附件	2 浙江水	、利水电学院	"南浔青年学	者"申请表	
姓名	李昊文	出生年月年龄 (年龄)	<u>1994</u> _年 <u>7</u> 月岁 ( 29 岁)	参加工作时间	<u>2023</u> 年 <u>4</u> 月
现所在单位 (部门)	机械与汽车工程学 院	最高学历/学位	博士研究生/博士	专业技术职务	讲师
符合条款		例:符合教学类,	教学成果奖,省级,	二等奖, 绩点 200。	
	教学类 绩点: <u>0</u>				
所涉业绩	科研类 绩点: <u>170</u>	科研类,3 类论文3 1. Haowen Li,Zh modules based o 244,123218.(S 2. Haowen Li,Zh supercapacitor fa (SCI,3类论文 3. Haowen Li,L characteristics o validation. Appli 文) 4. Haowen Li,Z responsive actua analysis. Journa 14(12):125001.	a 篇, 绩点 150; 科研 eng Bo, et al. Entro on a three-dimensiona <b>CI</b> , 3类论文) eng Bo, et al. Ener or different operating ( <i>J</i> ) ijun Wu, et al. N f the plate air heater <b>ied Thermal Engineer</b> heng Bo, et al. Ex ator based on metal <i>al of Thermal Scient</i> (SCI, 5类论文)	f类, 5 类论文 1 篇, ppy generation analy al coupled thermal n gy and entropy gene conditions. <i>Energy</i> , umerical investigation with variable chann <i>ring</i> , 2017, 122, 546 perimental investigation <i>ce and Engineering</i>	绩点 20: sis in supercapacitor nodel. <i>Energy</i> , 2022, eration analysis in a 2022, 260, 124932. on on heat transfer els and experimental - 554. (SCI, 3类论 tion of a thermally ulfide: a conceptual <i>Applications</i> , 2022,
	八才林亏矣 绩点:				
	总绩点		17	70	
	备注:请严格按照《浙江水利水电学院"南浔青年学者"实施办法(试行)》(浙水院〔2022〕114 号)附件《业绩成果绩点赋值项目库》的类目、分类、等级(等次)、绩点填写。				
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个人承诺			签名: 苍臭>	时间: 2	2023.11.15
	以上部分由申请	人填写,所在单位	审核。以下由单位(	部门)和学校填写。	
所在单位 (部门)意见	1.经审核,申请人所 2.是否符合所申请的 3.是否同意推荐:[	所填内容: □属实 り类别: □符合 □ 〕同意 □不同意。	□不属实; □不符合;	负责人签名: 年	(部门盖章) 月日
科技处 审核意见	负责人签名:	(部门盖章) _年月日	教务处 审核意见	负责人签名:	(部门盖章) 年月日

其他相关职能	负责人签名: (部门盖章)
部门审核意见	年月日
学校意见	(学校盖章) 年月日

备注:表格请用 A4 纸打印,有关佐证材料附后。

经检索《Web of Science》和《Journal Citation Reports (JCR)》数据库,《Science Citation Index Expanded (SCI-EXPANDED)》收录论文及其期刊影响因子、分区情况如下。(检索时间: 2022 年 9 月 19 日)

# 第1条,共4条

标题:Entropy generation analysis in supercapacitor modules based on a three-dimensional coupled thermal model

作者:Li, HW(Li, Haowen);Yang, HC(Yang, Huachao);Xu, CX(Xu, Chenxuan);Yan, JH(Yan,

Jianhua);Cen, KF(Cen, Kefa);Ostrikov, K(Ostrikov, Kostya (Ken));Bo, Z(Bo, Zheng);

来源出版物:ENERGY 卷:244 文献号:123218 DOI:10.1016/j.energy.2022.123218 出版年:APR 1 2022 入藏号:WOS:000792041500003

文献类型:Article

地址:

[Li, Haowen; Yang, Huachao; Xu, Chenxuan; Yan, Jianhua; Cen, Kefa; Ostrikov, Kostya (Ken); Bo, Zheng] Zhejiang Univ, Coll Energy Engn, State Key Lab Clean Energy Utilizat, Hangzhou 310027, Zhejiang, Peoples R China.

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709

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期刊《ENERGY》2021年的影响因子为8.857,五年影响因子为8.234。 期刊《ENERGY》2021年的JCR分区情况为:

Edition	JCR® 类别	类别中的排序	JCR 分区
SCI	ENERGY & FUELS	24/119	Q1
SCI	THERMODYNAMICS	3/63	Q1

# 第2条,共4条

标题:Energy and entropy generation analysis in a supercapacitor for different operating conditions 作者:Li, HW(Li, Haowen);Yang, HC(Yang, Huachao);Yan, JH(Yan, Jianhua);Cen, KF(Cen, Kefa);Ostrikov, K(Ostrikov, Kostya (Ken));Bo, Z(Bo, Zheng);

来源出版物:ENERGY 卷:260 文献号:124932 DOI:10.1016/j.energy.2022.124932 出版年:DEC 1 2022

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文献类型:Article

地址:

[Li, Haowen; Yang, Huachao; Yan, Jianhua; Cen, Kefa; Ostrikov, Kostya (Ken); Bo, Zheng] Zhejiang Univ, Coll Energy Engn, State Key Lab Clean Energy Utilizat, Hangzhou 310027, Zhejiang, Peoples R China.

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24/119	Q1
3/63	Q1
	24/119

# 第3条,共4条

标题:Combinatorial atomistic-to-AI prediction and experimental validation of heating effects in 350 F supercapacitor modules

作者:Bo, Z(Bo, Zheng);Li, HW(Li, Haowen);Yang, HC(Yang, Huachao);Li, CW(Li, Changwen);Wu, SH(Wu, Shenghao);Xu, CX(Xu, Chenxuan);Xiong, GP(Xiong, Guoping);Mariotti, D(Mariotti, Davide);Yan, JH(Yan, Jianhua);Cen, KF(Cen, Kefa);Ostrikov, K((Ken) Ostrikov, Kostya);

来源出版物:INTERNATIONAL JOURNAL OF HEAT AND MASS TRANSFER 卷:171 文献 号:121075 提前访问日期:FEB 2021 DOI:10.1016/j.ijheatmasstransfer.2021.121075 出版年:JUN 2021 入藏号:WOS:000641140800025

文献类型:Article

地址:

[Bo, Zheng; Li, Haowen; Yang, Huachao; Li, Changwen; Wu, Shenghao; Xu, Chenxuan; Yan, Jianhua; Cen, Kefa; (Ken) Ostrikov, Kostya] Zhejiang Univ, Coll Energy Engn, State Key Lab Clean Energy Utilizat, Hangzhou 310027, Zhejiang, Peoples R China.

[Bo, Zheng; Li, Haowen; Yang, Huachao; Li, Changwen; Wu, Shenghao; Xu, Chenxuan; Yan, Jianhua; Cen, Kefa] ZJU Hangzhou Global Sci & Technol Innovat Ctr, Hangzhou 311215, Zhejiang, Peoples R China.

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IDS 号:RO6GH

ISSN:0017-9310

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# 期刊《INT J HEAT MASS TRAN》2021年的影响因子为 5.431,五年影响因子为 5.428。 期刊《INT J HEAT MASS TRAN》2021年的 JCR 分区情况为:

Edition	JCR® 类别	类别中的排序	JCR 分区
SCI	ENGINEERING, MECHANICAL	17/137	Q1
SCI	MECHANICS	15/138	1:01
SCI	THERMODYNAMICS	9/63	Q1

MIL

# 第4条,共4条

标题:Numerical investigation on heat transfer characteristics of the plate air heater with variable channels and experimental validation 209

作者:Li, HW(Li, Haowen);Wu, LJ(Wu, Lijun);Yuan, ZC(Yuan, Zhicheng);Yuan, ZK(Yuan, Zhaokuo); 来源出版物:APPLIED THERMAL ENGINEERING 卷:122 页:546-554

DOI:10.1016/j.applthermaleng.2017.03.136 出版年:JUL 25 2017

入藏号:WOS:000403855200051

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地址:

[Li, Haowen; Wu, Lijun; Yuan, Zhicheng; Yuan, Zhaokuo] Tongji Univ, Sch Mech Engn, Shanghai 201804, Peoples R China.

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# 《SCI-EXPANDED》收录及《JCR》期刊影响因子、分区情况证明

期刊《APPL THERM ENG》2021 年的影响因子为 6.465,五年影响因子为 5.906。 期刊《APPL THERM ENG》2021 年的 JCR 分区情况为:

Edition	JCR® 类别	类别中的排序	JCR 分区
SCI	ENERGY & FUELS	43/119	Q2
SCI	ENGINEERING, MECHANICAL	10/137	Q1
SCI	MECHANICS	11/138	Q1
SCI	THERMODYNAMICS	6/63	Q1

注:

1. 期刊影响因子及分区情况最新数据以 JCR 数据库最新数据为准。

2. 以上检索结果来自 CALIS 查收查引系统。

3. 以上检索结果均得到委托人及被检索作者的确认。



# 《SCI-EXPANDED》收录及《JCR》期刊影响因子、分区情况证明

经检索《Web of Science》和《Journal Citation Reports (JCR)》数据库,《Science Citation Index Expanded (SCI-EXPANDED)》收录论文及其期刊影响因子、分区情况如 下。(检索时间: 2022年12月15日)

第1条,共1条

标题:Experimental Investigation of a Thermally Responsive Actuator Based on Metallic Molybdenum Disulfide: A Conceptual Analysis

作者:Li, HW(Li, Haowen);Yang, HC(Yang, Huachao);Xu, CX(Xu, Chenxuan);Cheng, XN(Cheng, Xiangnan);Yan, JH(Yan, Jianhua);Cen, KF(Cen, Kefa);Bo, Z(Bo, Zheng);Ostrikov, K(Ostrikov, Kostya

(Ken)); 来源出版物:JOURNAL OF THERMAL SCIENCE AND ENGINEERING APPLICATIONS 卷:14 期:12 文献号:125001 DOI:10.1115/1.4055152 出版年:DEC 1 2022

入藏号:WOS:000878271300008

文献类型:Article

地址:

[Li, Haowen; Yang, Huachao; Xu, Chenxuan; Cheng, Xiangnan; Yan, Jianhua; Cen, Kefa; Bo, Zheng;
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期刊《JTHERM SCIENG APPL》2021年的影响因子为1.879,五年影响因子为1.754。

# 期刊《JTHERM SCI ENG APPL》2021年的 JCR 分区情况为:



Contents lists available at ScienceDirect

# Energy

journal homepage: www.elsevier.com/locate/energy

# Energy and entropy generation analysis in a supercapacitor for different operating conditions



<sup>a</sup> State Key Laboratory of Clean Energy Utilization, College of Energy Engineering, Zhejiang University, Hangzhou, Zhejiang Province, 310027, People's Republic of China <sup>b</sup> School of Chemistry and Physics and QUT Centre for Materials Science, Queensland University of Technology (QUT), Brisbane, Queensland, 4000, Australia

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Keywords: Entropy generation 3D coupled thermal model Thermodynamic analysis Supercapacitor

#### ABSTRACT

Sustainability and efficiency are the key issues concerning performance and lifetime of supercapacitors. In this work, an entropy generation analysis is implemented for the first time in the supercapacitor cell, aiming at facilitating the design and optimization of the supercapacitor systems. Entropy generation analysis accurately quantifies the irreversibilities due to heat transfer, mass transfer and ohmic loss of the supercapacitor cell, satisfying direct identification of the inefficiency mechanisms that cannot be achieved by the conventional energy analysis. The effects of the electrolyte, porosity and charge/discharge current on the thermodynamic irreversibilities and heat transfer characteristics are investigated. The optimal design scheme of the supercapacitor cell is obtained using the optimization approaches based on the combined energy and entropy generation analyses. Results indicate that the main contribution to the irreversibilities is due to ohmic loss, followed by the mass transfer effect. The 1 M TEMABF<sub>4</sub>/ACN electrolyte with the porosity of 0.4 is found to be the optimal choice, corresponding to the entropy generation rate of 2230 W/(m<sup>3</sup>·K) and the temperature rise of 0.20974 °C. The outcomes of this work provide a fundamental and computational framework for the development and optimization of clean and renewable energy conversion and storage systems with reduced irreversibilities.

#### 1. Introduction

Supercapacitors, a fast-rising class of energy storage devices based on rapid ion adsorption and separation at the solid/liquid interface, have shown prominent merits in terms of ultrahigh rate capability, high power density (>10 kW/kg), and wide operating temperature window  $(-40 \ ^{\circ}\text{C} \sim 70 \ ^{\circ}\text{C})$  [1,2]. These particularly attractive features make supercapacitors commonly envisaged as a promising candidate for high-power demanding applications, such as hybrid electrical vehicles, uninterruptable power supply, emergency power support and braking energy regeneration [3,4]. Considering the key roles of the rate capabilities of the supercapacitor in these cases, continuous endeavors have been devoted towards the improvement of power densities of supercapacitors. For instance, the rate capability of supercapacitors can be significantly enhanced by developing advanced active materials and electrolyte fluids to accelerate the transportation of ions and electrons [5–8]. However, the role exerted by the cycle life and sustainability in implementing long-term development of supercapacitors has not received the attention it deserves.

It is well known that the service life of supercapacitors is mainly

affected by two aspects: temperature and irreversibilities. Temperature rise or even overheating issues are accompanied by electrolyte solvent evaporation [9] and separator decomposition [10], leading to unsatisfactory reliability and deteriorated service life of supercapacitors [11]. For example, the cycle life of a supercapacitor is halved for every 10  $^\circ\text{C}$ increment above the temperature of 25 °C [12]. Thus, the efficient thermal management schemes are highly desirable to reduce the temperature rise. Recently, extensive research has focus on developing the effective thermal management approaches based on air-cooling [13], liquid-cooling [14], and phase change materials (PCMs)-based cooling [15]. For example, Voicu et al. [16,17] adopted forced air-cooling modules to manage the temperature distribution in a supercapacitor stack for different geometries. Zhang et al. [18] investigated the effect of external airflow on air distribution in the supercapacitor module compartment designed for an electric bus and optimized the air-cooling system. Further to active cooling methods, passive techniques based on PCMs, such as paraffin waxes [19,20] and porous materials foams [21], were also implemented to prevent the undesired temperature increases. However, additional energy consumption costs and low thermal conductivity had limited the successful implementations on the thermal

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management of supercapacitors. And the proposed approaches were mainly focused on the methods to enhance the heat dissipation between the supercapacitor and cooling medium. Therefore, there is an increasing need for the design of supercapacitors with smaller temperature rise in the absence of thermal management approaches, in other words, with lower heat generation during the charge/discharge cycles.

According to the first-law of thermodynamics, for the energy conversion and storage systems, the input power is converted into the stored electrical energy and the dissipation. And the irreversibilities of the system are equivalent to their corresponding dissipation [22]. Therefore, the reduction of irreversibilities results in a more efficient and energy efficient system. Entropy generation according to the second law of thermodynamics determines the mechanisms of thermodynamic irreversibilities and quantifies the decrease of the available work [23-25]. Thus, besides the energy analysis, the entropy generation analysis has become a very popular tool to study and design efficient energy conversion and storage systems. For example, Sergio et al. [26] developed an entropy generation analysis of the performance of a fuel cell using different configurations to find a geometry that provided a good distribution of species for fuel cells. Ramírez-Minguela et al. [26] performed an entropy generation analysis in the solid oxide fuel cell to evaluate the effects of four different biogas compositions in the thermodynamic irreversibilities. Giangaspero et al. proposed an entropy generation minimization method to optimize the forced convective cooling system of LED-based spotlight [27] and to optimize the solar heat exchanger configurations [28]. However, although some entropy generation studies are implemented in the literature [29-33], the entropy generation analysis related to the supercapacitor cell accounting for the thermodynamic irreversibilities, to the best of our knowledge, has never been reported yet.

In this work, an entropy generation analysis based on a new multiscale coupled model is conducted for the first time to accurately quantify the heat transfer irreversibility, the mass transfer irreversibility and the ohmic loss irreversibility of the supercapacitor cell. The coupled model consists of a one dimensional (1D) electrochemical model, a three dimensional (3D) thermal model and a local entropy generation rate model, taking into account a complete solution of the electrochemical, heat generation, heat transfer and entropy generation rate equations. In particular, the entropy generation mechanisms associated with heat transfer, mass transfer and ohmic loss are investigated and the main irreversibility occurring inside the supercapacitor cell is revealed. Then, the effects of the electrolyte, porosity and charge/discharge current on the thermodynamic irreversibilities and heat transfer characteristics of the supercapacitor are analyzed. Finally, the entropy generation analysis and energy analysis are combined to make the multi-attribute decision on the optimization of the supercapacitor system with lower irreversibilities and temperature rise. The outcomes of this work can pave the way for designers to develop more efficient supercapacitors and other energy conversion and storage systems.

