章: 成绩校核人:张梦依 (60) 田 打印日期: 2025-06-03 成绩校校登室

## (19) 国家知识产权局



## (12)发明专利申请



(10)申请公布号 CN 118099465 A (43)申请公布日 2024.05.28

- (21)申请号 202410254978.4
- (22)申请日 2024.03.06
- (71)申请人 浙江大学
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   38号
- (72)发明人 宋浩 高翔 吴卫红 郑成航 林俊宏 王丁振 宋思慧 刘少俊 张霄 林赛赛 刘鹏 岑可法
- (74) 专利代理机构 浙江永航联科专利代理有限 公司 33304

专利代理师 张进

(51) Int.CI.

H01M 8/0273 (2016.01) H01M 8/0271 (2016.01) H01M 8/0258 (2016.01)

(54)发明名称

一种大功率液流电池用电极框

(57)摘要

本发明提供一种大功率液流电池用电极框, 包括电极框本体,所述电极框本体设有电极腔、 电解液入口和电解液出口,所述电解液入口和所 述电解液出口均分别连通电极腔与电极框本体 外部;所述电解液入口和所述电极腔之间、所述 电极腔和所述电解液出口之间均分别设有多级 分流通道,所述多级分流通道包括依次设置的一 级分流通道、二级分流通道和三级分流通道,所 述一级分流通道依次通过所述二级分流通道和 所述三级分流通道连通所述电极腔。本发明提高 电解液分布均匀性且降低电堆压降。 H01M 8/04276 (2016.01) H01M 8/04186 (2016.01) H01M 8/18 (2006.01)

权利要求书1页 说明书5页 附图3页



CN 118099465 A

#### 1 $Effect \cdot of \cdot flow \cdot frames \cdot on \cdot performance \cdot of \cdot a \cdot 10 \cdot kW \cdot 40 kWh \cdot vanadium$

- 2  $redox \cdot flow \cdot battery \cdot based \cdot on \cdot a \cdot multi-scale \cdot model \leftrightarrow$
- 3 Junhong Lin<sup>1</sup>, Zihao Ma<sup>2</sup>, <u>Shaojun Liu<sup>1,\*</sup></u>, Hao Song<sup>1,\*</sup>, <u>Guoneng Li<sup>3</sup></u>, <u>Weihong Wu<sup>1</sup></u>, <u>Chenghang</u> 4
  - Zheng<sup>1</sup>, Xiang Gao<sup>1,\*</sup>⊖
- 5 1. State Key Laboratory of Clean Energy Utilization, State Environmental Protection Center for Coal-Fired Air
- 6 Pollution Control, Zhejiang University, Hangzhou 310027, China
- 7 2. Shanghai Electric Group Co. LTD, Shanghai, 200336, China#
- 3. Department of Energy and Environment System Engineering, Zhejiang University of Science and Technology, 8
- 9 Hangzhou, 310023, China-



With kind regards

Thank you for submitting your work to this journal.

收件人: shaojun liu<phoenix205@zju.edu.cn

时间: 2025年4月7日 (周一) 10:22 🕥

Chunjie Zhang Managing Editor Journal of Zhejiang University-SCIENCE A

Reviewer #2: The manuscript has been improved and it can be accepted as it is.

010f01960e0e8a45-7cd52849-3898-441f-b642-971bd65b2a5a-000000 代表 Chunjie Zhang

010f01960e0e8a45-7cd52849-3898-441f-b642-971bd65b2a5a-000000<010f01960e0e8a45-7cd52849-3898-441f-b642-971bd65b2a5a-000000

Ref.: Ms. No. ZUSA-D-24-00544R1 Effect of flow frames on performance of a 10 kW 40kWh vanadium redox flow battery based on a multi-scale model Journal of Zhejiang University-SCIENCE A

I am pleased to tell you that your work has now been accepted for publication in Journal of Zhejiang University-SCIENCE A.

Reviewer #4: Accept.

发给 sl

发件人:

大小: 6 KB CC: menglian\_zheng@zju.edu.cn

Dear Dr. Liu,

#### 11 12 • Abstract

10

13

The structure of the cell in vanadium redox flow batteries (VRFBs) is a critical determinant of

Graphical-abstract

- 14 battery efficiency. A multi-scale model combining a three-dimensional model of the cell with an
- 15 equivalent circuit model (ECM) of the stack were established. The effect of different flow frames
- 16 was evaluated in terms of both cell performance and system efficiency. The results show that during
- 17 the discharge process, increasing flow rate can boost the discharge voltage, but it also leads to a
- 18
- higher pumping power consumption. The pressure drop of the cell decreases as the number of

|     | A  | В         | С         | D                  | E F                               | G H I J K L  |
|-----|--|-----------|-----------|--------------------|-----------------------------------|--|
| 326 | JOURNAL OF WATERWAY PORT COASTAL AND OCEAN ENGINE    | 0733-950X | 1943-5460 | ASCE-AMER SOC C    | 1801 ALEXANDER BELL DR, English   | Water Resources   Engineering, Civil   Engineering, Ocean  |
| 327 | JOURNAL OF WEB ENGINEERING                           | 1540-9589 | 1544-5976 | RIVER PUBLISHERS   | ALSBJERGVEJ 10, GISTRUP, English  | Computer Science, Software Engineering   Computer Scien    |
| 328 | JOURNAL OF WEB SEMANTICS                             | 1570-8268 | 1873-7749 | ELSEVIER           | RADARWEG 29, AMSTERD/ English     | Computer Science, Software Engineering   Computer Scien    |
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### Effect of flow frames on performance of a 10 kW 40kWh vanadium

### redox flow battery based on a multi-scale model

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#### Graphical abstract

#### Abstract

The cell structure in vanadium redox flow batteries (VRFBs) critically impacts battery efficiency. Therefore, in this study we evaluate the effect of different flow frames on VFRB cell performance and system efficiency. This is accomplished through a multi-scale model combining a 3D model of the cell and an equivalent circuit model (ECM) of the stack. The results reveal that during the discharge process, increasing the flow rate can boost the discharge voltage, but also leads to higher pumping power consumption. Moreover, the pressure drop of the cell decreases as the

number of flow frame channels increases. Due to improved reactant concentration distribution, the five-channel flow frame has a higher discharge voltage than other configurations at low reactant concentrations, even exceeding that of the twenty-channel flow frame. The flow frame structure and flow rate were optimized for a 10kW/40kWh VRFB with a genetic algorithm approach. Using a five-channel flow frame, a peak discharge efficiency of 93.70% was obtained at a flow rate of 960 mL/min. The results of this study may aid future design of kilowatt-scale VRFBs.