#### 2. Mathematical model

#### 2.1. Model description and assumptions

The modeling approach is based on a 1D electrochemical model, a 3D thermal model and a local entropy generation rate model. The computational domain of the supercapacitor is shown in Fig. 1. The electrochemical model is composed of a negative electrode, a positive electrode, electrolyte, and a separator (Fig. 1(a)). The thermal model computational domain is shown in Fig. 1(b), which is composed of the air core, the active zoon and the aluminum shell.

To simplify the mathematical analysis, the following assumptions are made [34]: (1) the porous electrode theory is applied; (2) the electrolyte is completely infiltrated in porous electrodes and separator; (3) the Faradaic reactions are neglected; (4) self-discharge and charge effects are negligible; (5) the heat generation is uniform because the properties



Fig. 1. Illustration of (a) 1D electrochemical model and (b) 3D thermal model.

of materials are isotropic; (6) thermal interface resistances are neglected; (7) the cover plate is adiabatic; (8) the computational domains of both electrochemical model and thermal model are completely symmetrical.

#### 2.2. Governing equations

#### 2.2.1. 1D electrochemical model

The charge conservation equation can be obtained based on the concept of current continuity:

$$\frac{\partial i_{s}(x,t)}{\partial x} + \frac{\partial i_{1}(x,t)}{\partial x} = 0$$
(1)

$$\frac{\partial i_{1}(x,t)}{\partial x} = \delta C_{\rm dl} \frac{\partial (\varphi_{\rm s}(x,t) - \varphi_{\rm l}(x,t))}{\partial t}$$
(2)

where  $i_s$  and  $i_l$  are the solid phase current density and liquid phase current density, respectively,  $\varphi_s$  and  $\varphi_l$  are the solid electrical potential and liquid electrical potential, respectively,  $C_{dl}$  is the capacitance per interfacial area, and  $\delta$  is the surface area per unit volume of electrode.

The solid current density in the electrodes follows the Ohm's Law:

$$i_{s}(x,t) = -\sigma_{\text{elec}} \frac{\partial \varphi_{s}(x,t)}{\partial x}$$
(3)

where  $\sigma_{\text{elec}}$  is the solid phase conductivity.

The liquid current density can be described by:

$$i_1(x,t) = F \sum_{i=1}^{n} e_i N_i(x,t)$$
 (4)

where *F* is the Faraday's constant,  $e_i$  is the charge,  $N_i$  is the flux of ions. The flux of ions  $N_i$  of the supercapacitor can be expressed as:

$$N_i(x,t) = N_{i,\text{trans}}(x,t) + N_{i,\text{diff}}(x,t) = -\frac{e_i F}{RT} D_i c_i \frac{\partial \varphi_1(x,t)}{\partial x} - D_i \frac{\partial c_i(x,t)}{\partial x}$$
(5)

where *R* is the universal gas constant,  $c_i$  is the ion concentration,  $D_i$  is the diffusion coefficient,  $u_i$  is the migration rate, and for a binary electrolyte i = 1, 2.

Considering the porosity of electrodes and separator, the electrode conductivity and the electrolyte diffusion coefficient within the electrodes and separator can be modified:

$$\sigma_{\text{elec}} = \sigma_{s} (1 - \varepsilon_{\text{elec}})^{\beta}$$

$$D_{i,\text{elec}} = D_{i} \cdot \varepsilon_{\text{elec}}^{\beta}$$

$$D_{i,\text{sep}} = D_{i} \cdot \varepsilon_{\text{sep}}^{\beta}$$
(6)

where  $\sigma$ s is the solid phase conductivity,  $\beta$  is the Bruggeman porosity exponent,  $\varepsilon_{\text{elec}}$  is the porosity of the electrodes, and  $\varepsilon_{\text{sep}}$  is the porosity of the separator.

The mass conservation is described as follows:

$$\frac{\partial c_i(x,t)}{\partial t} = -\frac{\partial N_i(x,t)}{\partial x} + R_i$$
(7)

where  $R_i$  is the source term (here  $R_i = 0$  without chemical reactions).

#### 2.2.2. 3D thermal model

Considering the electrolyte is completely infiltrated in electrodes and separator with no fluid flow, thermal conduction is the dominant heat transfer mechanism of the supercapacitor. In view of its symmetrical structure in 3D cylindrical coordinates, the transient equation can be written as:

$$\frac{\partial \left(\rho C_{\rm p} T(r,z,t)\right)}{\partial t} = \lambda_r \frac{\partial^2 T(r,z,t)}{\partial r^2} + \frac{\lambda_r}{r} \frac{\partial T(r,z,t)}{\partial r} + \lambda_z \frac{\partial^2 T(r,z,t)}{\partial z^2} + q \tag{8}$$

where  $\rho = \sum \rho_i V_i / \sum V_i$ ,  $C_p = \sum \rho_i C_{p_i} V_i / \rho \sum V_i$ , *i* is the number of layers, and *q* is the heat generation per unit volume, with the assumption of the uniform heat generation,  $q = \frac{Q_{\text{generation}}}{H \times \pi \times [(r_o - l_a)^2 - r_{\text{core}}^2]}$ ,

The heat generation is given by the following equation:

$$\frac{dQ_{\text{generation}}}{dt} = I^2 \times ESR - T\frac{k_B}{e} \left[ \ln\left(\frac{d_{\text{EDL}}^+ \times \frac{C}{C_{\text{positive}}}}{V_0}\right) + \ln\left(\frac{d_{\text{EDL}}^- \times \frac{C}{C_{\text{negative}}}}{V_0}\right) \right] I$$
(9)

where *ESR* is the equivalent serial resistance, *I* is the current, *T* is the temperature,  $k_{\rm B}$  is Boltzmann constant,  $d_{\rm EDL}$  is the effective electric double layer thickness of the negative electrode,  $d^+_{\rm EDL}$  is the effective electric double layer thickness of the positive electrode,  $C_{\rm negative}$  is the areal capacitance of the negative electrode,  $C_{\rm positive}$  is the areal capacitance of the negative electrode,  $C_{\rm positive}$  is the areal capacitance of the positive electrode, and  $V_0$  is total electrolyte volume.

#### 2.3. Local entropy generation rate formulation

The entropy generation analysis that derived from the second-law of thermodynamics is an effective tool to quantify the thermodynamic irreversibilities occurring in supercapacitor cell and to determine separately the causes of irreversibilities. The expression of the local entropy generation rate in terms of fluxes and the corresponding gradients of the physical quantities can be obtained from the entropy generation balance equation [35]:

$$\rho \frac{dS_e}{dt} = -\frac{1}{T^2} J_q \cdot \nabla T - \frac{1}{T} \sum_{k=1}^n J_k \cdot \left( T \nabla \frac{\mu_k}{T} - F_k \right) - \frac{1}{T} \Pi : \nabla \nu - \frac{1}{T} \sum_{j=1}^r J_j A_j$$
(10)

where  $S_e$  is the entropy generation rate, T is the temperature,  $J_q$  is the heat flux,  $J_k$  is the diffusion flux,  $\mu$  is the chemical potential,  $F_k$  is the external force,  $\Pi$  is the viscous pressure tensor,  $\nu$  is the velocity,  $J_j$  is the chemical reaction rate,  $A_j$  is the chemical affinity.

The first term of equation (10) represents the contribution of heat transfer in the presence of temperature difference, the second term refers to the entropy generation due to mass transfer driven by species concentration difference and external forces, the third term is due to fluid friction caused by the viscous dissipation and body forces, whereas the fourth term is due to chemical reaction driven by the chemical potential gradients. Since the electrolyte is assumed to be completely infiltrated in the porous electrodes and separator, there is no fluid flow in the computational domain. The fluid is assumed to be a Newtonian fluid because there are no chemical reactions. In this case, the equation for entropy generation in the supercapacitor can be simplified as follows:

$$S_{\rm e} = -\frac{1}{T^2} \boldsymbol{J}_q \cdot \nabla T - \frac{1}{T} \sum_{k=1}^n \boldsymbol{J}_k \cdot \left( T \nabla \frac{\mu_k}{T} - \boldsymbol{F}_k \right) = S_{\rm e,h} + S_{\rm e,m} + S_{\rm e,ohm}$$
(11)

In the case of a supercapacitor, the additional external force is the chemical potential difference, the entropy generation due to ohmic losses can be defined as:

$$S_{\rm c,ohm} = \frac{1}{T} \sigma \cdot \nabla U^2 = \frac{1}{T} \left( \frac{j^2}{\sigma} \right)$$
(12)

where  $\nabla U$  is the chemical potential difference, *j* is the current density, and  $\sigma$  is the electrical conductivity.

Consequently, the rate of entropy generation can be split into three main contributions, the heat transfer entropy generation rate which is driven by temperature difference, the mass transfer entropy generation rate which is driven by concentration difference and the ohmic loss entropy generation rate which is driven by the chemical potential difference:

$$S_{\rm e} = S_{\rm e,h} + S_{\rm e,m} + S_{\rm e,ohm}$$

$$= \frac{k(\nabla T)^2}{T^2} + \sum_{k=1}^n \rho R D_k \nabla \omega_k \cdot \frac{\nabla X_k}{X_k} + \frac{1}{T} \left(\frac{j^2}{\sigma}\right)$$
(13)

where *k* is the thermal conductivity,  $\rho$  is the density, *R* is the universal gas constant,  $D_k$  is the mass diffusion coefficient,  $\omega_k$  is the mass fraction, and  $X_k$  is the molar fraction.

#### 2.4. Boundary conditions and parameters

The boundary conditions for the 1D electrochemical model are set as follows:

(1) The liquid electrical potential is equal to zero at time t = 0, and the solid electrical potential is equal to zero at the interface between the current collector and the negative electrode:

$$\varphi_l(x,t)|_{t=0} = 0, \ \varphi_s(x,t)|_{x=0} = 0$$
(14)

(2) The current density is applied at the electrode/current collector interface, and the liquid current density is equal to zero at the electrode/current collector interface:

2 ( )

$$-\frac{\partial\varphi_{s}(x,t)}{\partial x}\Big|_{x=0} = \frac{i_{\rm app}}{\sigma_{\rm ele}}, \ \frac{\partial\varphi_{s}(x,t)}{\partial x}\Big|_{x=l_{\rm neg}+l_{\rm sep}+l_{\rm pos}} = \frac{i_{\rm app}}{\sigma_{\rm ele}}\frac{\partial\varphi_{l}(x,t)}{\partial x}\Big|_{x=0,x=l_{\rm neg}+l_{\rm sep}+l_{\rm pos}} = 0,$$
$$\frac{\partial\varphi_{s}(x,t)}{\partial x}\Big|_{x=l_{\rm neg},x=l_{\rm neg}+l_{\rm sep}} = 0$$
(15)

(3) The concentrations of ions are at the same uniform concentration and the fluxes of ions are equal to zero at the electrode/current collector interface:

$$\begin{aligned} c_1(x,t)|_{t=0} &= c_2(x,t)|_{t=0} = c_0 \\ N_1(x,t)|_{x=0,x=l_{\text{neg}}+l_{\text{sep}}+l_{\text{pos}}} &= N_2(x,t)|_{x=0,x=l_{\text{neg}}+l_{\text{sep}}+l_{\text{pos}}} = 0 \end{aligned}$$
 (16)

The boundary conditions for 3D thermal model are introduced as follows:

(1)  $T_{\text{init}} = 298.15$  K;

(2) The surface of the core is adiabatic:

$$\lambda_r \frac{\partial T(r,z,t)}{\partial r} \bigg|_{r=r_{\text{core}}} = 0$$
(17)

(3) Both the natural convection and the adiabatic boundary conditions are applied on the surfaces of the supercapacitor cell:

For the natural convection,

$$-\lambda_{a} \frac{\partial T(r,z,t)}{\partial z}\Big|_{z=0} = h_{\text{conv}}(T(r,z,t) - T_{\text{amb}})$$

$$-\lambda_{a} \frac{\partial T(r,z,t)}{\partial r}\Big|_{r=r_{o}} = h_{\text{conv}}(T(r,z,t) - T_{\text{amb}})$$
(18)

For the adiabatic part,

$$-\lambda_{a} \frac{\partial T(r,z,t)}{\partial r}|_{r=r_{0}} = 0$$

$$-\lambda_{a} \frac{\partial T(r,z,t)}{\partial z}|_{z=0} = 0$$
(19)

The structural parameters measured for the 1D electrochemical model and 3D thermal model are shown in Table 1.

The relevant parameters and properties of materials are listed in Table 2.

#### 3. Results and discussion

#### 3.1. Mesh selection

A pressure-based finite volume approach is utilized to solve the governing equations using the commercial COMSOL Multiphysics 5.5 software. In such a numerical simulation, a reliable result should not be considerably affected by the size of the mesh. A grid independence test has been performed for the whole computational domain which is meshed using structured hexahedral grids and unstructured tetrahedral grids. The porosity of the porous electrode is 0.5, the electrolyte is 1 M SBPBF<sub>4</sub>/ACN, and the applied current is 100 A. Fig. 2 presents the results obtained for the local heat transfer, mass transfer and ohmic loss entropy generation rates based on different mesh sizes. As presented, the entropy generation rates are almost the same when the grid size is 0.005 m. Therefore, the grid size of 0.005 m is utilized in this work in consideration of the high calculation precision and relatively low computational costs.

#### 3.2. Model validation

To corroborate the validity of the mathematical model, a comparison

#### Table 1

Parameters used in tl	ne electrochemical model
-----------------------	--------------------------

Parameter	Notation	Value
Particle radius of the positive electrode	r <sub>pos</sub>	5 µm
Particle radius of the negative electrode	r <sub>neg</sub>	5 µm
Solid phase conductivity	$\sigma_{\rm s}$	135 S/cm <sup>2</sup>
Initial electrolyte concentration	$c_0$	$1000 \text{ mol/m}^3$
Thickness of separator	$l_{sep}$	30 µm
Thickness of the negative electrode	lneg	100 µm
Thickness of the positive electrode	lpos	100 µm
Thickness of the collector	l <sub>c</sub>	10 µm
Bruggeman porosity exponent	β	1.5
Specific areal capacitance	$C_{dl}$	0.2566 F/cm <sup>2</sup>
Surface area per unit volume of the electrode	δ	$7.583  imes 10^7 \text{ m}^2/$
		m <sup>3</sup>
Minimum stop voltage	$V_{\min}$	0 V
Maximum stop voltage	V <sub>max</sub>	2.7 V
Height of the cell	H	62 mm
Radius of the air core	$r_{\rm core}$	1 mm
External radius	ro	17.75 mm
Thickness of aluminum shell	la	88 µm
The ambient temperature	$T_{\rm amb}$	288.15 K
Boltzmann constant	$k_{\rm B}$	$1.3807  imes 10^{-23}$
		J/K
Effective electric double layer thickness of the	$d_{\rm EDL}$	0.810 nm
negative electrode		
Effective electric double layer thickness of the positive electrode	$d^+_{\rm EDL}$	0.771 nm
Areal capacitance of the negative electrode	Counting	$4.04  \mu F/cm^2$
Areal capacitance of the positive electrode	Constitute	$5.30 \mu \text{F/cm}^2$
Total electrolyte volume	Vo	25.58 cm <sup>3</sup>

Table 2	
Material properties	[36,37].

Material	Property	Symbol	Value
Activated carbon	Density	$\rho_{\rm e}$	843 kg/m <sup>3</sup>
	Specific heat capacity	$C_{\rm pe}$	539 J/(kg·K)
	Thermal conductivity	$\lambda_{e}$	0.11 W/(m·K)
Cellulose	Density	$\rho_{s}$	415 kg/m <sup>3</sup>
	Specific heat capacity	$C_{\rm ps}$	13,591 J/(kg·K)
	Thermal conductivity	$\lambda_s$	0.042 W/(m·K)
Aluminum	Density	$\rho_{a}$	2700 kg/m <sup>3</sup>
	Specific heat capacity	$C_{\rm pa}$	898.15 J/(kg·K)
	Thermal conductivity	$\lambda_{\mathbf{a}}$	237 W/(m·K)



Fig. 2. FEM mesh model and entropy generation variations with different mesh sizes.

study between the numerical results and the experimental data is implemented for the supercapacitor temperature variations under the adiabatic and natural convection conditions. The electrolyte is 1 M SBPBF<sub>4</sub>/ACN, and the porosity of the porous electrode is 0.5. For the numerical simulations, the initial temperature of the supercapacitor is set as 298.15 K, the applied current is 10 A and the cycling time is 800 s. For the experimental tests, the adiabatic temperatures of the supercapacitor are obtained by the accelerated rate calorimetric techniques, while the temperature variations in natural convection conditions are measured by T-type thermocouples pasted onto the surface of the supercapacitor cell. The comparison results for the mean temperature variations of the supercapacitor cell under thermal insulation and natural convection conditions are shown in Fig. 3(a) and (b).