#### Keywords

Flow frames; All vanadium redox flow battery; Multi-scale model; Genetic algorithm; Battery efficiency

#### 1 Introduction

With the rapid growth of clean energy sources such as solar and wind power, energy storage technologies such as lithium-ion (Guo et al., 2023; Park et al., 2023; Guo et al., 2025) and flow batteries (Zerrahn and Schill, 2017; Albertus et al., 2020; Lu et al., 2024) are receiving increasing attention. In addition to benefits of long-duration energy storage and recyclability, flow batteries are intrinsically safer than lithium-ion batteries, due to the non-flammable aqueous electrolyte used. The flow field design critically affects the performance of flow batteries. For vanadium flow batteries, elaborate electrolyte channels are typically carved in the electrodes (Yang et al., 2022; Bhattarai et al., 2019) or bipolar plates (Zeng et al., 2019; Zheng et al., 2016); specially designed flow frames are also used to regulate the flow field. The configuration of such channels has been extensively investigated. For instance, Ali et al. (2022) compared the performance of serpentine and interdigitated channels, showing that 2 mm serpentine flow channels resulted in higher cell voltage and more uniform  $V^{2+}$  concentration in the negative electrode. However, serpentine flow channels also resulted in higher pump power consumption. Thus, Lu et al. (2021) optimized the discharge power and system efficiency of serpentine flow channels, demonstrating power and system efficiency that reached 16.73 W and 87.8%, respectively; these values were 2.85 W and 3.7% higher than the conventional approach. Sharma and Kumar (2021) also improved serpentine channels with two new designs: split serpentine and split-merged serpentine channels. Their experiments demonstrated better electrolyte distribution in the split serpentine channel. Additionally, Gundlapalli and Jayanti (2021) proposed a flip-flop directional split serpentine flow field, which demonstrated excellent distribution and quick evacuation of product species. Their novel channel configuration ensures scaling from 900 to 2200 cm<sup>2</sup> of active cell area.

Designing cells with large active area is becoming more common in industrial practice, but has not been extensively studied (Ponce de León et al., 2006; Sun et al., 2019). A large active area can effectively reduce the number of cells in the stack, which is more conducive to VRFB applications (Sun et al., 2019). Several cells can be connected to form a stack, enabling kW-scale VRFB systems. In such systems, channels carved in the flow frames (instead of bipolar plates) are extensively employed to reduce material costs (Kim et al., 2013; Delgado et al., 2022; Guarnieri et al., 2018). However, this usually leads to kW-scale VRFBs having lower energy efficiency compared to batteries employing laboratory-scale equipment (Kapoor and Verma, 2022).

Only a few studies have investigated the effect of flow frames and achieved kW-scale VRFBs. For example, Kim et al. (2013) demonstrated a flow frame design in a 1kW/1kWh VRFB, with the average energy efficiency reaching 82% at a low current density of 80 mA/cm<sup>2</sup>. And Guarnieri et al. (2018) tested a 9kW/26kWh VRFB with 40 cells, in which the flow frames followed an equal path length design.

In a typical cell, good electrochemical performance is always accompanied by an increased pressure drop. However, the conventional equivalent circuit model (ECM) for VRFBs (Delgado et al., 2022; Zhao et al., 2023) is unable to calculate this pressure drop. It also cannot represent the dynamics of cell voltage during battery operation. For this reason, we propose a multi-scale model which combines a 3D model of the cell with an ECM of the stack. Using this model, the effect of different flow frames on the performance of a 10 kW/40kWh vanadium redox flow battery are investigated. Varying structures are compared in terms of VRFB system efficiency and cell performance.

#### 2 Numerical modeling

#### 2.1 VRFB description

As shown in Fig. 1, the studied VRFB consists of an electrical stack, two electrolyte storage tanks, two pumps, and connecting pipes. The stack is comprised of multiple cells connected in series. A cell consists of two porous electrodes and an ion exchange membrane between them. Two tanks are used to store the anolyte and catholyte. During operation, electrolyte is pumped into the porous electrode and electrochemical reactions take place. The reactions at the electrodes are described by Eqs. (1) and (2).

Positive electrode:

$$VO^{2+} + H_2O - e^- \rightleftharpoons VO_2^+ + 2H^+$$
 (1)

Negative electrode:

$$V^{3+} + e^- \rightleftharpoons V^{2+} \tag{2}$$

The state of charge (SOC) is used to describe the relative amount of vanadium ions with different valences, in both the porous electrodes and the tanks. It is defined as follows:

$$SOC = \frac{c(V^{2+})}{c(V_{\text{total}})} = \frac{c(V^{5+})}{c(V_{\text{total}})}$$
(3)

where  $c(V^{2+})$  and  $c(V^{5+})$  are the molar concentrations of  $V^{2+}$  and  $VO_2^+$ , respectively, and  $c(V_{\text{total}})$  is the total vanadium ion concentration in a given electrolyte.