The relative square errors (Equation (20)) are introduced to quantify the performance of the model:

$$\Gamma = \frac{1}{i} \sum_{i} \left( \frac{y_i - \hat{y}_i}{y_i} \right)^2$$
(20)

where *i* is the number of measurements,  $y_i$  is the theoretical value, <sup>*y*</sup>*i* is the predicted value. It can be seen that the predicted temperatures match well with the experimental values, with the errors of 0.159% in the adiabatic condition and 0.182% in the natural convection condition. In the light of the validation tests above, the mathematical model and numerical method can be adopted for describing the performance of the supercapacitor owing to their reliability and precision.

# 3.3. Temperature distribution and entropy generation rate of the supercapacitor

Fig. 4 shows the temperature distribution fields of the supercapacitor at cycling times of 30 s, 130 s and 300 s, respectively, for the charging/discharging current of 100 A. The electrolyte is 1 M TEABF<sub>4</sub>/ACN, the porosity of the porous electrode is 0.5, and the initial temperature of the supercapacitor and the ambient temperature are both set as 298.15 K. The heat generation appears to be homogeneous and the temperature

Ι



Fig. 3. Comparisons between experiments and simulation results under the (a) thermal insulation and (b) natural convection conditions.



Fig. 4. Distribution of the temperature at the time of (a) 30 s, (b) 130 s and (c)300 s.

distribution field is quite similar for all the cases except that the temperature is enhanced with the cycling time. In the process of charging and discharging, the maximum temperature area appears towards the center of the core region, and the temperature around the core region is higher than that near the aluminum shell. Besides, the temperature is lower closer to the outer surface. The reason is that there is natural convection effect between the aluminum shell and surroundings, resulting in effective heat dissipation.

Fig. 5 presents the entropy generation rates caused by the heat transfer, mass transfer and ohmic loss versus the charging/discharging time. It can be seen that the entropy generation rate due to mass transfer varies periodically over cycling time. The reason is that as the concentration gradient increases, the mass transfer entropy generation rate also increases. The concentration gradient increases due to ion adsorption during charging and decreases due to ion desorption during discharging.



Fig. 5. Variations of the entropy generation rates due to heat transfer, mass transfer and ohmic loss versus the cycling time.

The entropy generation rates due to heat transfer and ohmic loss increase with an increase in the temperature of supercapacitor during charging, and decline when the temperature decreases during discharging.

The results also indicate that the main contribution to the entropy generation rate is due to ohmic loss, corresponding up to 80% of the overall entropy generation. The second major contribution is from the mass transfer, which corresponds to 19% of the rate of total entropy generation. It is observed that the contribution due to heat transfer is much lower than the ohmic loss and mass transfer. Since the temperature distribution in the supercapacitor cell is uniform, temperature gradients are not relevant, and the corresponding heat transfer entropy generation rate is not significant. Therefore, the use of electrolyte and porous electrode considering the ohmic loss and mass transfer entropy generation rate has a significant contribution in the thermodynamic irreversibilities that occur inside the supercapacitor.

#### 3.4. Optimization of the electrolyte and porosity

In this section, four commercial electrolyte fluids (SBPBF<sub>4</sub>/ACN, TEABF<sub>4</sub>/ACN, TEABF<sub>4</sub>/ACN, TEABF<sub>4</sub>/PC, TEMABF<sub>4</sub>/ACN) are chosen to evaluate the effect of the electrolyte and the porosity on the entropy generation rate and temperature rise, and to determine the most optimal porosity for all the electrolyte fluids. To perform a meaningful comparison, the different cases are solved at the same charging/discharging current of 100 A and the cycling time of 300 s. Furthermore, in consideration of the practical applications and the performance of the supercapacitor, the porosity in the range of 0.1–0.7 is adopted.

#### (1) The optimal porosity for SBPBF<sub>4</sub>/ACN electrolyte

Contributions of heat transfer, mass transfer and ohmic loss effects to

(a)

(b)

 $S_{e}$  (W/m<sup>3</sup>K)

1.4E-20

1.2E-20

1.0E-20

8.0E-21

6.0E-21

1.0E+04

8.0E+03

6.0E+03

4.0E+03

2.0E+03

0.0E+00

0.2 0.3

0.2

0.3 0.4

0.1

0.1

 $S_{e,h}$  (W/m<sup>3</sup>K)

the entropy generation rates for the 1 M SBPBF<sub>4</sub>/ACN electrolyte versus the porosity are presented in Fig. 6(a). It can be seen that the rate of the mass transfer entropy generation decreases with the porosity increasing from 0.1 to 0.7. The increase in porosity leads to the decline in the fluid friction, thus lowering the mass transfer entropy generation rate. When the pore sizes increase to the extent when the electrolyte within the porous electrode acts on ions as a bulk fluid, the decrease in the mass transfer entropy generation rate becomes insignificant. The heat transfer generation rate decreases slightly with the increase in the porosity in the range of 0.1-0.3, while increases significantly with the porosity increasing from 0.4 to 0.7. The entropy generation rate due to ohmic losses exhibits a similar behavior with the heat transfer entropy rate, which shows its minimum value for the porosity of 0.4. In addition, as can be seen in Fig. 6(a), the contributions of irreversibility effects to the mass transfer and ohmic losses are the predominant losses (representing up to 99% of the overall irreversibility contributions).

Fig. 6(b) shows the variations of the total entropy generation rate and the temperature difference with the porosity. It is noted that the temperature increases about 0.2 °C for all the porosities during the charging and discharging cycling period, which explains the reasons of the much lower heat transfer entropy generation rate. Noteworthy, as the porosity is 0.5, the total entropy generation rate reached its lowest value. And it can be seen that the deviation in the temperature difference is much smaller compared to the deviation in the total entropy generation rate. Therefore, the total entropy generation rate is the main contribution in determining the optimal porosities. Thus, the optimal porosity for the 1 M SBPBF<sub>4</sub>/ACN electrolyte is 0.5, corresponding to the entropy generation rate of 2398 W/(m<sup>3</sup>·K) and the temperature rise of 0.20908 °C.

#### (2) The optimal porosity for TEABF<sub>4</sub>/ACN electrolyte

The variations of entropy generation rates due to the heat transfer,

S<sub>e.h</sub>

S<sub>e,ohm</sub>

S<sub>e,m</sub>

0.5 0.6

0.7 0.8

Se

 $\Delta T$ 

0.4

Porosity

3.5E+03

3.0E+03

2.5E+03

2.0E+03

5E+03

0.2108

0.2104

0.2100

0.2096

0.2092

0.2088

0.2084

/m<sup>3</sup>K)

8.0E+03

6.0E+03

4.0E+03

2.0E+03

0.0E+00

 $(m^{3}K)$ 



Porosity

0.5

0.6 0.7

mass transfer and ohmic loss for the 1 M TEABF<sub>4</sub>/ACN electrolyte with different porosities are plotted in Fig. 7(a). As shown, the entropy generation rates due to heat transfer and ohmic loss for the TEABF<sub>4</sub>/ACN electrolyte exhibit a similar trend to the TEABF<sub>4</sub>/ACN electrolyte. The heat transfer entropy generation rate declines with porosity in the range of 0.1–0.3 whereas it is enhanced for porosity ranging from 0.4 to 0.7. Meanwhile, the ohmic loss entropy generation rate is also suppressed with porosity changing from 0.1 to 0.4 whereas it is elevated in the range of 0.5–0.7. Conversely, the same increase in porosity results in the lower mass transfer entropy generation rate. Larger porosity leads to an undesirable decrease in fluid friction of the electrolyte ions. Therefore, mass transfer entropy generation is minimized with larger porosity.

Fig. 7(b) presents the variations in total entropy generation rate and temperature difference with different porosities. These results suggest that the relative error of temperature difference is below 0.002% with the porosity increasing from 0.1 to 0.7, which is much smaller than the relative error of the total entropy generation rate. The lowest value of the total entropy generation rate is obtained at the porosity of 0.4. Therefore, the optimal porosity for the 1 M TEABF<sub>4</sub>/ACN electrolyte is 0.4, in which the entropy generation rate is 2240 W/(m<sup>3</sup>·K) and the temperature rise is 0.20941 °C.

Variations of the entropy generation rates due to heat transfer, mass transfer and ohmic loss against the porosity for the 1 M TEMABF<sub>4</sub>/ACN electrolyte are shown in Fig. 8(a). Interestingly, the contributions from entropy generation to ohmic loss and heat transfer share the similar trend. They first decrease as the porosity gets larger and the lowest value of the heat transfer and ohmic loss entropy generation rate corresponds to the porosity of 0.4. Then, both entropy generation rates rise with the increasing porosity from 0.5 to 0.7. On the other hand, the mass transfer entropy generation rate decreases significantly while the porosity is increasing from 0.1 to 0.7.



**Fig. 7.** (a) Variations of the entropy generation rates due to heat transfer, mass transfer and ohmic loss versus the porosity, and (b) the evolutions of entropy generation rate and temperature rise versus the porosity for the 1 M TEABF<sub>4</sub>/ ACN electrolyte.

(3) The optimal porosity for TEMABF<sub>4</sub>/ACN electrolyte



**Fig. 8.** (a) Variations of the entropy generation rates due to heat transfer, mass transfer and ohmic loss versus the porosity, and (b) the evolutions of entropy generation rate and temperature rise versus the porosity for the 1 M TEMABF<sub>4</sub>/ ACN electrolyte.

(4) The optimal porosity for TEABF<sub>4</sub>/PC electrolyte

Fig. 8(b) presents the evolutions of total entropy generation rate and temperature difference for the 1 M TEMABF<sub>4</sub>/ACN electrolyte versus the porosity value. It can also be observed that as the porosity becomes larger, the overall entropy generation rate first decreases and then increases faster when the porosity reaches 0.4. The variations of temperature rise with respect to the TEMABF<sub>4</sub>/ACN electrolyte change from 0.209023 °C at the porosity of 0.3 to 0.209933 °C at the porosity of 0.2, in which the derivation is too small to be considered. Thus, the optimal porosity for the 1 M TEMABF<sub>4</sub>/ACN electrolyte is 0.4, corresponding to the entropy generation rate of 2230 W/(m<sup>3</sup>·K) and the temperature rise of 0.20974 °C.

Fig. 9 shows the heat transfer entropy generation rate, the ohmic loss entropy generation rate, the mass transfer entropy generation rate, the overall entropy generation rate and temperature rise with variation in porosity for the 1 M TEABF<sub>4</sub>/PC electrolyte. According to Fig. 9(a), the ohmic loss entropy generation rate is much larger than for the other three electrolyte fluids, showing a rising trend with greater values of porosity. For lower porosity in the range of 0.1–0.4, the peak value of heat transfer entropy generation rate is observed for the porosity of 0.3. And then it shows a monotonic rise with the porosity increasing from 0.5 to 0.7. The mass transfer entropy generation rate exhibits a similar declining trend to the mass transfer entropy generation rate of the other three electrolyte fluids.

Inspection of the results in Fig. 9(b) reveals that the lowest value of the total entropy generation rate is observed at the porosity of 0.2. Furthermore, the temperature rise fluctuates slightly from 0.20900 °C to 0.20972 °C with the porosity, whose variation is much smaller than the total entropy generation rate. In this case, the optimal porosity for the 1 M TEABF<sub>4</sub>/PC electrolyte is 0.2, corresponding to the entropy generation rate of 7720 W/(m<sup>3</sup>·K) and the temperature rise of 0.20951 °C.

Moreover, it can also be seen in Figs. 6-9, for each of the electrolytes



**Fig. 9.** (a) Variations of the entropy generation rates due to heat transfer, mass transfer and ohmic loss versus the porosity, and (b) the evolutions of entropy generation rate and temperature rise versus the porosity for the  $1 \text{ M TEABF}_4/\text{PC}$  electrolyte.

analyzed, that the trend in the entropy generation rates due to heat transfer, mass transfer and ohmic loss are almost the same for all the physical phenomena occurring within the supercapacitor cell. This is mainly due to the similar ion transport and the similar temperature distributions among the cell during the charging and discharging cycle. After analyzing the entropy generation rates due to different mechanisms and the temperature differences for four different electrolytes, the optimal porosity for each electrolyte is determined. The total entropy generation rate and temperature analysis on the four electrolytes with the corresponding optimum porosity is carried out to optimize the characteristics of the supercapacitor cell. The total entropy generation rates and temperature differences of supercapacitor cells with different electrolytes are compared in Fig. 10. The smallest temperature rise corresponds to SBPBF4/ACN, which is approximately 0.315% lower



Fig. 10. Variations of entropy generation rate and temperature rise for all the electrolytes at the most optimal porosity value.

than for the TEMABF<sub>4</sub>/ACN electrolyte. Therefore, the electrolyte has little effect on the temperature difference variation. The largest thermodynamic irreversibilities occur in the TEABF<sub>4</sub>/PC electrolyte while the lower irreversibilities are present for the cases of TEABF<sub>4</sub>/ACN and TEMABF<sub>4</sub>/ACN electrolytes. It also can be seen that the deviations in the temperature differences between the four electrolytes are much smaller compared to the deviations in the entropy generation rates. Thus, the effect of temperature difference on determining the optimal electrolyte can be neglected. It is noted that compared to the TEABF<sub>4</sub>/ACN electrolyte, the TEMABF<sub>4</sub>/ACN electrolyte exhibits better electrochemical performance [38]. Therefore, the 1 M TEMABF<sub>4</sub>/ACN electrolyte with the porosity of 0.4 is the most optimal choice in consideration of the thermodynamic irreversibilities and the electrochemical behavior of the supercapacitor.

#### 3.5. Effect of the current

Apart from the electrolyte and porosity, the current also plays a significant role in the entropy generation within the supercapacitor. The electrolyte is the 1 M TEMABF<sub>4</sub>/ACN solution and the porosity is 0.4. The effect of changing the charging and discharging current of the



Fig. 11. (a) Variations of the entropy generation rates due to heat transfer, mass transfer and ohmic loss, (b) variations of entropy generation rate and temperature rise and (c) variation of parameter  $\Pi$  versus the current for the 1 M TEMABF<sub>4</sub>/ACN electrolyte.

supercapacitor is analyzed and presented in Fig. 11. Fig. 11(a) reveals the effect of the current on the entropy generation rates due to heat transfer, ohmic loss and mass transfer. It can be observed that the three different entropy generation rates show a similar behavior, increasing with the current increasing from 10 A to 200 A. The effect of current on the total entropy generation rate and temperature difference is presented Fig. 11(b). The temperature difference and total entropy generation rate both increase with the current in the range of 10 A–200 A. We note that the higher charging/discharging current would lead to more effective heat generation, whereas the stronger temperature and viscous effects will result in the higher total entropy generation rate. Furthermore, the small curve slope at low values of the current indicates minor irreversibilities, while the curve rises quickly at high values of the current as a consequence of stronger irreversibility effects.

To understand the relative dominance of the entropy generation rate owing to mass transfer irreversibilities, a dimensionless parameter is introduced, defined as:

$$\Pi = \frac{S_{e,m}}{S_e} \tag{21}$$

As noted, parameter  $\Pi$  defines the ratio of irreversibility due to mass transfer to the total irreversibilities. The values of parameter  $\Pi$  of supercapacitor with different currents is compared in Fig. 11(c), showing a declining trend with higher currents. Noteworthy, as the current increases, the entropy generation rates due to heat transfer, ohmic loss and mass transfer also increase. However, the increase of the ohmic loss entropy generation rate is much larger compared to other loss channels, leading to the lower mass transfer irreversibility ratio.

The results also indicate that when the current increases from 10 A to 25 A, the main contribution to the entropy generation rate is due to mass transfer, while the ohmic loss irreversibilities become dominant with the current in the range of 50 A–200 A. To understand the contributions of the mechanisms, the ohmic loss entropy generation rate and the mass transfer entropy generation rate are compared with the current in the range of 25 A–50 A, shown in Fig. 12. It can be seen that the mass transfer entropy generation rate is higher than the ohmic loss entropy generation rate at currents below 46 A. When the current value reaches 46 A, the ohmic loss becomes the main contribution to the total irreversibilities. Therefore, when the charging/discharging current is below 46 A, the optimization approaches considering the reduction of mass transfer entropy generation rate should be adopted. On the other hand, when the current is higher than 46 A, the ohmic loss entropy generation



Fig. 12. Variations of the entropy generation rates due to mass transfer and ohmic loss in the current range from 25 A to 50 A.

rate has a significant contribution in the reduction of thermodynamic irreversibilities.