Fig. 1 Schematic diagram of the vanadium redox flow battery

#### 2.2 Description of the VRFB cell

In this work, a three-dimensional (3D) VRFB cell was constructed. The VRFB cell consists of porous electrodes, current collectors, a proton exchange membrane, bipolar plates, and flow frames, as shown in Fig. 2. Flow channels were carved into the flow frame to provide a steady electrolyte supply. Three flow frames with different numbers of channels were compared in terms of flow field

and electrochemical performance. The 3D model enables detailed analysis of cell performance and can act as a reference for design optimization. In the assembly and sealing of the cells, the channel depth in the flow frames will be lower than the thickness of the electrodes. The width of the channels and the position of the inlet were kept consistent for the three flow frames. The geometric parameters of the VRFB cell are listed in Table 1.

| Parameters          | Value | Unit |
|---------------------|-------|------|
| Cell height         | 300   | mm   |
| Cell width          | 600   | mm   |
| Channel depth       | 1.3   | mm   |
| Electrode thickness | 3.5   | mm   |
| Membrane thickness  | 50    | μm   |

Table 1 Geometric parameters of the VRFB cell



1-End plate 2-Current collector 3-Bipolar plate 4-Flow frame 5-Porous electrode 6-Membrane



Fig. 2 Schematic diagram of the components used in a VRFB cell

#### 2.3 Governing equations

#### 2.3.1 Fluid flow in the frames and electrodes

The flow of electrolyte is described by the Navier-Stokes equation:

$$\rho(\boldsymbol{u}\cdot\nabla)\boldsymbol{u} = -\nabla p + \mu\nabla^2\boldsymbol{u} \tag{4}$$

$$\nabla \cdot \boldsymbol{u} = 0 \tag{5}$$

where  $\boldsymbol{u}$  is the velocity of the electrolyte,  $\rho$  is the electrolyte density,  $\mu$  is the dynamic viscosity of the electrolyte, and p is the pressure of the liquid.

The flow of electrolyte in a porous electrode is described by Darcy's law:

$$\boldsymbol{u} = -\frac{K}{\mu} \nabla p \tag{6}$$

Where K is the permeability of the porous electrode. This permeability can be described by the Kozeny-Carman equation as follows:

$$K = \frac{d_{\rm f}^2 \varepsilon^3}{K_{\rm ck} (1 - \varepsilon)^2} \tag{7}$$

where  $K_{ck}$  is the Kozeny-Carman constant,  $d_f$  is the fiber diameter, and  $\varepsilon$  is the electrode porosity.

#### 2.3.2 Ions in the electrodes

The ions in the positive and negative electrodes are mainly comprised of vanadium ions  $(V^{2+}, V^{3+}, VO^{2+}, VO^{2+}_2, VO^{2+}_2)$  and ion products from sulfuric acid  $(H^+, HSO^-_4, SO^{2-}_4)$ . Each ion obeys the following conservation equation:

$$\frac{\partial}{\partial t} (\varepsilon c_j) + \nabla \cdot \left( -D_j^{\text{eff}} \nabla c_j - z_j \kappa_j F c_j \nabla \varphi_j + \boldsymbol{u} c_j \right) = -S_j \tag{8}$$

Here, *j* represents a particular ion,  $c_j$  is the concentration of the ion, and  $D_j^{eff}$  denotes the effective diffusion coefficient of the ion in the porous electrode;  $z_j$  and  $\kappa_j$  indicate the valence of the ion and the ionic mobility, respectively; *F* is the Faraday constant;  $\varphi_j$  denotes the ionic potential in the electrodes, and  $S_j$  indicates the source term of the ion.

Electrolyte solutions are electrically neutral, therefore:

$$\sum_{j} z_j c_j = 0 \tag{9}$$

The diffusion of ions in VRFBs occurs in porous electrodes, which differs from behavior in free space. The ideal ion diffusion coefficients are corrected with the Bruggeman equation:

$$D_i^{\rm eff} = \varepsilon^{1.5} D_j \tag{10}$$

where  $D_j$  denotes the diffusion coefficient of the ion in free space.

The source term describes how ion concentrations change in the electrolyte due to electrochemical reactions or the dissociation of sulfuric acid, as shown in Table 2 (here, *i* denotes the current density).

#### Table 2 The source terms of the conservation equation

| Source term          | Positive electrode               | Negative electrode   |
|----------------------|----------------------------------|--|
| $S_{V^{2+}}$         | -                                | $ abla \cdot \boldsymbol{i}/F$                               |
| $S_{V^{3+}}$         | -                                | $-\nabla \cdot \boldsymbol{i}/F$                             |
| $S_{V^{4+}}$         | $ abla \cdot \boldsymbol{i}/F$   | -  |
| $S_{V^{5+}}$         | $-\nabla \cdot \boldsymbol{i}/F$ | -  |
| $S_{\mathrm{H}^{+}}$ | $-S_{ m HSO_4^-}$                | $-2\nabla \cdot \boldsymbol{i}/F - S_{\mathrm{HSO}_{4}^{-}}$ |
| $S_{ m HSO_4^-}$     | $S_{\mathrm{HSO}_{4}^{-}}$       | $S_{\mathrm{HSO}_{4}^{-}}$                                   |

#### 2.3.3 Transport in the membrane

In a proton exchange membrane, it is assumed that protons are the only mobile ions. The current conservation equation can be expressed as follows:

$$\boldsymbol{N}_{\mathrm{H}^{+}} = -\frac{\sigma_{\mathrm{mem}}}{F} \nabla \phi_{\mathrm{mem}} \tag{11}$$

where  $\sigma_{mem}$  is the membrane conductivity,  $\phi_{mem}$  is the membrane potential, and  $N_{H^+}$  is the flux density vector of protons.

#### 2.3.4 Electrochemical kinetics

The Butler-Volmer model was used to describe the relationship between current density and overpotential during the electrochemical reactions:

$$J_{1} = A_{\rm S} F k_{1} (c_{\rm V^{4+}})^{(1-\alpha_{1})} (c_{\rm V^{5+}})^{\alpha_{1}} \left[ \frac{c_{\rm V^{4+}}^{s}}{c_{\rm V^{4+}}} \exp\left(\frac{(1-\alpha_{1})F\eta_{1}}{R_{\rm g}T}\right) - \frac{c_{\rm V^{5+}}^{s}}{c_{\rm V^{5+}}} \exp\left(-\frac{\alpha_{1}F\eta_{1}}{R_{\rm g}T}\right) \right]$$
(12)

$$J_{2} = A_{\rm S} F k_{2} (c_{\rm V^{2+}})^{(1-\alpha_{2})} (c_{\rm V^{3+}})^{\alpha_{2}} \left[ \frac{c_{\rm V^{2+}}^{s}}{c_{\rm V^{2+}}} \exp\left(\frac{(1-\alpha_{2})F\eta_{2}}{R_{\rm g}T}\right) - \frac{c_{\rm V^{3+}}^{s}}{c_{\rm V^{3+}}} \exp\left(-\frac{\alpha_{2}F\eta_{2}}{R_{\rm g}T}\right) \right]$$
(13)

Here,  $J_1$  and  $J_2$  are the transfer current densities of the positive and negative electrodes respectively;  $A_s$  is the specific surface area of the electrode;  $k_1$  and  $k_2$  are the reaction rate constants;  $\alpha_1$  and  $\alpha_2$  are the charge transfer coefficients; *s* represents the liquid-solid interfaces of the porous region;  $R_g$  is the universal gas constant; *T* is the temperature;  $\eta_1$  and  $\eta_2$  represent the activation overpotentials of the positive and negative reactions, respectively.  $\eta_1$  and  $\eta_2$  are defined as:

$$\eta_1 = \phi_s - \phi_l - E_1 \tag{14}$$

$$\eta_2 = \phi_{\rm s} - \phi_{\rm l} - E_2 \tag{15}$$

where  $\phi_s$  is the electrode potential,  $\phi_l$  is the electrolyte potential, and  $E_1$  and  $E_2$  are the standard equilibrium potentials for positive and negative redox reactions, respectively.

 $E_1$  and  $E_2$  are expressed by the Nernst equation:

$$E_1 = E_1^0 + \frac{R_g T}{F} \ln\left(\frac{c_{V^{5+}}(c_{H^+})^2}{c_{V^{4+}}}\right)$$
(16)

$$E_2 = E_2^0 + \frac{R_{\rm g}T}{F} \ln\left(\frac{c_{\rm V^{3+}}}{c_{\rm V^{2+}}}\right) \tag{17}$$

where  $E_1^0$  and  $E_2^0$  are the positive and negative standard potentials, respectively.

The cell voltage is calculated as follows:

$$E_{\text{cell}} = E_1^0 - E_2^0 + \frac{R_g T}{F} \ln\left(\frac{c_{\text{V}^2} + c_{\text{V}^5} + (c_{\text{H}^+})^2}{c_{\text{V}^3} + c_{\text{V}^4}}\right) - \eta_1 - \eta_2 - IR_{\text{cell}}$$
(18)

Here, I denotes the discharge current of the cell, and  $R_{cell}$  is the cell's resistance.

#### 2.4 Model parameters

Table 3 and Table 4 list the relevant parameters used in the 3D model of the VRFB cell.

| Parameters                        | Symbols                       | Value                  | Unit               | Reference          |
|-----------------------------------|-------------------------------|------------------------|--------------------|--------------------|
| Total vanadium ion concentration  | $c(V_{\rm total})$            | 1700                   | mol/m <sup>3</sup> | -                  |
| Initial proton concentration      | $c^{0}(H^{+})$                | 4000                   | mol/m <sup>3</sup> | -                  |
| $HSO_4^-$ ion concentration       | $c^{0}(\mathrm{HSO}_{4}^{-})$ | 4000                   | mol/m <sup>3</sup> | -                  |
| $V^{2+}$ diffusion coefficient    | $D_{V^{2+}}$                  | $2.4 \times 10^{-10}$  | m²/s               | (Lee et al., 2019) |
| $V^{3+}$ diffusion coefficient    | $D_{V^{3+}}$                  | $2.4 \times 10^{-10}$  | m²/s               | (Lee et al., 2019) |
| $V^{4+}$ diffusion coefficient    | $D_{V^{4+}}$                  | $3.9 \times 10^{-10}$  | m²/s               | (Lee et al., 2019) |
| $V^{5+}$ diffusion coefficient    | $D_{V^{5+}}$                  | $3.9 \times 10^{-10}$  | m²/s               | (Lee et al., 2019) |
| $H^+$ diffusion coefficient       | $D_{\mathrm{H}^+}$            | $9.312 \times 10^{-9}$ | m²/s               | (Lee et al., 2019) |
| $SO_4^{2-}$ diffusion coefficient | $D_{{ m SO}_{4}^{2-}}$        | $1.065 \times 10^{-9}$ | m²/s               | (Lee et al., 2019) |
| $HSO_4^-$ diffusion coefficient   | $D_{\mathrm{HSO}_{4}^{-}}$    | $1.33 \times 10^{-9}$  | m²/s               | (Lee et al., 2019) |
| Universal gas constant            | R <sub>g</sub>                | 8.3145                 | J/(mol<br>· K)     | -                  |
| Faraday constant                  | F                             | 96485                  | C/mol              | -                  |
| State of charge                   | SOC                           | 0.8                    | -                  | -                  |
| Kozeny-Carman constant            | K <sub>ck</sub>               | 9                      | -                  | -                  |
| Density                           | ρ                             | 1354                   | kg/m <sup>3</sup>  | (Lee et al., 2019) |