#### 4. Conclusion

In this paper, an entropy generation analysis of the supercapacitor cell with four different electrolytes has been implemented for the first time to evaluate the effects of thermodynamic irreversibilities. The research is based on a coupled CFD model that consists of a 1D electrochemical model and a 3D thermal model for a single cell, taking into account the transport of species, heat and mass. The local entropy generation rate is derived from the second-law of thermodynamics to single out the mechanisms of irreversibilities and to quantify the thermodynamic irreversibilities occurring in a supercapacitor cell. In particular, the contributions of heat transfer, mass transfer and ohmic loss effects are considered in the entropy generation model. The present model results show a good agreement with the experimental values, with the very low errors of 0.159% in the adiabatic condition and 0.182% in the natural convection condition, respectively. The effects of the electrolyte, porosity and current have been investigated and then optimized. The following results have been obtained:

- (1) The irreversibilities that occur in the supercapacitor cell, according to the entropy generation analysis are mainly due to three physical phenomena: ohmic losses, mass transfer, and heat transfer.
- (2) The results indicate that the entropy generation rates due to the ohmic loss, mass transfer and heat transfer vary periodically over cycling time. The reason is that the temperature and concentration gradient experience periodical variations. These variations increase during charging as ions form the electric double layer and decrease during discharging as the ions resume their uniform concentration. The most important generation of global entropy in the system is due to the ohmic loss corresponding to the 80% of the total global entropy generation. The ohmic loss is followed by the mass transfer, representing almost 19% of the total entropy generation. It is observed that the contribution due to heat transfer is much lower than the ohmic loss and mass transfer.
- (3) The trend in the entropy generation rates due to heat transfer, mass transfer and ohmic loss are almost the same for all the four electrolytes. This is mainly due to the similar ion transport and the similar temperature distributions among the cell during the charging and discharging cycle. The optimal porosities for the 1 M SBPBF<sub>4</sub>/ACN, 1 M TEABF<sub>4</sub>/ACN, 1 M TEABF<sub>4</sub>/PC, and 1 M TEMABF<sub>4</sub>/ACN electrolytes are 0.5, 0.4, 0.4 and 0.2, respectively. Compared with other electrolytes, the 1 M TEMABF<sub>4</sub>/ACN electrolyte with the porosity of 0.4 is the most proper choice in consideration of the thermodynamic irreversibilities and the electrochemical behavior of the supercapacitor, corresponding to the entropy generation rate of 2230 W/(m<sup>3</sup>·K) and the temperature rise of 0.20974 °C.
- (4) The entropy generation rates caused by heat transfer, ohmic loss and mass transfer increase with the current increasing from 10 A to 200 A. Furthermore, when the current increases from 10 A to 46 A, the main contribution to the entropy generation rate is due to the mass transfer, while the ohmic loss related irreversibilities become dominant within the current range of 46 A–200 A.

This analysis confirms that the thermodynamic performance of the supercapacitor is significantly affected by the choice of electrolytes and operating conditions, which must be carefully selected. The outcomes of this work are useful in the development of efficient supercapacitor cells with lower irreversibilities. In the future, it can be anticipated that many studies will be devoted to this issue to optimize other energy conversion and storage systems.

#### Author statement

Haowen Li: Conceptualization; Methodology; Investigation; Formal analysis; Data curation; Writing – original draft. Huachao Yang: Methodology; Writing – review & editing. Jianhua Yan: Resources. Kefa Cen: Supervision. Kostya (Ken) Ostrikov: Writing – review & editing; Supervision. Zheng Bo: Supervision; Resources; Writing – review & editing.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data availability

Data will be made available on request.

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# Entropy generation analysis in supercapacitor modules based on a three-dimensional coupled thermal model



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#### ABSTRACT

Efficient and sustainable thermal management systems are crucial to ensure thermal stability and a long lifespan of supercapacitors. In this work, an entropy generation analysis based on an improved coupled model is implemented for the first time, with the aim to facilitate the design and optimization of the supercapacitor thermal management systems. Entropy generation analysis accurately quantifies the irreversibilities due to heat transfer and fluid friction of the supercapacitor module, allowing for direct identification of the causes of inefficiency that cannot be achieved by the conventional energy analysis. The improved coupled model incorporates a one-dimensional electrochemical model and three-dimensional thermal models at cell and module levels. The optimal thermal management scheme of the supercapacitor module is obtained by the multiparametric optimization based on four evaluation criteria derived from the coupled energy and entropy generation analyses. Results indicate that the supercapacitor module with lower entropy generation and higher heat transfer rate can be achieved with the geometry of transverse pitch equal to longitudinal pitch twice the diameter with the 293.15 K cooling water. This work provides the advanced fundamental and computational frameworks for the development of next-generation, energy-smart thermal management systems for clean and renewable energy conversion and storage modules.

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#### 1. Introduction

Supercapacitors, the highly efficient energy storage devices based on the electrostatic attraction of ions at the solid/liquid interface, have shown prominent advantages over rechargeable batteries in terms of high power density and ultrahigh charge/ discharge rate [1]. These beneficial features make them particularly attractive for high-power-demanding applications, such as braking energy regeneration, automotive vehicles, and non-interrupted emergency power supply. In these cases, high-power charging and discharging conditions are accompanied by inevitable temperature rise or even overheating issues, resulting in electrolyte

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solvent evaporation [2] and unsatisfactory cycling performance [3]. For example, the lifetime of supercapacitors decreases by a half every 10 °C above the standard temperature of 25 °C [4,5]. Besides, the temperature differences between the cells in the module must be controlled within 5 °C for a full lifespan [6]. Considering the key role of temperature on the performance of supercapacitors, continuous endeavors have been directed towards the development of efficient thermal management schemes. Consequently, the evaluation criteria, as the preliminary object, should be introduced to assist the design and optimization of the supercapacitor thermal management systems. For example, the evaluation parameters based on the energy analysis such as temperatures of the supercapacitors, Nusselt number (Nu), and friction factor have been proposed to evaluate the effectiveness of supercapacitor thermal management approaches [3,7–10]. However, such evaluation processes do not account for the limitations stipulated by the second law of thermodynamics. Indeed, the contribution of the second-law thermodynamic principles in the assessment of supercapacitor





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thermal management performance has not received the attention it merits.

Entropy generation, that is derived from the second law of thermodynamics, properly quantifies thermodynamic irreversibility and efficiency, thus providing an insight that cannot be achieved by the energy analysis alone [11]. The entropy generation determines the mechanisms of irreversibilities associated with heat transfer, fluid flow, and mass transfer, and can help maximize the gainful use of the available resources. Furthermore, due to its direct derivation from the basic thermodynamic principles and attractive features, entropy generation analysis can be in principle used for optimizing any type of energy conversion and storage systems [12–18]. For example, Giangaspero et al. [19] proposed an entropy generation minimization method to optimize the configuration of the solar heat exchanger surfaces. Entropy generation maps were also investigated by Giangaspero et al. [20] to perform an optimization of the forced convective cooling of LED-based spotlight. Ramírez-Minguela et al. [21] carried out an entropy generation analysis in the solid oxide fuel cell with four different biogas compositions in order to evaluate the effects of the different biogas compositions in the prediction of thermodynamic irreversibilities. Despite these advances, the entropy generation analysis related to the supercapacitor module, rigorously accounting for the irreversibilities arising due to heat transfer and fluid friction processes, to the best of our knowledge, has never been reported yet.

Besides the lack of the entropy generation analyses of supercapacitor thermal management systems, the mathematic models used in predicting supercapacitor heat generation and heat transfer performance have several limitations. For example, a threedimensional (3D) model [3,7] was developed to investigate the heat transfer performance inside and around the supercapacitor module, in which only the Joule heat is considered in the heat generation of supercapacitor. The coupled model of a onedimensional (1D) equivalent circuit model and a 3D temperature distribution model [22] were used to predict and optimize the thermal performance of the supercapacitor module. However, the transport of species in the cell was neglected by the 1D equivalent circuit model, and heat generation only due to the Joule effect was considered. Considering the importance of precisely predicting the supercapacitor thermal behavior in the evaluation of thermal management systems, an accurate mathematic model is indispensable.

In this work, an entropy generation analysis based on a detailed coupled model is for the first time conducted to accurately quantify the heat transfer irreversibility and the fluid friction irreversibility of the supercapacitor module. The coupled model consists of a 1D electrochemical model, the 3D thermal models for a single cell and a supercapacitor module. This coupled model is developed with the finite volume method taking into account a complete solution of the electrochemical, heat generation, mass, momentum and energy equations. The entropy generation mechanisms associated with the heat transfer and viscous dissipation are investigated. In addition, four evaluation criteria based on the energy and entropy generation analyses, namely heat transfer entropy generation, fluid friction entropy generation, Nu, and ecological coefficient of performance (ECOP), are proposed for the evaluation and optimization of the thermodynamic performance of the supercapacitor thermal management systems. Finally, the effects of physical parameters such as structural geometries, cooling liquids, inlet temperature, current, and inlet water velocity on the thermodynamic irreversibilities and heat transfer characteristics of the supercapacitor modules are examined. The results can pave the way for more efficient thermal management of supercapacitor and other energy conversion and storage systems.

#### 2. Methodology

The modeling approach is based on the coupling of a 1D electrochemical model, a 3D thermal model for the supercapacitor cell and a 3D forced convective heat transfer model for the supercapacitor module. Fig. 1 represents the computational domains and the way that they are coupled to form the coupled mathematical model.

#### 2.1. 1D electrochemical model

#### 2.1.1. Model assumptions

As illustrated in Fig. 1(a), a supercapacitor cell typically has a negative electrode, a positive electrode, electrolyte, a separator, and current collectors at both the anode and cathode. The current collectors are eliminated in the computational domain of the electrochemical model in consideration of their higher electrical conductivity. To simplify the model and concentrate on the key phenomena and parameters, several assumptions are adopted for this model: 1) porous electrode theory is adopted; 2) Faradaic reactions are negligible; 3) the computational domain is symmetrical; 4) the electrolyte is completely infiltrated in the porous electrodes and separator; 5) self-discharge and charge redistribution effects are neglected.

#### 2.1.2. Governing equations

The charge conservation equation can be obtained as:

$$\frac{\partial i_s(x,t)}{\partial x} + \frac{\partial i_l(x,t)}{\partial x} = 0$$
(1)

$$\frac{\partial i_l(x,t)}{\partial x} = \delta C_{dl} \frac{\partial (\varphi_s(x,t) - \varphi_l(x,t))}{\partial t}$$
(2)

The solid current density follows Ohm's Law:

$$i_{s}(x,t) = -\sigma_{ele} \frac{\partial \varphi_{s}(x,t)}{\partial x}$$
(3)

$$\sigma_{\rm ele} = \sigma_s (1 - \varepsilon_{\rm ele})^{\beta} \tag{4}$$

The relation between liquid current density and ion flux can be described as:

$$i_{l}(x,t) = -\frac{F^{2}}{RT} \frac{\partial \varphi_{l}(x,t)}{\partial x} \sum_{i=1,2} e_{i}^{2} D_{i} c_{i}(x,t) - F \sum_{i=1,2} e_{i} D_{i} \frac{\partial c_{i}(x,t)}{\partial x}$$
(5)

, where the subscript i is the electrolyte ions, for a binary electrolyte i = 1, 2.

Based on the Nernst-Planck equation, the flux of ions  $N_i$  can be expressed by the following equation. The two terms on the righthand side of the equation represent ion migration and diffusion in the electrolyte solution.

$$N_{i}(x,t) = -\frac{e_{i}F}{RT}D_{i}c_{i}\frac{\partial\varphi_{l}(x,t)}{\partial x} - D_{i}\frac{\partial c_{i}(x,t)}{\partial x}$$
(6)

$$D_{i,\text{ele}} = D_i \cdot \varepsilon_{\text{ele}}^{\beta}, D_{i,\text{sep}} = D_i \cdot \varepsilon_{\text{sep}}^{\beta}$$
(7)

The mass balance in the electrolyte phase can be described as follows:

$$\frac{\partial c_i(x,t)}{\partial t} = -\frac{\partial N_i(x,t)}{\partial x} + R_i$$
(8)

, where  $R_i$  is the source term (here  $R_i = 0$  with no chemical reactions).



Fig. 1. The coupled mathematical model incorporates (a) 1D electrochemical model, 3D thermal model for the supercapacitor (b) cell and (c) module.

#### 2.1.3. Boundary conditions

The appropriate boundary conditions are introduced as follows:

1) The interface between the negative electrode and the current collector is electrically grounded and the liquid electrical potential is equal to zero at time t = 0:

$$\begin{aligned} \varphi_s(x,t)|_{x=0} &= 0 \\ \varphi_l(x,t)|_{t=0} &= 0 \end{aligned} \tag{9}$$

2) The current density is imposed at the current collector/electrode interface, and the liquid current density is vanished at the current collector/electrode interface:

$$\frac{\partial \varphi_{s}(x,t)}{\partial x}\Big|_{x=0} = \frac{i_{app}}{\sigma_{ele}}$$

$$\frac{\partial \varphi_{s}(x,t)}{\partial x}\Big|_{x=l_{neg}+l_{sep}+l_{pos}} = \frac{i_{app}}{\sigma_{ele}}$$

$$\frac{\partial \varphi_{s}(x,t)}{\partial x}\Big|_{x=l_{neg},x=l_{neg}+l_{sep}} = 0$$

$$\frac{\partial \varphi_{l}(x,t)}{\partial x}\Big|_{x=0,x=l_{neg}+l_{sep}+l_{pos}} = 0$$
(10)

3) Both ions of the binary electrolyte that at time t = 0 are at the same uniform concentration and the fluxes of ions are zero at the current collector/electrode interface:

$$\begin{aligned} c_1(x,t)|_{t=0} &= c_2(x,t)|_{t=0} = c_0 \\ N_1(x,t)|_{x=0,x=l_{\text{neg}}+l_{\text{sep}}+l_{\text{pos}}} &= N_2(x,t)|_{x=0,x=l_{\text{neg}}+l_{\text{sep}}+l_{\text{pos}}} = 0 \end{aligned} \tag{11}$$

#### 2.1.4. Parameters

The parameters of electrodes and electrolyte for the 1D electrochemical modeling are summarized in Table 1.

#### 2.2. 3D thermal model for a single cell

#### 2.2.1. Model assumptions

The 3D thermal model for a single cell is shown in Fig. 1(b). The mathematical model is simplified by the following assumptions: 1) thermal conduction is the dominant mechanism of the heat transfer; 2) thermal resistances at interfaces are neglected; 3) the heat generation is uniform during the charging and discharging processes; 4) the heat transfer of the cover plate is considered as adiabatic due to its low thermal conductivity.

#### 2.2.2. Governing equations

Since the structure is symmetrical in 3D cylindrical coordinates, the transient equation can be written as shown below:

Table 1	
Parameters used in the electrochemical model	[23-25].

Parameter	Notation	Value
Volume fraction for porous electrode	<i>e</i> ele	45%
Volume fraction for separator	$\varepsilon_{sep}$	70%
Solid phase conductivity	$\sigma_{\rm s}$	135 S/cm <sup>2</sup>
Initial electrolyte concentration	<i>c</i> <sub>0</sub>	1000 mol/m <sup>3</sup>
Thickness of separator	lsep	30 µm
Thickness of negative electrode	lneg	100 µm
Thickness of positive electrode	lpos	100 µm
Thickness of collector	l <sub>c</sub>	10 µm
Thickness of aluminum shell	la	88 µm
Bruggeman porosity exponent	β	1.5
Specific areal capacitance	C <sub>dl</sub>	0.2566 F/cm <sup>2</sup>
Surface area per unit volume of electrode	δ	$7.5827 \times 10^7 \text{ m}^2/\text{m}^3$

$$\frac{\partial \left(\rho C_p T(r,z,t)\right)}{\partial t} = \lambda_r \frac{\partial^2 T(r,z,t)}{\partial r^2} + \frac{\lambda_r}{r} \frac{\partial T(r,z,t)}{\partial r} + \lambda_z \frac{\partial^2 T(r,z,t)}{\partial z^2} + q$$
(12)

, where  $\rho = \sum \rho_i V_i / \sum V_i$ ;  $C_p = \sum \rho_i C_{p_i} V_i / \rho \sum V_i$ ; the subscript *i* is the number of layers; *q* refers to the heat generation per unit volume,  $q = Q_{\text{generation}} / [H \times \pi \times ((r_0 - l_a)^2 - r_i^2)]$ .

The heat generation consists of Joule heat and the heat generation due to entropy change:  $Q_{\text{generation}} = Q_{\text{Joule}} + Q_{\text{entropy}}$ .