Table 3 Model parameters of the VRFB cell

| Parameters             | Symbols          | Value                  | Unit | Reference              |
|------------------------|------------------|------------------------|------|------------------------|
| Viscosity              | μ                | $4.928 \times 10^{-3}$ | Pa·s | (Lee et al., 2019)     |
| Specific surface area  | $A_{s}$          | $1.62 \times 10^{4}$   | 1/m  | (Lee et al., 2019)     |
| Carbon fiber diameter  | $d_{\mathrm{f}}$ | $1.76 \times 10^{-5}$  | m    | (Yuan et al.,<br>2020) |
| Electrode conductivity | $\sigma_{ m ed}$ | 1000                   | S/m  | (Lee et al., 2019)     |
| Porosity               | ε                | 0.90                   | -    | -                      |

| <b>Table 4</b> Parameters used in the simulation | on |
|--|----|
|--|----|

| Parameters                    | Symbols     | Value                | Unit | Reference          |
|-------------------------------|-------------|----------------------|------|--------------------|
| Positive rate constant        | $k_1$       | $6.8 \times 10^{-7}$ | m/s  | (Lee et al., 2019) |
| Negative rate constant        | $k_2$       | $1.7 \times 10^{-7}$ | m/s  | (Lee et al., 2019) |
| Positive transfer coefficient | $\alpha_1$  | 0.55                 | -    | (Lee et al., 2019) |
| Negative transfer coefficient | α2          | 0.45                 | -    | (Lee et al., 2019) |
| Positive standard potential   | $E_{1}^{0}$ | 1.004                | V    | (Lee et al., 2019) |
| Negative standard potential   | $E_{2}^{0}$ | -0.255               | V    | (Lee et al., 2019) |

#### 2.5 Boundary conditions

The structure of the flow frame has a critical impact on the performance of the VFRB cells. In this study, the VRFB performance was tested for three different flow frame structures, as illustrated in Fig. 3. The walls of both the flow frames and the porous electrodes were assumed to satisfy noslip boundary conditions. The positive and negative electrolyte flow rates were kept equal and ranged from 240-1080 mL/min. The mass flow inlet and outlet pressure boundary conditions were defined for the flow frames. The current density used in the discharge simulation was 105 mA/cm<sup>2</sup>. The system temperature was held at 293.15 K. The equations governing the 3D numerical model of the cell were solved using COMSOL Multiphysics® software.



Fig. 3 Flow frame designs for the numerical simulation

#### 2.6 Mesh independence analysis

Effective mesh division can improve the computational efficiency and accuracy of simulations. The 3D model of the VRFB was therefore investigated for mesh independence. The model was mainly divided into tetrahedral meshes, and the mesh was refined in corners and areas of complex flow, as shown in Fig. 4. To verify the independence between the simulation results and the number of meshes, four different mesh numbers were selected for comparative simulations. The mesh number increased proportionally from  $1.15 \times 10^6$  to  $1.22 \times 10^7$ , with the obtained results shown in Fig. 5. Considering both the computational cost and accuracy, a mesh number over  $6.20 \times 10^6$  was used for future comparison and discussion. We also compared structured and unstructured grids to validate the effectiveness of unstructured grids in this context. Fig. S1 shows the structured grid for Channel 1, and Fig. S2 compares the results of structured and unstructured grids. Both methods produce essentially the same results, showing that unstructured grids can flexibly and accurately treat complex geometries.



Fig. 4 Mesh refinement in the corners



Fig. 5 Grid independence analysis

#### 2.7 Multi-scale model of the VRFB

In the ECM of the VRFB, each cell is simplified to consist of a series-connected internal resistance and a controlled voltage source, where the internal resistance ( $R_{int}$ ) is comprised of the component resistance and the contact resistance between components. The voltage of the controlled source is obtained by linear interpolation of the cell model simulation results under different operating conditions. The resistance of the porous electrodes and the proton exchange membrane are calculated in the cell model, so  $R_{int}$  represents only the resistance of the bipolar plate between the cells. The ECM uses the Simscape block in *MATLAB* to construct a circuit model with parallel and series ohmic resistors connected to controlled voltage sources (represented as cells of the VRFB), and a constant current source (used to simulate constant current discharge).

Shunt currents are generated by the flow of electrolytes in the stack manifolds, which form paths between individual cells connected in series. The paths can be simplified to equivalent resistances in the ECM. These resistances are calculated using Eq. (19):

$$R = \frac{l}{\sigma A} \tag{19}$$

where l is the effective length, A is the cross-sectional area, and  $\sigma$  is the electrolyte conductivity.  $R_{pc_j}$ ,  $R_{nc_j}$ ,  $R_{pm_j}$ , and  $R_{nm_j}$  denote the equivalent resistances of the electrolyte in the positive channel, the negative channel, the positive manifold, and the negative manifold, respectively, and j represents the j<sup>th</sup> cell in the stack. The electrical conductivity  $\sigma$  depends on the SOC of the solution in each cell (Trovò et al., 2019):

$$\sigma_{\rm n} = \text{SOC} \cdot \sigma_{\rm V^{2+}} + (1 - \text{SOC}) \cdot \sigma_{\rm V^{3+}} \tag{20}$$

$$\sigma_{\rm p} = \text{SOC} \cdot \sigma_{\rm V^{5+}} + (1 - \text{SOC}) \cdot \sigma_{\rm V^{4+}} \tag{21}$$

where  $\sigma_n$  and  $\sigma_p$  is the catholyte and anolyte conductivity, respectively.

Additionally, species are exchanged between the stack and the tank during battery operation. The mass balance of the VRFB model is based on the following assumptions: (1) The electrolyte is completely mixed in the pipe, stack, and tank. (2) The charge loss during battery operation is caused only by the shunt current. The transport of species across the membrane is not considered. (3) Electrolyte parameters such as density, viscosity, and mobility remain constant during battery operation. (4) Flow rate differences between cells are not considered.

The ion concentration in the tanks is determined by the flow rate and the ion concentration at

the stack outlet. The concentration in the tanks is calculated using the following equation (Han et al., 2024):

$$\frac{\mathrm{d}c_i^{\mathrm{tank}}}{\mathrm{d}t} = \frac{Q}{V_{\mathrm{tank}}} \left( c_i^{\mathrm{stack}} - c_i^{\mathrm{tank}} \right) \tag{22}$$

where  $c_i^{\text{tank}}$  is the concentration of the valence vanadium ion *i* in the tank, *Q* is the flow rate of the stack,  $V_{\text{tank}}$  is the volume of electrolyte in the positive/negative tank, and  $c_i^{\text{stack}}$  is the ion concentration at the outlet of the stack.

The concentration in the cell is controlled by both the flow rate and the electrochemical reaction. Moreover, it is assumed that the concentration distribution in the cell varies linearly, which is expressed as follows:

$$\frac{\mathrm{d}c_i^{\mathrm{cellave}_j}}{\mathrm{d}t} = \frac{Q}{MV_{\mathrm{cell}}} \left( c_i^{\mathrm{tank}} - c_i^{\mathrm{cellout}_j} \right) \pm \frac{I_j}{zFV_{\mathrm{cell}}}$$
(23)

$$c_i^{\text{cellave}_j} = \left(c_i^{\text{cellin}_j} + c_i^{\text{cellout}_j}\right)/2 \tag{24}$$

$$c_i^{\text{stack}} = \sum_{j=1}^{M} c_i^{\text{cell}_j} / M$$
(25)

Here,  $c_i^{\text{cellin}_j}$ ,  $c_i^{\text{cellout}_j}$ , and  $c_i^{\text{cellave}_j}$  represent the average concentrations of the *i*-valent ions at the inlet, outlet, and the entire cell within the *j*<sup>th</sup> cell, respectively, *M* is the number of cells, *z* is the unit activity coefficient, *F* is Faraday's constant,  $V_{\text{cell}}$  is the volume of electrolyte in the cell, and  $I_j$  is the charge/discharge current that flows through the *j*<sup>th</sup> cell (I > 0 for charging). The sign of the value is determined by the species. When *i* equals 2 or 5, the value is positive; when *i* equals 3 or 4, the value is negative.

Therefore, the multi-scale model of the VRFB can be represented as shown in Fig. 6.  $I_{\rm L}$  is the total discharge current of the stack. The parameters used in the multi-scale model are shown in Table 5. The currents and voltages in the circuit are calculated using Kirchhoff's law.



Fig. 6 The multi-scale model of a VRFB with 40 cells Table 5 Parameters used in the multi-scale model of the VRFB

| Parameters                  | Value   | Unit           | Reference     | Meaning                                |
|-----------------------------|---------|----------------|---------------|--|
| D                           | 1 6E 7  | 0              |               | Bipolar plate resistance               |
| <i>k</i> <sub>int</sub>     | 1.0E-7  | 52             | -             | between cells                          |
| -                           | 27.5    | S /m           | (Moro et al., | W <sup>2+</sup> solution can destinity |
| 0 <sub>V<sup>2+</sup></sub> | 21.5    | 5/m            | 2017)         | v-* solution conductivity              |
| _                           | 175     | C /m           | (Moro et al., | ¥3+                                    |
| 0 V3+                       | 17.5    | 5/m            | 2017)         | ver solution conductivity              |
| _                           |         |                | (Moro et al., | 174+1_4;                               |
| $\sigma_{V^{4+}}$           | 21.1    | <b>S</b> /m    | 2017)         | v <sup>++</sup> solution conductivity  |
| -                           | 41.2    | C /m           | (Moro et al., | ¥75+                                   |
| 0 V2+                       | 41.3    | <b>S</b> /m    | 2017)         | v <sup>or</sup> solution conductivity  |
| 17                          | 0.75    | 3              |               | Electrolyte volume in the              |
| <i>V</i> tank               | 0.75    | III            | -             | tank                                   |
| 17                          | 0.00062 | 3              |               | Electrolyte volume in the              |
| <i>v</i> <sub>cell</sub>    | 0.00063 | III°           | -             | cell                                   |
| $l_{\rm m}$                 | 0.008   | m              | -             | Manifold length                        |
| A <sub>m</sub>              | 4.91E-4 | m <sup>2</sup> | -             | Manifold area                          |
| IL                          | 189     | А              | -             | Stack discharge current                |
|                             | 0.0     |                | (Ali et al.,  | D                                      |
| $arphi_{	ext{pump}}$        | 0.9     | -              | 2020)         | rump eniciency                         |

#### 3 Results and discussion

A major goal of this study is to analyze how the flow frame structure affects the VRFB performance. A properly distributed flow field in the porous electrodes of a VRFB cell can improve the battery performance. Hence, battery performance across different flow field distributions was investigated in terms of discharge voltage, pressure drop, and vanadium concentration. Finally, the effect of flow frame structure on system efficiency was analyzed using the multi-scale model of the VRFB. Given that mass transfer and electrochemical reactions in the positive and negative electrodes are essentially similar, only the negative electrode was used for comparative analysis.

#### 3.1 Model validation

To validate the accuracy of the cell model, the discharge data from a single cell with an electrode size of 600 mm x 300 mm was tested. The initial and final vanadium ion concentrations were quantified by potentiometric titration to determine the corresponding state of charge (SOC). Constant current density discharge tests were conducted on the single cell experimental platform (shown in Fig. 7). During the experiment, the electrolyte flow rate was set to 1080 mL/min, and the applied current density was 105 mA/cm<sup>2</sup>, which is consistent with the conditions in the numerical simulation. The VRFB discharge cutoff voltage was 1.1 V.

Fig. 8 compares the simulation and experimental results for the discharge voltage at different states of charge. The results indicate that the numerical model accurately reflects the behavior of the battery during discharge. In the low state of charge region, the discrepancy between the numerical results and the experimental results is more obvious. This is because the cell discharge voltage decreased rapidly at the end of discharge, which increased the error in estimating the reactant concentration during the discharge process.



Fig. 7 Test platform for a cell of the VRFB



Fig. 8 Comparison of the numerical and experimental results

#### 3.2 Velocity and pressure drop

The velocity distributions within the electrodes are significantly influenced by the configuration of the flow frames, as illustrated in Fig. 9. A large area of high values is clearly observed for Channel 1, which is located in the middle of the electrode. In contrast, high value areas for Channels 5 and 20 are only located near the top and bottom of the electrodes.