The heat generation due to the Joule effect is given by the following equation [23]:

Table 3		
Material	properties	[28,29]

Property	Symbol	Value
Density	ρ <sub>e</sub>	843 kg/m <sup>3</sup>
Specific heat capacity	Cpe	539 J/(kg·K)
Density	$\rho_{\rm s}$	415 kg/m <sup>3</sup>
Specific heat capacity	$C_{ps}$	13591.3 J/(kg·K)
Density	$\rho_{\rm a}/\rho_{\rm c}$	2700 kg/m <sup>3</sup>
Specific heat capacity	$C_{\rm pa}/C_{\rm pc}$	898.15 J/(kg·K)
	Property Density Specific heat capacity Density Specific heat capacity Density Specific heat capacity	$\begin{array}{c c} Property & Symbol \\ \hline Density & \rho_e \\ Specific heat capacity & C_{pe} \\ Density & \rho_s \\ Specific heat capacity & C_{ps} \\ Density & \rho_a   \rho_c \\ Specific heat capacity & C_{pa}   C_{pc} \\ \end{array}$

2.2.4. Parameters

The parameters for the 3D thermal model are presented in

$$\frac{dQ_{\text{Joule}}}{dt} = I^2 \times (0.05035 - 0.002151 \times I - 0.000126 \times T + 0.000006304 \times T \times I)$$
  
= 0.05035I^2 - 0.002151I^3 - 0.000126I^2T + 0.000006304I^3T

The heat generation due to entropy change is expressed as follows [23,26]:

$$\frac{dQ_{\text{entropy}}}{dt} = -T\frac{k_B}{e} \left[ \ln\left(\frac{d_{\text{EDL}}^+ \times \frac{C}{C_{\text{positive}}}}{V_0}\right) + \ln\left(\frac{d_{\text{EDL}}^- \times \frac{C}{C_{\text{negative}}}}{V_0}\right) \right] I$$
(14)

#### 2.2.3. Boundary conditions Boundary conditions are:

Table 2

- 1) The initial temperature is uniform and equal to the ambient temperature:  $T(\mathbf{r}, \mathbf{z}, \mathbf{0}) = T_{amb}$ ;
- 2) The surface of inner core is thermally insulated and the natural convection is applied on the surfaces of the supercapacitor cell:

$$\lambda_{r} \frac{\partial T(r, z, t)}{\partial r} \Big|_{r=r_{i}} = 0$$

$$-\lambda_{a} \frac{\partial T(r, z, t)}{\partial r} \Big|_{r=r_{o}} = h_{\text{conv}}(T(r, z, t) - T_{\text{amb}})$$

$$-\lambda_{a} \frac{\partial T(r, z, t)}{\partial z} \Big|_{z=0} = h_{\text{conv}}(T(r, z, t) - T_{\text{amb}})$$
(15)

Parameters used in the thermal model [23,27].

Table 2 and the related properties of supercapacitor materials are listed in Table 3.

#### 2.3. 3D thermal model for supercapacitor module

#### 2.3.1. Model assumptions

As presented in Fig. 1(c), a 3D supercapacitor module in the staggered pattern of five rows is created for numerical computations. Symmetrical planes are used to simplify the computational domain and reduce the computation time. To simplify this numerical simulation, the following assumptions are made: 1) the fluids are incompressible Newtonian fluids and there is a constant volume flow rate; 2) the gravity and buoyancy lift effects are negligible; 3) viscous dissipation is negligible; 4) the radiation is neglected because of the small temperature differences between the supercapacitor surfaces and the cooling fluids; 5) the no-slip conditions are used on all solid walls of supercapacitors.

#### 2.3.2. Governing equations

The continuity equation, momentum equations, and energy equation are solved in this model, the expressions of which are listed as follows.

Conservation of mass:

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} = 0$$
(16)

Conservation of momentum:

Parameter	Notation	Value
Height	Н	62 mm
External radius	ro	17.75 mm
Inner core radius	r <sub>i</sub>	0.5 mm
Thickness of aluminum shell	la	88 µm
Boltzmann constant	$k_{ m B}$	$1.3807 \times 10^{-23} \text{ J/K}$
Effective electric double layer thickness of negative electrode	$d_{EDL}^{-}$	0.810 nm
Effective electric double layer thickness of positive electrode	$d^+_{\rm EDL}$	0.771 nm
Areal capacitance of negative electrode	Cnegative	4.04 μF/cm <sup>2</sup>
Areal capacitance of positive electrode	C <sub>positive</sub>	5.30 µF/cm <sup>2</sup>
Total electrolyte volume	V <sub>0</sub>	25.58 cm <sup>3</sup>

(13)

$$\frac{\partial u}{\partial \tau} + u \frac{\partial u}{\partial x} + v \frac{\partial u}{\partial y} + w \frac{\partial u}{\partial z} = -\frac{1}{\rho} \frac{\partial p}{\partial x} + v \left( \frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} + \frac{\partial^2 u}{\partial z^2} \right) + \frac{1}{\rho} F_x$$

$$\frac{\partial v}{\partial \tau} + u \frac{\partial v}{\partial x} + v \frac{\partial v}{\partial y} + w \frac{\partial v}{\partial z} = -\frac{1}{\rho} \frac{\partial p}{\partial y} + v \left( \frac{\partial^2 v}{\partial x^2} + \frac{\partial^2 v}{\partial y^2} + \frac{\partial^2 v}{\partial z^2} \right) + \frac{1}{\rho} F_y$$

$$\frac{\partial w}{\partial \tau} + u \frac{\partial w}{\partial x} + v \frac{\partial w}{\partial y} + w \frac{\partial w}{\partial z} = -\frac{1}{\rho} \frac{\partial p}{\partial z} + v \left( \frac{\partial^2 w}{\partial x^2} + \frac{\partial^2 w}{\partial y^2} + \frac{\partial^2 w}{\partial z^2} \right) + \frac{1}{\rho} F_z$$
(17)

Conservation of energy:

$$\frac{\partial T}{\partial \tau} + u \frac{\partial T}{\partial x} + v \frac{\partial T}{\partial y} + w \frac{\partial T}{\partial z} = \frac{\lambda}{\rho c_p} \left( \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} \right) + q$$
(18)

#### 2.3.3. Boundary conditions

The boundary conditions are:

- 1) At the inlet of extended domain, uniform velocities and temperatures profiles are considered:  $v_x = v_{inlet}$ ,  $v_y = v_z = 0$ ,  $T = T_{inlet}$ ;
- 2) At the outlet of the extended domain, pressure is equal to the atmospheric pressure:  $P_{\text{outlet}} = P_{\text{atm}}$ ;
- 3) At the interfaces, the uniform heat flux and temperature are applied:  $\lambda_i(\partial T_i / \partial n) = \lambda_j(\partial T_j / \partial n)$  and  $T_i = T_j$ , where *i* and *j* are the indices of the parts separated by the interface.

#### 2.4. Entropy generation model

The entropy generation analysis is implemented once the CFD simulations are accomplished. The entropy generation is defined as [30,31]:

$$S_{\text{gen}} = -\frac{1}{T^2} J_q \cdot \nabla T - \frac{1}{T} \sum_{k=1}^n J_k \cdot \left( T \nabla \frac{\varphi_k}{T} - F_k \right) - \frac{1}{T} \Pi : \nabla \nu - \frac{1}{T} \sum_{j=1}^r J_j A_j$$
(19)

Since the fluid is Newtonian fluid and there is no chemical reaction, the expression of entropy generation can be simplified as follows:

$$S_{\text{gen}} = S_{\text{gen},h} + S_{\text{gen},f} \tag{21}$$

where  $S_{\text{gen,h}}$  and  $S_{\text{gen,f}}$  are the heat transfer entropy generation and fluid friction entropy generation, respectively. Thus, the expressions for the heat transfer and fluid friction entropy generations are as follows:

$$S_{\text{gen},h} = \frac{\lambda}{T^2} \left[ \left( \frac{\partial T}{\partial x} \right)^2 + \left( \frac{\partial T}{\partial y} \right)^2 + \left( \frac{\partial T}{\partial z} \right)^2 \right]$$
(22)

$$S_{\text{gen},f} = \frac{\mu}{T} \left[ 2 \left( \left( \frac{\partial u}{\partial x} \right)^2 + \left( \frac{\partial v}{\partial y} \right)^2 + \left( \frac{\partial w}{\partial z} \right)^2 \right) + \left( \frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right)^2 + \left( \frac{\partial u}{\partial z} + \frac{\partial w}{\partial x} \right)^2 + \left( \frac{\partial w}{\partial y} + \frac{\partial v}{\partial z} \right)^2 \right]$$
(23)

The relative dominance of entropy generation owing to heat transfer and fluid friction irreversibilities is described by the Bejan number (Be) [12], a dimensionless parameter, defined as:

$$Be = \frac{S_{gen,h}}{S_{gen}}$$
(24)

Therefore, Be > 0.5 indicates the dominance of heat transfer irreversibility, Be < 0.5 implies irreversibility is dominated by fluid friction effects, and Be = 0.5 is the case wherein heat transfer irreversibility and fluid flow irreversibility are of equal importance.

#### 2.5. Evaluation criteria

In this section, four evaluation criteria are introduced for assessing the heat transfer characteristics and irreversibilities of supercapacitor thermal management systems.

- (1) The entropy generation due to heat transfer ( $S_{\text{gen},h}$ ) and fluid friction ( $S_{\text{gen},f}$ ) are selected as the first two criteria. The minimization of entropy generation is an optimization approach of the thermodynamic imperfections and fluid flow irreversibility [32].
- (2) Nu is the third criterion to evaluate the heat transfer rate.
- (3) A criterion is recently presented as a benchmark for the evaluation of performance, which is the so-called ECOP based on the second law of thermodynamics [17,33]. ECOP specifies the ratio of heat transfer rate and total irrevers-

$$S_{\text{gen}} = -\frac{1}{T^2} J_q \cdot \nabla T - \frac{1}{T} \Pi : \nabla \nu$$
  
=  $\frac{\lambda}{T^2} \left[ \left( \frac{\partial T}{\partial x} \right)^2 + \left( \frac{\partial T}{\partial y} \right)^2 + \left( \frac{\partial T}{\partial z} \right)^2 \right]$   
+  $\frac{\mu}{T} \left[ 2 \left( \left( \frac{\partial u}{\partial x} \right)^2 + \left( \frac{\partial v}{\partial y} \right)^2 + \left( \frac{\partial w}{\partial z} \right)^2 \right) + \left( \frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right)^2 + \left( \frac{\partial u}{\partial z} + \frac{\partial w}{\partial x} \right)^2 + \left( \frac{\partial w}{\partial y} + \frac{\partial v}{\partial z} \right)^2 \right]$ 

, where the first term on the right-hand side accounts for the entropy generation due to heat transfer; the second term accounts for the entropy generation due to fluid friction.

Equation (20) can be presented as:

ibility rate while the Bejan number does not give any information about the rate of heat transfer. Hence, increasing the average *Nu* and decreasing entropy generation are simultaneously desirable. ECOP can be defined as follows:

(20)



Fig. 2. (a) Computational grid and (b) the outlet air temperature with different element grid sizes.

$$ECOP = \frac{Nu}{S_{gen}}$$
(25)

#### 3. Results and discussion

#### 3.1. Grid independence

A pressure-based finite volume approach is utilized to solve the governing equations by the commercial software COMSOL Multiphysics 5.5. In such a numerical simulation, a reliable result should not be affected by the size of the element mesh. As shown in Fig. 2(a), structured hexahedral and unstructured tetrahedral grids are utilized in our computational domain. In consideration of the module size and the computation cost, the grid is initially set as 0.00796 m and is encrypted. The module geometry and operating conditions of the simulations, in which the transverse pitch is equal to the diagonal pitch 1.25 times the cell diameter ( $S_T = S_D = 1.25 D$ ), the inlet air temperature is 288.15 K, the inlet air velocity is 0.5 m/s and the current is 100 A, are established [3,7]. Fig. 2(b) presents the simulation results obtained for the outlet air temperatures (x = 0.227 m, z = 0.031 m, y = -0.04, -0.03, -0.02, -0.01 and 0 m)



**Fig. 3.** Comparisons between the simulation results and the correlation values for Nu and f at different values of Re.

with different element grid sizes. As presented, the temperature difference between the grid sizes of 0.001 m and 0.0005 m is below 1.41%. Therefore, the grid size of 0.001 m is selected in the consideration of the precision and computation costs.

#### 3.2. Model validation

To verify the validity of the present computational model, a comparison study between the numerical results and available data is performed. For the numerical simulations, the initial temperature of the supercapacitor is 298.15 K, the inlet air temperature is 288.15 K and the module structure is  $S_T = S_L = 2$  D. The inlet air velocity changes from 0.1 m/s to 1 m/s, thus, the Reynolds number (Re) varies between 1050 and 10,501. The comparison study has been performed by comparing the computed Nu and friction factor (f) with those predicted by the theoretical correlations [34,35]. The comparison results for Nu and f at different Re are presented in Fig. 3. The errors between the CFD model and the theoretical correlation are calculated using the sum of the relative squared errors (Equation (26)). It can be seen that the present numerical predictions are in a good agreement with the corresponding correlation values, with the errors of 5.96% and 0.024% for Nu and f, respectively. In the light of the validation tests above, the mathematical model and numerical method can be adopted owing to the reliability.

$$\Gamma = \sum_{i} \left(\frac{y_i - \hat{y}_i}{y_i}\right)^2 \tag{26}$$

where *i* is the number of measurements;  $y_i$  is the theoretical value;  $y_i$  is the predicted value.

#### 3.3. Structural optimization based on entropy generation analysis

In most supercapacitor cooling cases, space requirements are important parameters. Wilson et al. [36] studied two longitudinal rows of tubes, in which the ratio of longitudinal pitch and transverse pitch of less than 1 ( $S_L/S_T < 1$ ) is recommended for the staggered arrangement to reduce the friction factor and enhance the heat transfer. Thereby, eight geometries of staggered supercapacitor module ( $S_T = S_D = 1.25$ , 1.5, 1.75 and 2 D;  $S_T = S_L = 1.25$ , 1.5, 1.75 and 2 D) are studied, accounting for the compactness as well as heat transfer performance. After determining the structural parameters, the entropy generation analyses on the supercapacitor



Fig. 4. Heat transfer entropy generation contours of supercapacitor modules for the configuration with eight different geometries.



Fig. 5. Fluid friction entropy generation contours of supercapacitor modules for the configuration with eight different geometries.



Fig. 6. Bejan number contours of supercapacitor modules for the configuration with eight different geometries.

module with different geometries are carried out. The inlet air temperature is 288.15 K, the inlet air velocity is 0.5 m/s and the current is 100 A.

Fig. 4 presents the heat transfer entropy generation contours of supercapacitor modules with different geometries. It is observed that the heat transfer entropy generation decreases when the space between the cells becomes larger. The decrease in pitch increases the temperature gradient and subsequently causes greater heat transfer entropy generation. The entropy generation due to the heat transfer is concentrated in the region near the wavy supercapacitor shells, where the temperature gradient is larger. The heat transfer entropy generation increases from near zero at the center of the channel to a maximum value around the wavy shell. The reason is the insignificant temperature and velocity gradients at the centerline, and the high values of such gradients near the supercapacitor shells. The comparisons indicate that the lowest entropy generation due to thermal effects is obtained for the geometry described by  $S_T = S_L = 2 D$ .

Fig. 5 shows the fluid friction entropy generation contours of supercapacitor modules with different geometries. As it is observed, an increase in space between cells leads to the reduction of the fluid friction entropy generation. The increase in the pitch leads to the velocity gradients in the flow field become more gradual and, subsequently, fluid friction entropy generation reduce. The entropy generation due to viscous effects is significant in the region near the wavy supercapacitor shells, where the velocity gradient is much considerable. The results also indicate that the

lowest frictional irreversibility is obtained for the geometry described by  $S_T = S_L = 2$  D.

Fig. 6 presents the Bejan number contours at different geometries, which is an indicator of the entropy generation due to heat transfer effects. The values of the Bejan number are symmetrically distributed along the centerline. The fluid friction entropy generation is the main term in the front of the flow field while heat transfer entropy generation becomes dominant in most parts of the domain. Moreover, heat transfer entropy generation is the main term near the wavy shells and in the rear of the cells. The contribution of the heat transfer entropy generation being dominant in this region. In consideration of the reduction of heat transfer and fluid friction irreversibilities, the geometry of  $S_T = S_L = 2$  D is adopted in this study.

# 3.4. Cooling liquid optimization based on entropy generation analysis

Fig. 7 presents the results for the entropy generations, *Nu*, and ECOP versus the inlet temperature for different cooling liquids. In consideration of the cooling performance and the irreversibilities, the air and water are chosen as the cooling media with the inlet temperatures of 278.15 K, 280.15 K, 283.15 K, 285.15 K, 288.15 K, 291.15 K, and 293.15 K. The inlet velocity is 0.1 m/s and the current is 100 A. The variations of the entropy generations of heat transfer and fluid friction for the air and water at different inlet



Fig. 7. The variations of (a) the heat transfer entropy generation, (b) the fluid friction entropy generation, (c) Nu, and (d) ECOP versus the inlet temperature for the different cooling liquids.



Fig. 8. The variations of (a) the heat transfer and fluid friction entropy generations and (b) Nu and ECOP via the current.

temperatures are presented in Fig. 7(a) and (b). The results show that the entropy generation of water cooling is higher than that of air cooling. Moreover, as temperature increases, the heat transfer entropy generation decreases while the fluid friction entropy generation shows no apparent fluctuations with temperatures. And the differences between the water and air heat transfer entropy generations also decrease with the increase in temperature, while the differences of fluid friction entropy generations stay the same.

Apart from the irreversibilities, heat transfer performance is another concern. The comparisons in Nu of air cooling and water cooling via the temperature are depicted in Fig. 7(c). The Nu of water cooling is larger than that of air cooling, owing to the stronger disturbance and thus a higher heat transfer coefficient of water. The trends in *Nu* values of air and water with changes in the temperatures are similar to Fig. 7(b). Fig. 7(d) shows the ECOP versus the temperature of air and water. As seen, the ECOP goes up via increasing the inlet temperature and the enhancement is more significant for temperatures in the range of 288.15 K-293.15 K. There is a maximum value for ECOP at 293.15 K for both air and water. Therefore, in consideration of the heat transfer and fluid friction irreversibilities, Nu and ECOP, the optimal cooling approach should be based on the water cooling with the inlet temperature of 293.15 K. Thus, the water with the temperature of 293.15 K is selected as the cooling liquid in the following analyses.

#### 3.5. Effects of the current

Fig. 8 presents the effect of the current on the entropy generations, Nu and ECOP at the range of 50 A–200 A. The inlet water

velocity is 0.5 m/s and the inlet water temperature is 293.15 K. From the 3D thermal model for a single cell, it is noted that the larger charging and discharging current would lead to the larger heat generation, thus the higher temperature of the supercapacitor. As shown in Fig. 8(a), the heat transfer entropy generation increases slightly with an increase in the current, while the fluid friction entropy generation decreases 0.236% with the current increasing from 50 A to 200 A. The reason is that the effect of temperature variations of supercapacitors on the flow field is too small to cause a change in the velocity gradient. Similar to the fluid friction entropy generation, *Nu* and ECOP also go down slightly with the increase in the current (Fig. 8(b)). The results show that the current has no apparent effect on the irreversibilities and heat transfer performance of the water-cooled supercapacitor module.

#### 3.6. Effects of the inlet water velocity

Fig. 9 portrays the effects of inlet water velocity on the entropy generations, *Nu* and ECOP. The current is 200 A, the inlet water temperature is 293.15 K, and the inlet water velocity ranges from 0.1 m/s to 1 m/s. The results suggest that the inlet water velocity plays a key role in the supercapacitor cooling system. Fig. 9(a) shows that the heat transfer entropy generation goes up via ascending the inlet water velocity and there is a maximum value for heat transfer entropy generation at the velocity of 0.5 m/s. Then it gradually decreases as the velocity of water increasing from 0.6 m/s to 1 m/s. It can be concluded that when the inlet water velocity is 0.5 m/s, the temperature gradient would be the most significant, thus the largest heat transfer irreversibility. The fluid friction



Fig. 9. The variations of (a) the heat transfer and fluid friction entropy generations and (b) Nu and ECOP via the inlet water velocity.

entropy generation variations via the inlet water velocity exhibit like an exponential curve, where the slope becomes sharper with the increase in the inlet water velocity. It is noticeable that increasing inlet water velocity is undesirable whereas increasing the entropy generation is undesirable. The effects of the inlet water velocity on *Nu* and ECOP are illustrated in Fig. 9(b). As it is observed, an increase in the inlet water velocity leads to the increasing of *Nu* and ECOP.

#### 4. Conclusion

In this paper, the entropy generation analysis based on a detailed coupled model has been implemented for the first time to evaluate thermodynamic irreversibilities within the supercapacitor modules. The coupled model consists of a 1D electrochemical model, a 3D thermal model for a single cell and a 3D forced convective heat transfer model for a supercapacitor module, which can be used to accurately predict the transport of species, heat generation and heat transfer of supercapacitor at cell and module levels. The present model results show a good agreement with theoretical Nu and f, with the errors of 5.96% and 0.024%, respectively. The entropy generation analysis is conducted once the results of CFD simulations for the supercapacitor module are obtained. In particular, four evaluation and optimization criteria based on energy and entropy generation analyses, namely heat transfer entropy generation, fluid friction entropy generation, Nu and ECOP, are proposed to evaluate the thermodynamic irreversibilities and heat transfer performance of supercapacitor thermal management schemes. The effects of structural geometries, cooling liquids, inlet temperature, current and inlet water velocity have been investigated and then optimized. The following results have been obtained:

- (1) Entropy generation due to heat transfer effects is dominant in most parts of the flow field compared with fluid friction effects. A higher value of Be is obtained near the wavy shells and in the rear of the cells. Moreover, the contribution of the heat transfer entropy generation reduces near the centerline and the fluid friction entropy generation dominates in this region. In consideration of the reduction of heat transfer and fluid friction irreversibilities, the geometry described by  $S_T =$  $S_L = 2$  D is the optimal.
- (2) The heat transfer entropy generation of water cooling is larger than that of air cooling, and the value of them, as well as the differences between them, decrease with the increase in temperature. The fluid friction entropy generation and *Nu* of air cooling are lower than that of water cooling, and their values show no changes via the temperature. ECOP enhances with increasing temperature and the differences between water and air become more significant. Thus, the water with the temperature of 293.15 K is chosen as the cooling liquid taking the irreversibilities and heat transfer rate into account.
- (3) The current has no apparent effect on the irreversibilities and heat transfer characteristics of the water-cooled supercapacitor module.
- (4) The heat transfer entropy generation increases with increasing the inlet water velocity in the range of 0-0.5 m/s and it reduces with inlet water velocity increasing from 0.6 m/s to 1 m/s. The enhancement in fluid friction entropy generation becomes more prominent with increasing inlet water velocity. And an increase in inlet water velocity leads to an increase in *Nu* and ECOP.

efficient supercapacitor thermal management systems with lower irreversibilities and higher heat transfer rates. It is found that under the requirement of compactness, the supercapacitor module with larger longitudinal pitch would lead to the smaller entropy generation. And under the premise of satisfying the cooling demand, increasing the inlet temperature of cooling liquid would result in the reduction of the entropy generation. In the future, we anticipate that many studies can be related to this issue such as optimization analysis based on artificial neural networks to find the optimum thermal management schemes for supercapacitor modules, as well as other energy conversion and storage systems.

#### Author statement

**Haowen Li**: Conceptualization; Methodology; Investigation; Formal analysis; Data curation; Writing – original draft. **Huachao Yang**: Methodology; Writing – review & editing. **Chenxuan Xu**: Writing – original draft; Visualization. Jianhua Yan: Resources. **Kefa Cen**: Supervision. **Kostya (Ken) Ostrikov**: Writing – review & editing; Supervision. **Zheng Bo**: Supervision; Resources; Writing – review & editing.

#### **Declaration of competing interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### Nomenclature

i <sub>s</sub>	Solid phase current density
<i>i</i> 1	Liquid phase current density
δ	Surface area per unit volume of the electrode
$\varphi_{s}$	Solid electrical potential
$\varphi_1$	Liquid electrical potential
C <sub>dl</sub>	Capacitance per interfacial area
$\sigma_{\rm s}$	Solid phase conductivity
$\Sigma_{elec}$	Conductivity of electrodes
$\varepsilon_{ele}$	Porosity of electrodes
$\varepsilon_{sep}$	Porosity of separator
lsep	Thickness of separator
lneg	Thickness of negative electrode
lpos	Thickness of positive electrode
Î <sub>c</sub>	Thickness of collector
la	Thickness of aluminum shell
β	Bruggeman porosity exponent
F	Faraday's constant
R	Universal gas constant
Di	Diffusion coefficient
ei	Charge
Ci	Ion concentration
u <sub>i</sub>	Migration rate
i <sub>app</sub>	Applied current density
q	Heat generation per unit volume

The outcomes of this work are useful in the development of

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2	Padial thermal conductivity
Λr	
Λ <sub>Z</sub>	
	lemperature
н	Height
r <sub>o</sub>	External radius
r <sub>i</sub>	inner radius
Q <sub>Joule</sub>	Heat generation due to Joule effect
Qentropy	Heat generation due to entropy change
1	Current
k <sub>B</sub>	Boltzmann constant
$d_{\rm EDL}$	Thickness of effective electric double layer
С	Capacitance
$V_0$	Total electrolyte volume
C <sub>negative</sub>	Areal capacitance of negative electrode
C <sub>positive</sub>	Areal capacitance of positive electrode
T <sub>amb</sub>	Ambient temperature
$\lambda_{a}$	Thermal conductivity of aluminum
ρ	Density
Cp	Specific heat capacity
u, v, w	Velocity in the x, y and z coordinates
ν	Kinematic viscosity
$F_{\rm x}, F_{\rm v}, F_{\rm z}$	Volume force in the x, y and z coordinates
J	<sub>a</sub> Heat flow
Jĸ	Diffusion flow
φ <sub>k</sub>	Thermodynamic potential
$F_k$	External force
П	Momentum flows tensor
Ii	Chemical reaction rate
A <sub>i</sub>	Chemical affinity
Seenh	Heat transfer entropy generation
Seen f	Fluid friction entropy generation
u u	Dynamic viscosity
Ве	Beian number
Nu	Nusselt number
ST	Transverse pitch
Sn	Diagonal pitch
S	Longitudinal pitch
D	Diameter of supercapacitor cell
f	Friction factor
J	

Re Reynolds number

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# Experimental Investigation of a Thermally Responsive Actuator Based on Metallic Molybdenum Disulfide: A Conceptual Analysis

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Actuators that convert external stimuli to mechanical energy have aroused strong attention for emerging applications in robotics, artificial muscles, and other fields. However, their limited performance under harsh operating conditions evidenced by the low cycle life and hysteresis has restricted their practical applications. Here, a thermal-driven actuator based on layered metallic molybdenum disulfide  $(1T MoS_2)$  nanosheets is demonstrated. The active actuator film exhibits fully reversible and highly stable (>99.296% in 2700 cycles) thermal-mechanical conversion over a wide temperature window (from -60 °C to 80 °C). Importantly, *IT MoS*<sub>2</sub> film shows a fast response with the bending rate and the recovery rate of >1.090 rad  $s^{-1}$  and >0.978 rad  $s^{-1}$ , respectively. The assembled actuator can lift 20 times its weight over several centimeters for more than 200 cycles. This work, for the first time, demonstrates the thermoresponsive characteristics of 1T MoS<sub>2</sub> in developing the thermal actuator, which may open new opportunities for various applications, such as robotics, artificial muscles, and human assist devices. [DOI: 10.1115/1.4055152]

Keywords: actuation system, thermally responsive characteristics, metallic molybdenum disulfide, bending performances, energy efficiency, energy systems, thermal systems

#### 1 Introduction

Innovative actuation devices that convert external stimuli (e.g., electric [1-4], solvent adsorption/desorption [5,6], heat [7-10], or light [11-13]) to mechanical energy have shown advantages over natural muscles in terms of the higher mechanical stresses and superior adaptability to complex environments [14,15]. To date, innovative actuation devices based on various mechanisms and advanced functional materials have been developed. The most widely used actuation devices are electroresponsive actuators (e.g., electric actuators [1,16–18] and electrochemical actuators [19–22]), which are driven by electric fields. For example, the dielectric elastomer actuators [16,23,24], based on the electrostatic interaction between conductive layers, are particularly attractive because of their large actuation strains. Albeit the advantages, high driven voltages (~1 kV), substantial hysteresis, and difficulty in synthesis have limited their practical applications. The liquid crystal elastomers [17], with significantly lower driven voltage, are restricted by the relatively low stiffness and high cost. The shape memory polymers (SMPs) [25] are also promising electrically induced materials considering their low manufacturing cost, good elasticity, and light weight. However, SMPs also suffer from a long response time, short cycle life, and unsatisfactory environmental stability. Recently, electrochemically driven actuators, including the organic conducting polymers [26-28], ionic polymer metal composites [29-31], and graphene materials [32,33], have been demonstrated with outstanding mechanical properties and large strokes. However, their reversibility cycle rates are restricted by the intrinsic conductivity of the materials. Another metallic molybdenum disulfide (1T MoS<sub>2</sub>) nanosheet-based electrochemical actuator that is based on the intercalation of ions exhibits low driven voltage, excellent reversibility, and stability [19]. Despite that, the actuation system suffers from high cost and heavy weight with counter electrodes, electrolyte, and containment system.

Considering the drawbacks of the electroresponsive actuators in practical applications, increasing attention has been paid to the thermal actuators, which feature low cost, fast response, high reversibility, and long cycle life [34-37]. Herein a thermalresponsive actuator based on the two-dimensional 1T MoS<sub>2</sub> film prepared by a versatile hydrothermal method is for the first time demonstrated and experimentally investigated. The as-prepared actuator exhibits fast thermal-mechanical response, high reversibility, low cost, and long cycle life. The bending performances of 1T MoS<sub>2</sub> films with different thicknesses at a wide temperature window of -60 °C to -80 °C are measured and compared. The 1T MoS<sub>2</sub> film presents the highly reversible and stable (recovery ratio >99.296%) and rapid response (bending rate >1.090 rad s<sup>-</sup> and recovery rate >0.978 rad  $s^{-1}$ ) over the 2700 cycles. Importantly, it is capable of lifting 20 times its weight over several centimeters during hundreds of cycles. In the end, the thermomechanical characteristics of the 1T MoS<sub>2</sub> film actuator are measured by the dynamic mechanical analysis (DMA) test.

#### 2 Methodology

2.1 Fabrication of 1T MoS<sub>2</sub> Film. 1T MoS<sub>2</sub> is synthesized by the customized hydrothermal method. Thirty-five milligram thioacetamide, 25 mg molybdenum oxide (MoO<sub>3</sub>), and 120 mg urea are dissolved in a solvent of 20 ml de-ionized water under magnetic stirring to form a homogeneous suspension. Then, the solution is transferred into a 40 ml polytetrafluoroethylene tank sealed with stainless steel autoclave, maintaining at 180 °C for 18 h. Next, the reaction is terminated by rapidly cooling the solution to room temperature by removing the autoclave from the oven. The resultant sample is mixed with de-ionized water to form a dispersion with a concentration of  $1 \text{ mg ml}^{-1}$ . The solution is ultrasonicated for 1 h to agitate the exfoliation and is centrifuged several times with de-ionized water to remove residual salts. 1T MoS<sub>2</sub> films are prepared by filtering the diluted suspension through a polyvinylidene difluoride (PVDF) membrane (millipore, 25 nm pore size), the thicknesses of which can be controlled by adjusting the volume of the solution.

**2.2** Materials Characterization. Scanning electron microscope (SEM; Hitachi SU-8010) is utilized to characterize the morphology and structure of 1T  $MoS_2$  with an acceleration voltage of 8 kV. The Raman spectra are collected by a Raman spectrometer (LabRAM HR Evolution) using an argon laser beam with an excitation wavelength of 532 nm. Surface composition is analyzed by X-ray photoelectron spectroscopy (XPS) using a VG ESCALAB MARK II spectrometer.

**2.3 Bending Performance Measurements.** To track the displacement and bending curvature, the thermoresponsive deformation tests are performed. For the bending tests in the high temperature range of 20–80 °C, the bilayer films (1T MoS<sub>2</sub>/substrate) with a dimension of  $\phi$ 50 mm in different thicknesses are prepared. The films are then placed on a heating stage within the vacuum drying oven to measure the bending angle by applying a temperature increment rate of 1 °C min<sup>-1</sup>. For the bending tests in the low temperature range –60 °C to 10 °C, the prepared films are placed in the high-low temperature test chamber by applying a temperature decrement rate of 1 °C min<sup>-1</sup>. The changes of the bending angle, produced as a thermal response, are recorded by a digital camera every 5 °C with a resolution of 0.01 deg.

For the dynamic bending tests from 40–80 °C, the bilayer film with a dimension of  $\phi$ 50 mm is prepared and placed in a dish on the heating stage to measure the bending angle. Once the bending curvature reaches stable, the film is immediately transferred to the dish held at room temperature (~14 °C), where it recovers its original state, and so forth. For the dynamic bending experiments in the range of -60 °C to 10 °C, the prepared bilayer film is transferred from the high-low temperature test chamber to the dish held at

room temperature. The entire process is recorded by a digital camera, especially the thermal response time is recorded along with the bending angle of the film.