Fig. 10 depicts the effect of flow rate on velocity distribution. Over Section 1, velocities along a red line which is offset 30 mm from the top of the electrode (Line 1) were chosen for comparison. As expected, velocity differences increase with increasing flow rates for all flow frames. Interestingly, the number of velocity peaks is the same as the number of channels for Channels 1 and 5, coinciding with the velocity contour shown in Fig. 9. As channel numbers continue to increase, velocity peaks transform into velocity plateaus at Channel 20. A similar phenomenon can be observed along other lines, as evidenced by the velocity changes along lines 150 mm (Line 2) and 270 mm (Line 3) offset from the top of the electrode. Meanwhile, the velocity fluctuates relatively gently along Line 2. This is mainly due to the significant resistance exerted on the fluid by the porous electrode. According to Darcy's law, the higher the fluid velocity, the greater the resistance it encounters. This causes the fluid in regions with higher velocity to diffuse towards adjacent regions, leading to a more uniform velocity distribution. Additionally, we assessed velocity fluctuations by dividing the standard deviation of the velocity by its mean value. Along Line 1 and Line 3, Channels 5 and 20 show similar values. But along Line 2, Channel 5 has a more uniform velocity than Channel 20, implying a more even distribution of concentration.

Another key performance parameter for the cell is the pressure drop, which affects the amount of pump consumption during battery operation. An excessive pressure drop will reduce the system efficiency during the charge/discharge cycle, and also necessitate stronger mechanical properties for the components, which may increase manufacturing costs. Fig. 11 presents a comparison of the total pressure drops of different flow frames, at flow rates ranging from 240-1080 mL/min. An approximate positive linear relationship between total pressure drop and flow rate can be observed. The total pressure drop for Channel 1 reaches  $5.3 \times 10^4$  Pa at a flow rate of 1080 mL/min, while the pressure drops for Channels 5 and 20 are relatively low. This may be because Darcy's law governs fluid flow in porous electrodes. The pressure drop in the electrodes is further illustrated in Fig. S3. A high value for Channel 1 is obvious, and Channel 5 and Channel 20 have almost the same values. Note that the total pressure drop of Channel 5 is higher than that of Channel 20, implying that the differences in the flow frames are responsible for this effect. Indeed, Channel 20 has a shorter electrolyte path in its flow frame compared to Channel 5, as shown in Fig. S4.



Fig. 9 The distribution of velocity (Section 1) at the negative electrode for all three flow frames (Q = 600 mL/min). (a) Channel 1, (b) Channel 5, (c) Channel 20









(c) Channel 20

Fig. 10 Velocity change along the red line for different flow frames (Section 1): (a) Channel 1, (b)





Fig. 11 Total pressure drop for different flow frames across various electrolyte flow rates

#### 3.3 Discharge voltage and V<sup>2+</sup> concentration distribution

Discharge voltage is an important factor in evaluating battery performance. Therefore, we compare the discharge voltage for the three flow frame structures across different flow rates. In the numerical simulations, the SOC is kept between 0.2-0.8 and the discharge current density is set at 105 mA/cm<sup>2</sup>. The cut-off voltage was set to 1.1 V. The flow rates of catholyte and anolyte were equivalent, and were increased from 240 mL/min to 1080 mL/min. The discharge voltage of batteries with different flow frames across varying flow rates and SOCs is shown in Fig. 12. With an increasing flow rate, the discharge voltages of all three flow frames rise. This is because the higher flow rate can increase the concentrations of reactants in the electrodes. However when the reactant concentrations become sufficient, this effect becomes weaker; also, the voltage difference between the three flow frames decreases. Thus, the electrolyte flow rate is the main factor influencing the discharge process of the VRFB. The Channel 5 structure has a higher discharge voltage than the other structures, especially at low flow rates and SOCs. Interestingly, this contrasts with the speculation that more channels would lead to a more even distribution of velocity and a higher discharge voltage.



Fig. 12 3D scatter plot of discharge voltage for the three flow frames at varying flow rates and SOCs

We aim to explain this phenomenon by investigating the reactant concentration distribution in the negative electrode. In Fig. 13, the concentration distributions of  $V^{2+}$  in Sections 1, 2, and 3 are shown when the flow rate is 360 mL/min and the SOC is 0.3. Compared to Channel 5 and Channel

20, Channel 1 shows a large low-concentration region of  $V^{2+}$ , which would significantly hinder battery performance. For Channels 5 and 20, the  $V^{2+}$  concentration distributions are similar, with obvious high values near the diagonal lines. This phenomenon can be explained by the direction of the pressure gradient in the porous electrode. As depicted in Fig. S5, the pressure is high near the inlet and low near the outlet; this pressure gradient causes the fluid to flow from the inlet towards outlet. Accordingly,  $V^{2+}$  ions are transported along with the fluid, and follow the trend of the diagonal line.

To evaluate the reactant concentrations, total values were calculated and are shown in Fig. 13. The values over different sections follow the same order, that is, Channel 5 > Channel 20 > Channel 1. Moreover, this trend persists with increasing flow rates, as shown in Fig. S6. The superior value for Channel 5 may be a consequence of the more uniform velocity distribution. As mentioned above, along different lines for Channel 5, the velocity fluctuation values  $(\frac{\sigma_u}{u_{average}})$  are 0.24, 0.013, and 0.25, which are always lower than their counterparts in Channel 20. Thus, the unique design of Channel 5 promotes a higher reactant concentration within the electrode, leading to lower concentration polarization and higher discharge voltage.



Fig. 13  $V^{2+}$  concentration distribution and total values at different sections: (a) Channel 1, (b) Channel 5, and (c) Channel 20

#### 3.4 Flow frame impact on system efficiency

Efficiency is a crucial parameter for evaluating the performance of a battery system. In the operation of a VRFB, increasing the electrolyte flow rate can enhance the stack voltage, but also leads to higher pump power consumption. Therefore, the selected flow rate needs to balance the electrochemical reaction and the pump power consumption. We use the system efficiency to comprehensively assess the impact of flow rate on these two aspects. To ascertain the optimal power

efficiency using different cell structures, a 10kW/40kWh VRFB model with 40 cells was built. Then, the pump power of the entire stack and the power loss due to overpotential were investigated. The VRFB system efficiency ( $\psi_{power}$ ) was calculated as follows:

$$\psi_{\text{power}} = \frac{P_{\text{net}}}{P_{\text{total}}} = 1 - \frac{P_{\text{loss}} + P_{\text{pump}}}{P_{\text{total}}}$$
(26)

$$P_{\rm loss} = \sum_{j=1}^{M} I_j A \left( \eta_{\rm p_j} + \eta_{\rm n_j} \right) + P_{\rm R}$$
(27)

$$P_{\text{pump}} = \frac{2(\Delta P_{\text{cell}} + \Delta P_{\text{G}}) \cdot Q}{\psi_{\text{pump}}}$$
(28)

where  $\psi_{power}$  is the VRFB system efficiency,  $\psi_{pump}$  is the pump efficiency,  $P_{net}$  is the output power,  $P_{total}$  is the total input power,  $P_{pump}$  is the pump power consumption,  $P_{loss}$  is the stack power loss,  $P_R$  is the stack ohmic power loss,  $\Delta P_{cell}$  is the pressure drop in the cells, and  $\Delta P_G$  is the gravitational potential difference between the tank and the stack that needs to be overcome.

The system efficiency of each flow frame at varying flow rates can be obtained from the VRFB model, as shown in Fig. 14. As the flow rate increases, the system efficiency of each flow frame gradually rises, and the gap between the efficiency of Channel 5 and Channel 20 gradually disappears. After the flow rate reaches 960 mL/min, increasing the flow rate has little effect on the system efficiency. A peak efficiency of 93.51% was observed at the electrolyte flow rate of 1080 mL/min for Channel 5 and Channel 20.



**Fig. 14** System efficiency for various flow frame structures at differing flow rates From the ECM model, it was ascertained that an important parameter affecting the efficiency

of the stack is the resistance of each cell. Among these resistances,  $R_{pm}$  and  $R_{nm}$  are determined by the thickness of the porous electrodes and bipolar plates; unfortunately, these are difficult to change.  $R_{pc}$  and  $R_{nc}$  can be increased by extending the lengths of the inlet/outlet pipes and shirking the pipe cross-section in the cell, so as to reduce the shunt current. However, this also increases the pressure drop as well as the pump consumption of the stack. Therefore we decide to use the genetic algorithm (GA) to optimize the parameters such as pipe length and cross-section width. Table 6 lists the optimization parameter ranges, and more detailed information on the GA workflow can be found in the Supporting Information. The upper limit of the flow rate is based on the pump's rated power, while the lower limit is set such that the system can operate normally without damaging the electrolyte. The maximum values for pipe length and cross-section width are constrained by the manufacturing precision. Our minimization objective (or objective function) is the VRFB's system loss efficiency in the discharging process.

Table 6 Optimization parameters and ranges

| Parameters  | range      | Unit   |
|---|------------|--------|
| Pipe length of the positive side $(L_p)$              | 0 - 3      | m      |
| Pipe length of the negative side $(L_n)$              | 0 - 3      | m      |
| Pipe cross-section width of the positive side $(W_p)$ | 10 - 60    | mm     |
| Pipe cross-section width of the negative side $(W_n)$ | 10 - 60    | mm     |
| Flow rate $(Q)$                                       | 360 - 1080 | mL/min |

The optimal solutions obtained are shown in Table 7. Each solution includes the optimal sizes and the corresponding system efficiency of the discharge process. The highest efficiency of 93.70% was observed for Channel 5 at an electrolyte flow rate of 960 mL/min.

| <b>Table</b> / Optimization results |
|-------------------------------------|
|-------------------------------------|

| Channel | $L_{n}(m)$ | <i>L</i> <sub>p</sub> (m) | $W_{n}(mm)$ | W <sub>p</sub> (mm) | Q (mL/min) | $oldsymbol{\psi}_{	extsf{power}}$ |
|---------|------------|---------------------------|-------------|---------------------|------------|-----------------------------------|
| 1       | 1.263      | 1.708                     | 58          | 59                  | 1080       | 92.65%                            |
| 5       | 1.555      | 1.697                     | 60          | 52                  | 960        | 93.70%                            |
| 20      | 1.141      | 1.709                     | 45          | 53                  | 977        | 93.63%                            |

#### 4 Conclusions

The performances of three different flow frame structures in a vanadium redox flow battery

(VRFB) were evaluated at both the cell and stack level. To analyze the flow field and species concentration distribution at the cell level, a 3D model of the VFRB cells was developed. With this model, the cell performance was compared across different flow rates and states of charge (SOC), investigating factors such as pressure drop, velocity distribution, reactant concentration distribution, and discharge voltage. The results showed that the pressure drop of the cell decreases gradually with increasing numbers of channels in the flow frame. The cell with a five-channel flow frame was superior to the other configurations at low reactant concentration. We also found that despite a high flow rate enabling satisfactory discharge voltage, it also causes extra pumping consumption. Next, we proposed an equivalent circuit model (ECM) of the stack to simulate the operation of the battery. The system efficiency of the discharge process was used to evaluate different cell structures. The voltage differences between cells and the conductivity of the electrolyte generates shunt currents within the stack and piping system, which lead to energy losses and reduced system efficiency. The cell parameters affecting the shunt current in the discharge process were therefore optimized using a genetic algorithm. The final optimized configuration, using a five-channel flow frame, achieved a maximum system efficiency of 93.70% at a flow rate of 960 mL/min.

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