To investigate the thermomechanical behavior of the 1T  $MoS_2$ -based film, a DMA test is performed on a DMA analyzer (Q800, TA Instrument Inc., New Castle, DE). The test is performed in the tension film mode on a strip sample with dimensions of 40 mm × 7 mm × 0.11 mm. The sample is first heated to 35 °C and stabilized for 10 min to reach thermal equilibrium, and then the temperature increases from 35 °C to 85 °C at a rate of 2 °C min<sup>-1</sup>. A 0.5% oscillating strain is applied to the samples.

#### **3** Results and Discussion

**3.1** Characterization of the 1T  $MoS_2$  Film. As depicted in Fig. 1(*a*), the thin flake comprises an active layer (1T  $MoS_2$ ) deposited on a substrate (i.e., the PVDF beam). When heated, the film bends towards the 1T  $MoS_2$  side; when cooled, it bends in the reverse direction thus making a thermoresponsive circle. The interactions between the nanosheets of 1T  $MoS_2$  initiate the bending motion in response to heating and cooling. The micromorphology of the  $MoS_2$  layer is characterized by SEM (Fig. 1(*b*)). A high-magnification cross-sectional image of the film reveals the well-packed layered structure and the thickness of  $MoS_2$  film.

Raman spectroscopy is performed to confirm the crystalline structure of the MoS<sub>2</sub> films. Figure 1(*c*) shows the Raman spectra of 1T MoS<sub>2</sub> and semiconducting molybdenum disulfide (2H MoS<sub>2</sub>), respectively. A distinctive Raman peak is observed at 153.2 cm<sup>-1</sup>, attributed to Mo–Mo stretching vibrations in 1T MoS<sub>2</sub>. The other three Raman peaks (i.e., ~229.5 cm<sup>-1</sup>, 326.6 cm<sup>-1</sup>, and 405.0 cm<sup>-1</sup>) are also associated with the phonon modes in 1T MoS<sub>2</sub>, which are substantially different from those of 2H MoS<sub>2</sub> [38].

The detailed elemental composition and the phase identification on the surface of  $MoS_2$  film are further characterized by XPS. Highresolution XPS from the Mo 3d region (Fig. 1(*d*)) and the S 2p region (Fig. 1(*e*)) of 1T MoS<sub>2</sub> film are presented. The Mo 3d spectrum shows two characteristic peaks and a small peak at the binding energies of 228.6 eV, 231.8 eV, and 226.2 eV corresponding to Mo  $3d_{5/2}$ , Mo  $3d_{3/2}$ , and S 2s, respectively. The Mo 3d peaks of 1T MoS<sub>2</sub> shift to lower binding energies by 0.9 eV with respect to the corresponding peaks in 2H MoS<sub>2</sub>. Similarly, the S 2p peaks are located at 161.5 eV and 162.6 eV associated with S  $2p_{3/2}$  and S  $2p_{1/2}$ , respectively. It has been observed that the S 2p peaks from the 1T phase appear at a binding energy that is 1 eV lower than for their 2H counterparts. Deconvolution of the Mo 3d and S 2p regions of chemically exfoliated MoS<sub>2</sub> indicates that the 1T phase concentration of the nanosheets is ~70% [39,40].

**3.2** Dynamic Thermoresponsive Bending Property. As presented in Figs. 2(a)-2(f), the film features the thermoresponsive bending property. Under the thermal stimulus, the film bends towards the 1T MoS<sub>2</sub> side when heated, and the higher the temperature, the larger the bending angle. The film bends in the reverse direction when cooled, the lower the temperature, the larger the bending angle. Through the bending tests, it is found that the film thermoresponsive bending phenomenon begins at  $-60 \text{ }^{\circ}\text{C}$  and ceases at 80 °C, thus the responsive temperature window of the 1T MoS<sub>2</sub>-based film is from  $-60 \text{ }^{\circ}\text{C}$  to 80 °C. Meanwhile, the bending angles of the substrate at different temperatures are also measured, and the results show that there is no deformation versus temperature. The comparisons of the bending performances reveal that the bending curvature is induced by the expansion and contraction of 1T MoS<sub>2</sub> nanosheets.

The bending curvatures of the 1T MoS<sub>2</sub>/substrate films (with different thicknesses of 4  $\mu$ m, 5  $\mu$ m, 7  $\mu$ m, 9  $\mu$ m, and 10  $\mu$ m 1T MoS<sub>2</sub> film) via temperatures are presented in Fig. 2(g). The resulting curves coincide in the temperature range from -60 °C to 80 °C. The bending curvatures of the films with different thicknesses



Fig. 1 (a) Schematic illustration of the thermoresponsive characteristics of 1T MoS<sub>2</sub>-based film, (b) side view of the 1T MoS<sub>2</sub> observed by SEM showing the layered nature of the film and the high-magnification image of restacked 1T MoS<sub>2</sub> nanosheets, (c) Raman spectroscopy of 1T MoS<sub>2</sub> and 2H MoS<sub>2</sub>. High-resolution XPS spectra of (d) Mo 3d and (e) S 2p for 1T MoS<sub>2</sub> and 2H MoS<sub>2</sub>.

increase from  $-0.266 \text{ cm}^{-1}$  to  $0.282 \text{ cm}^{-1}$  with the temperature increasing from  $-60 \text{ }^{\circ}\text{C}$  to  $80 \text{ }^{\circ}\text{C}$ . Furthermore, the rate of increment tends to slow down when the temperature reaches  $-50 \text{ }^{\circ}\text{C}$  and  $70 \text{ }^{\circ}\text{C}$ . At the same temperature, the absolute value of the film bending curvature increases slightly with the increase of the

1T MoS<sub>2</sub> film thickness. However, the difference between the curvatures between the 4  $\mu$ m and 10  $\mu$ m thick 1T MoS<sub>2</sub> film is <8.16% throughout the entire temperature range, indicating that the thicknesses of films have little effect on the bending curvatures. The reason is that the van der Waals force between the nanosheets is



Fig. 2 Images of curvature induced by thermal stimulus in 1T MoS<sub>2</sub> film on substrate at (a) -50 °C, (b) -30 °C, (c) -10 °C, (d) 40 °C, (e) 65 °C, (f) 80 °C (scale bar, 1 cm). (g) Curvatures versus temperature of films with different thicknesses and (h) curvatures versus temperature during heating and cooling.

too weak to cause the difference in the bending curvatures of different thicknesses. In consideration of the flakes with different thicknesses sharing the same bending characteristics, the flake loaded with  $10 \,\mu\text{m}$  1T MoS<sub>2</sub> film is selected for the following tests and applications.

The film is also fully reversible over the temperature ranges. The reversible bending curvature towards and outwards the 1T MoS<sub>2</sub> direction is attributed to the strain induced by the expansion and contraction of the 1T MoS<sub>2</sub> nanosheets. In Fig. 2(*h*), the notations a, b, c, d, e, and f represent the bending curvatures at the temperatures indicated by Figs. 2(a)-2(f), respectively. It can be seen that the curvature gradually increases with the increase in temperature, and the curvature gradually decreases with the decrease in temperature. In particular, the bending curvatures have little deviations (<0.0214 cm<sup>-1</sup>) at any temperature during the heating and cooling process, which reveals that the film is reversible when heated and cooled throughout the studied broad temperature ranges.

For the reversibility and stability tests at high temperatures (40-80 °C), the film is placed on a heating stage to measure the bending angles. Temperatures of the heating stage change from room temperature to high temperatures (40, 45, 50, 55, 60, 65, 70, 75, and 80 °C) by applying a temperature increment rate of 1 °C min<sup>-1</sup>. The tests at low temperatures (-60 °C to 10 °C) are carried in the high-low temperature test chamber. The temperatures in the test chamber change from room temperature to low temperatures (-60, -50, -40, -30, -20, -10, 0, and 10 °C) by applying a temperature decrement rate of 1 °C min<sup>-1</sup>. The process is repeated five times at each temperature. The bending curvature changes with respect to the temperatures are presented in Figs. 3(a) and 3(b). The results show that under any specified temperature, the bending curvatures are constant within the cycles, and the deviation is below 5.19%, indicating its stability and reversibility within the temperature ranges studied. In addition, the 1T MoS<sub>2</sub>-based film is stable for up to eight weeks, as shown in Figs. 3(c) and 3(d)for the temperature ranges of 40 °C to 80 °C and -60 °C to 10 °C, respectively. In both cases, the film exhibits a stable continuous bending curvature for eight weeks.

Besides the stability, the 1T MoS<sub>2</sub> film demonstrates the fast response characteristic in bending and recovering. For the dynamic bending tests at high temperatures (20–80 °C), the film quickly bends towards the 1T MoS<sub>2</sub> side once transferred to the dish held on a heating stage. When the bending curvature reaches equilibrium, the film is immediately returned to the dish recovering the original state at room temperature. For the dynamic bending test at low temperatures (-60 °C to 10 °C), the film is transferred from the high-low temperature test chamber to the dish maintained at room temperature. To obtain the bending rate, the thermal response time ( $t_b$ ) is recorded along with the bending angle ( $\theta_b$ ) of the film. To obtain the recovery rate, the recovery time ( $t_r$ ) is recorded along with the recovery angle ( $\theta_r$ ). The bending rate and recovery rate can be defined as follows:

The bending rate:

$$v_b = \frac{\pi \cdot \theta_b}{180 \cdot t_b} \tag{1}$$

The recovery rate:

$$v_r = \frac{\pi \cdot (\theta_b - \theta_r)}{180 \cdot t_r} \tag{2}$$

Figure 4(*a*) shows the curves of the bending and recovery rates versus temperatures for the 1T MoS<sub>2</sub>-based film. It can be seen that the bending rate and recovery rate are >1.090 rad s<sup>-1</sup> and >0.978 rad s<sup>-1</sup>, respectively, which demonstrates the fast response of the film to the thermal stimulus. The bending and recovery rate increase with temperature increasing from  $-60 \,^{\circ}\text{C}$  to 20  $^{\circ}\text{C}$ , while decreasing with the increase in temperature in the range of 30–80  $^{\circ}\text{C}$ . Figure 4(*b*) shows the film shape recovery ratio as a function of temperatures. The definition of the recovery ratio is as follows. The shape recovery ratio is not sensitive to the temperature, maintaining between 99.296% and 99.615%. The results derived from the calculation of the recovery ratio also reveal the



Fig. 3 The curvature at different temperatures, demonstrating stability over the range of temperatures: (a) 40–80 °C and (b) –60 °C to 10 °C. Cycling stability of 1T  $MoS_2$ -coated film over the range of temperatures: (c) 40–80 °C and (d) –60 °C to 10 °C. The film exhibits stable continuous bending curvature for eight weeks.

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Fig. 4 (a) The bending and recovery rate of the film under different temperatures, and (b) the recovery ratio of the film versus temperature.

reversibility and stability of the 1T MoS<sub>2</sub>-based films.

$$R_r = \frac{\theta_b - \theta_i}{\theta_b} \times 100\% \tag{3}$$

3.3 Thermal Actuation Behavior. To further demonstrate the actuation behavior of the 1T MoS<sub>2</sub>-based film, a working actuator is designed and developed. The actuator consists of two 1T MoS<sub>2</sub>--based beams attached end to end via Cu strips, in which the active actuator films are applied to the opposite sides of the beams, as shown in Fig. 5. This design ensures each film bend with an equivalent magnitude to cancel out the rotational angle of each, so that the weights can be vertically elevated. With this configuration, under the fixed load, the actuator behaves as a low cost and light actuator with no additional parts. The 10  $\mu$ m 1T MoS<sub>2</sub> film is applied to each beam, with each film weighing approximately 20 mg. To demonstrate the vertical lifting, we suspend a 1000 mg weight from the actuator and initiate the heating and cooling cycles. It can be seen from the photographs in Fig. 5(b) that the 1T MoS<sub>2</sub> actuator is able to lift objects more than 20 times heavier than its own weight. The weight can be lifted up to 0.8 cm when the temperature changes from 40 °C to 80 °C. We measure hundreds of (>200) cycles and find that the actuation device is able to repeatedly and stably elevate and lower the weight. These results demonstrate the potential applications in artificial muscles and soft actuators.

3.4 Thermomechanical Behavior. To investigate the thermomechanical behavior of the actuator film in the range of 40-80 °C, a dynamic mechanical analysis test is performed using a DMA analyzer (Q800, TA Instrument Inc., New Castle, DE), by increasing temperature from 35 °C to 85 °C at a ramp rate of 2 °C min<sup>-</sup> Figure 6(a) shows the variations of storage modulus (corresponding) to elastic response) via the temperatures, increasing from  $\approx 65$  MPa at high temperature (80 °C) to ≈100 MPa at low temperature (40 °C). The temperature-dependent stiffness variations of the film are further examined by conducting uniaxial tensile tests with a strain rate of  $0.0005 \text{ s}^{-1}$ . The Young's modulus is measured as the initial slope of the material stress-strain curve in the linear regime (up to 2% strain). The stress-strain curves indicate that the actuator film exhibits a linear stress-strain behavior with a modulus of 64.7 MPa at 80 °C, and its stiffness dramatically increases to 98.6 MPa when the temperature drops to 40  $^{\circ}\mathrm{C}$ (Fig. 6(b)). The stiffness variations observed from the quasi-static uniaxial tensile tests agree well with the results from DMA tests. The 1T MoS<sub>2</sub>-based films present mechanical characteristics that are better than the skin (Young's modulus of ~0.2 MPa) and comparable to muscles (Young's modulus of  $10^{-2}$ – $10^3$  MPa) [16,41,42].

#### 4 Conclusion

In summary, this work for the first time presented the practical demonstration of the highly stable, fully reversible, fast response,







Fig. 6 (a) The storage modulus of the film under different temperatures and (b) stress-strain curves of the film at 40 °C and 80 °C.

low cost, and easy-to-fabricate thermal-driven actuators, benefiting from the unique thermoresponsive properties of 1T MoS<sub>2</sub> film. The bending performances of the 1T MoS<sub>2</sub>-based film are demonstrated upon exposure to a thermal stimulus (-60 °C to 80 °C). The bending curvatures of the films with different thicknesses increase with the increase of the temperature following the nearly linear trend, and the thickness plays no significant effect on the bending characteristics. Moreover, the bending curvature deviations are <5.190% with the recovery ratios >99.296% over 2700 cycles at different temperatures, and the bending rate and the recovery rate are >1.090 rad  $s^{-1}$  and >0.978 rad  $s^{-1}$ , respectively. These results suggest the stable, reversible, and rapid behaviors of the 1T MoS<sub>2</sub> film. Owing to the thermoresponsive characteristics, an actuator device is designed and developed. The device is based on the 1T MoS<sub>2</sub> film equipped with Cu tape, which can lift objects of more than 20 times its weight up to 0.8 cm over hundreds of cycles. The actuator film also reveals thermomechanical characteristics that are better than the skin and muscles. The measured Young's modulus is 64.7 MPa at 80 °C and 98.6 MPa at 40 °C. The thermoresponsive characteristics and the low cost manufacturing of the device make it promising for the development of next-generation integrated thermal actuator systems. This work opens the framework to develop new 2D materials-based thermal actuators with low cost, long cycle life, fast response, high modulus, and high stability.

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#### **Conflict of Interest**

There are no conflicts of interest.

#### **Data Availability Statement**

The datasets generated and supporting the findings of this article are obtainable from the corresponding author upon reasonable request.

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**Research** Paper

# Numerical investigation on heat transfer characteristics of the plate air heater with variable channels and experimental validation

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HIGHLIGHTS

• The model of the novel plate air heater with variable channels was developed.

• The model takes into account the phase transitions in the flow.

• Numerical and experimental investigations are treated.

• The model presented in this study shows obvious advantages in the condensation process.

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#### ABSTRACT

This paper presents a novel plate air heater using steam as the high temperature fluid. The heat transfer characteristics of the air heater were numerical simulated in different steam and air volume flow rates with Fluent and a user-defined function (UDF) as well as verification by experiment. The results showed the variable channel structure of the new plate air heater promptly condensed liquid discharge, reduced the thickness of the steam film and improved the speed of condensing steam. In this experimental model, when the temperature of the steam side reached 120 °C, the flow rate was 12  $m^3 \cdot h^{-1}$  and the air side flow was 480 m<sup>3</sup>  $\cdot$  h<sup>-1</sup>. The total heat transfer coefficient was approximately 70 W (m<sup>2</sup>  $\cdot$  K)<sup>-1</sup> and the drop in pressure of the steam side was less than 1 kPa.

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1. Introduction

Air heaters have been widely used in many industrial areas such as chemical, tobacco, food processing power systems, and other engineering applications [1,2]. Nowadays, the interest in the market of two-phase air heater is rising, due to the presence of latent heat from vaporization and that the heat transfer coefficient of two-phase air heater is much larger than that of single phase. Various types of air heaters have been used in engineering applications such as shell-and-tube, plate fin, finned tube and plate. Studies depict the most widely used air heaters are the finned tube [3,4]. However, these heaters have disadvantages such as their incompact structure, high production costs and that they occupy a large space. Further, during the process of heat exchange, they may waste latent heat because of incomplete condensation.

To date, researchers have successfully studied different types of air heater. Based on the similarity theory, Zhou [5] studied the heat transfer characteristics and corrosion resistance of a flat plate air

\* Corresponding author. E-mail address: ljwu@tongji.edu.cn (L. Wu). plate air heaters were significantly affected by the temperature of the inlet steam and the velocity of the steam and fluid, providing an important reference for the design and manufacture of plate heaters in the future. Khoshvaght-Aliabadi et al. [6] studied the heat transfer and flow performances of the vortex-generator channel with different geometrical parameters using numerical simulations and experimental validation. The correlations were derived for the Nusselt number of the vortex-generator channels plate-fin heat exchanger. Zeng et al. [7] carried out an experiment investigating the pressure drop and heat transfer characteristics in a plate-fin iron air heater by changing the mass flow rate and inlet temperature of flue gas. The heat transfer correlation was obtained with the Genetic Algorithm (GA) method by separating coefficients of the overall heat transfer coefficient. The conclusions they drew will be helpful for future development and design of a highefficiency plate-fin heat preheater. Li [8] conducted a theoretical and experimental study on heat and mass transfer characteristics of a basic plate heat exchanger, analyzing the different geometric parameters, flow parameters and dirt types of the heat exchanger and provided a theoretical basis for the practical application and

heater with an air curtain. They concluded that the properties of







#### Nomenclature

Symbols		h	heat transfer coefficient $(W/(m^2 \cdot K))$
p	density (kg/m <sup>3</sup> )	$\Delta P$	pressure drop (Pa)
λ	thermal conductivity (W/(m · K))	j	Colburn factor
$C_p$	specific heat at constant pressure $(kJ/(kg \cdot K))$	f	Finning friction factor
μ	dynamic viscosity (kg/(m · s))	Re	Reynolds number
k	thermal conductivity (W/(m · K))	Nu	Nusselt number
$G_k$	producing item of $k$ by average velocity gradient	Pr	Prandtl number
3	turbulent dissipation rate		
$\sigma_k$	Prandtl numbers corresponded to $k(-)$	Subscrip	ots
$\sigma_{arepsilon}$	Prandtl numbers corresponded to $\varepsilon$ (–)	wall	wall
$C_{1\varepsilon}$	empirical constant	х	span wise coordinate
$C_{2\varepsilon}$	empirical constant	у	normal coordinate
$S_m$	source term	Z	stream wise coordinate
$S_k$	user-defined source term	Ι	component on x, y, z coordinates
$S_{\omega}$	user-defined source term	ref	reference
Φ	thermal energy converted by the mechanical energy on		
	effect of viscous effect (kJ/kg)		

design of a plate heat exchanger. Wu et al. [9] studied the influence of plate heat exchanger channel structure and flue gas velocity on heat transfer characteristics, getting the highest heat transfer coefficient of plate heat exchanger 50 W ( $m^2 \cdot K$ )<sup>-1</sup> when the gas velocity was 8 m/s. Qin et al. [10] applied numerical simulation analysis on heat transfer and flow processes of a corrugated plate air heater. They studied the effect of the structure characteristics of a corrugated plate air heater, with the corrugated inclination angle  $\beta$ , wave height H, and the corrugated pitch and plate spacing. The heat transfer performances of a plate-fin catalytic combustion heat exchanger (PFCCHE) were investigated by Wang and Wang [11], through a numerical simulation and experiment. They drew the conclusion that plate-fin heat exchanger tended to reduce the heat resistance compared with tube-fined heat exchangers.

With the research on heat transfer characteristics of different air heaters intensifying, studies on two phase heat exchanger are naturally progressing. Zeng et al. [12] firstly introduced the definition of two-phase flow, the flow pattern and related parameters, then summarized the mathematical model and heat transfer calculation, analyzing the flow characteristics of two-phase flow in different devices. Huang et al. [13] researched the condenser shell side flow process of gas-liquid two-phase flow in a power plant. They then completed a numerical simulation and made comparisons between the simulation results and calculation results. Ramos et al. [14] applied theoretical analysis on the heat transfer characteristics of a heat exchanger equipped with thermosiphons, with a single-pass flow on the air side and a two-phase flow on the water side. They made numerical calculations as well as an experimental set-up, and compared the Computational Fluid Dynamics (CFD) results with the experimental ones, finding that the modeling predictions were within 10% deviation of the experimental results.

In the noted literature, there are few studies analyzing the heat transfer enhancement of plate air heater with variable channel structure. Also, a parametric study on plate air heaters has been not carried out when two-phase fluid is applied as a working media. Therefore, the design and development of a novel plate air heater blended from a number of types, shell-and-tube, plate fin and finned tube should be implemented. Further, more studies into flow and heat transfer of two-phase fluids are necessary. In this study, 3-D numerical simulations were carried out to provide a greater understanding of the mass and energy transformation of the novel plate air heater by using Fluent numerical calculation software. In addition to numerical simulations, an experimental study of heat exchange in a plate air heater was carried out. The first objective was to validate the feasibility and accuracy of both the physical model and the calculation model, by comparing the numerical simulation results with the experimental ones. The other objective was to evaluate the heat transfer and pressure drop performance of the variable channel structure plate air heater, when the flow rate and temperature of air and steam side were changed [15–17].

#### 2. Model description

#### 2.1. Novel plate air heater structure

Unconventional ideas in the design phase result in a novel air heater with a compact structure, a high heat transfer coefficient, high temperature resistance and bearing capacity, easy configuration, ease of cleaning and a low investment cost. The engineering applications demonstrate that this kind of air heater had superior performance.

Novel air heater plate beams are formed by stacking two pieces of the same plate, using a special process to form the elliptical or oval circulation channel. The inner loop direction channels are for water vapor, and two layer board beams are displaced in a dislocation mode, forming an air corrugated channel, with the flow of the two fluids being cross-flow. Fig. 1 displays the core part structure of the novel plate air heater.

The variable channel structure of the novel plate air heater is presented in Fig. 2(a). The elliptical circulation channels adopt the decreasing 4-3-2-1 distribution, which is equivalent to the decrease of the channel diameter. This design solves the problem



Fig. 1. Core architecture of the novel plate air heater.



Fig. 2. Variable channel structure of the novel plate air heater.

of the decline of flow velocity caused by a reduction in volume when the medium changed from gas to liquid. With the condensation of water vapor, the decreasing channel structure makes the condensed water flow rapidly, improving the velocity of the medium in the channel and enhancing the heat transfer. The consequence of the variable channel structure is illustrated in Fig. 2(b).

#### 2.2. Physical models

Selecting a representative physical model and applying appropriate boundary conditions in the CFD simulations are crucial to achieve the predicted characteristics of the novel plate air heater. Commercial software Gambit 2.4.6 was used to publish the geometry configuration. It is composed of five plates with each plate having 10 elliptical channels, with a 30 mm long axis and an 8 mm short axis. The length of the plate is 600 mm with a spacing of 15 mm. The elliptical circulation direct channels are for water vapor while the corrugated plate channels are for air creating a two way cross-flow. Fig. 3 and Fig. 4 display the physical model of the novel plate air heater and its geometric parameters.

#### 2.3. Mesh selection

The quantity and quality of mesh are important to the accuracy and the time of the CFD simulations. Therefore, we performed a grid independence study to determine the optimal mesh setting for these plates. The mesh selection was achieved by running the same numerical simulation with different mesh types and sizes and then comparing the accuracy of the results. Inlet temperature of the air-side and steam-side were set to 293 K and 393 K respectively, and the air-side mass flow rate was  $480\ m^3\cdot h^{-1}$  while steam-side was  $12\ m^3\cdot h^{-1}.$ 

The mesh for the air-side and steam-side were unstructured tetrahedral mesh. The mean temperature at the outlet for air side is exhibited in Fig 5. It can be seen that the grid cells at 8,329,473, 11,162,569, 13,327,549, 15,521,835 and 16,932,194 are mesh independent, because of the maximum error of the temperature remaining less than 2.041%. In addition, the heat transfer coefficient *h* and the pressure drop  $\Delta P$  of that 5 different mesh sizes are presented in Table 1. And according to the method proposed by Ref. [18], a mesh is characterized as "good" if the maximum skewness was lower than 0.7 for hexahedron and tetrahedrons. Considering both accuracy and workload, a grid quantity of 11162569 was used in all the numerical simulations.

#### 2.4. Boundary conditions

All numerical simulations were calculated by using the software FLUENT 6.3. The computational domain consists of a steam part and an air part. The applied boundary conditions in these parts are described as follows.

#### (1) In the air part

The Mass flow inlet and Pressure outlet boundary types were adopted on the inlet and outlet respectively. All the solid surfaces of the air region adopted a non-velocity slip boundary condition. The shell wall temperature obeyed the adiabatic boundary condition,  $\frac{\partial T}{\partial n}|w = 0$ , while the coupled wall temperature was set as constant,  $T_{wall} = const$ .

#### (2) In the steam part

The Mass flow inlet and Pressure outlet boundary types were adopted on the steam inlet and condensate water outlet respectively, assuming that the inlet flow distributed evenly. All the solid surfaces of the steam region adopted a non-velocity slip boundary condition. The present paper mainly aimed at the steam side and only considered the heat transfer perpendicular to the wall between the high temperature steam and air, ignoring the heat transfer in the other direction. For simplification, the steam side heat transfer process could be divided into steam cooling and water vapor condensation sections. Two segments of wall temperature were different but were set as constant [19–22]. The corresponding parameters of working fluids and materials are presented in Table 2.

#### 2.5. Numerical method and governing equations

To simplify the numerical simulation related to the phase change, the channels for air and steam were simulated respectively. The calculation model was simplified to only two-phase flow for the steam side and only single-phase flow for the air side.



1—air inlet 2—steam inlet 3—condensate water outlet 4—air outlet 5—wall 6 —heat transfer surface

Fig. 3. Sectional physical model of the novel plate air heater.



Fig. 4. Physical model of the novel plate air heater.



Fig. 5. Temperature comparisons of different grid cases.

To simply this 3-D numerical modeling, the following assumptions were made [23,24]:

- There was a constant volume flow rate in both flow sides
- The air was incompressible Newtonian fluid
- The gravity and buoyancy lift effect for air side was negligible while the gravity effect for steam side was considered
- Thermal radiation and viscous dissipation for air side was negligible while the viscous dissipation was considered
- The solid surface was adiabatic
- The air heater boundary condition and the external environment were negligible

#### 2.5.1. Governing equations for the air side

With consideration of the above assumptions, the governing equations of the mathematical model of air side are as follows.

Continuity equation:

$$\frac{\partial u_x}{\partial x} + \frac{\partial u_y}{\partial y} + \frac{\partial u_z}{\partial z} = 0 \tag{1}$$

Table 1Mesh dependency.

No. of cells	Skewness < 0.7	h	$\Delta P$	Time per iter. (s)
8,329,473	0.9745	71.33	85.74	4-7
11,162,569	0.9815	69.19	83.22	5-12
13,327,549	0.9823	69.17	83.00	8–17
15,521,835	0.9847	69.05	81.96	15-22
16,932,194	0.99	68.97	82.43	20-35

Momentum equation:

$$\rho \frac{du_i}{dt} = -\frac{\partial p}{\partial x_i} + di v (\mu \cdot gradu_i)$$
<sup>(2)</sup>

Energy equation:

$$\rho \frac{dE}{dt} = -p \cdot di v \, \vec{u} + di v (k \cdot gradT) \tag{3}$$

The standard k- $\varepsilon$  model is the simplest and the most widely validated turbulence model for which only boundary conditions need to be supplied. It has achieved notable success in calculating a wide variety of thin shear layer and recirculating flows without the need for case-by-case adjustment of the model constants. And at high Reynolds number, the standard k- $\varepsilon$  model avoids the need to integrate the model equations right through to the wall by making use of the universal behavior of near-wall flows. So the standard k- $\varepsilon$ model was adopted to simulate the turbulent flow for the air side in this study, which is presented as follows [25].

$$\frac{\partial(\rho k)}{\partial t} + \frac{\partial(\rho k u_i)}{\partial x_i} = \frac{\partial}{\partial x_j} \left[ \left( \mu + \frac{\mu_t}{\sigma_k} \right) \frac{\partial k}{\partial x_j} \right] + G_k - \rho \varepsilon$$
(4)

$$\frac{\partial(\rho\varepsilon)}{\partial t} + \frac{\partial(\rho k u_i)}{\partial x_i} = \frac{\partial}{\partial x_j} \left[ \left( \mu + \frac{\mu_t}{\sigma_\varepsilon} \right) \frac{\partial\varepsilon}{\partial x_j} \right] + \frac{C_{1\varepsilon}\varepsilon}{k} G_k - C_{2\varepsilon} \rho \frac{\varepsilon^2}{k}$$
(5)

#### 2.5.2. Governing equations for the steam side

It should be noted that the simulation of the phase change process for the steam side was quite different from the air side. The correctness of the steam side depended on the UDF, the addition of mass and an energy source item as well as the set of the model and the boundary conditions.

The governing equations for the mathematical model of steam side are as follows:

Continuity equation:

$$\frac{\partial \rho}{\partial t} + \rho \left( \frac{\partial u_x}{\partial x} + \frac{\partial u_y}{\partial y} + \frac{\partial u_z}{\partial z} \right) = S_m \tag{6}$$

Momentum equation:

$$\rho \frac{du_i}{dt} = -\frac{\partial p}{\partial x_i} + di v (\mu \cdot gradu_i) \tag{7}$$

**Energy equation:** 

$$\rho \frac{dE}{dt} = -p \cdot di \nu \, \vec{u} + di \nu (k \cdot gradT) + \Phi + S_E \tag{8}$$

The standard  $k-\omega$  model initially attracted attention because integration to the wall does not require wall-damping functions in low Reynolds number applications. The k- $\varepsilon$  model assumption

# Table 2 Physical parameters of materials and working fluid.

	$ ho~({ m kg}\cdot{ m m}^{-3})$	$\lambda (W(m \cdot K)^{-1})$	$C_p (J(kg \cdot K)^{-1})$
Steel	7840	49.8	465
Air	1.205	0.0259	1005
Steam	998	0.0256	2119
Water	998	0.599	4183

is that the flow is fully turbulent and the effects of viscosity are negligible, so it is not suitable to adopt k- $\varepsilon$  model for low Reynolds number flow. The standard k- $\omega$  model was adopted to simulate the turbulent flow for the steam side, because of its precision of near wall domain compared with the standard k- $\varepsilon$  model [26].

$$\frac{\partial(\rho k)}{\partial t} + \frac{\partial(\rho k u_i)}{\partial x_i} = \frac{\partial}{\partial x_j} \left[ \Gamma_k \frac{\partial k}{\partial x_j} \right] + G_k - Y_k + S_k \tag{9}$$

$$\frac{\partial(\rho\omega)}{\partial t} + \frac{\partial(\rho\omega u_i)}{\partial x_i} = \frac{\partial}{\partial x_j} \left[ \Gamma_\omega \frac{\partial\omega}{\partial x_j} \right] + \frac{C_{1\omega}\omega}{k} G_k - Y_\omega + S_\omega$$
(10)

The governing equations for both sides were discretized by the finite volume method (FVM). A coupled pressure-based solver was adopted and the coupling of velocity and pressure were solved with the semi implicit method for pressure linked equation (SIMPLE). The convection terms were discretized by using the second-order upwind scheme and the mixture model was adopted for the steam side. C programming languages were used to write UDF, in which steam was set as the main phase while condensate water was set as the secondary phase.

#### 3. Simulation results and discussions

The numerical results are presented in terms of the heat transfer coefficient and pressure drop, by applying the control variable methods when the steam and air inlet temperature were  $120 \,^{\circ}$ C and  $20 \,^{\circ}$ C respectively.

#### 3.1. Influence of the steam volume flow rate

The overall heat transfer coefficient and the pressure drop values with a variable steam volume flow rate are presented in Fig. 6. The air volume flow rate is  $480 \text{ m}^3 \cdot \text{h}^{-1}$ .

The results show that the overall heat transfer coefficient increased with the increasing steam volume flow rate. The cause of this phenomenon is that as the increased steam volume flow rate flow past the fluid surfaces, the effect of viscous forces is more apparent. The viscous forces would pull fluid surfaces making them thinner and enlarge the heat transfer coefficient, furthermore, viscous forces would strengthen the disturbance of fluid surfaces and reduce the thermal resistances. However, the increased condensate water which would lead to an increased film thickness, reducing the volume flow rate influence on the heat transfer coefficient. With the continuous condensation of steam, the steam partial pressure near the fluid surfaces constantly decreases while the partial pressure of non-condensable gas increased. Only through the non-condensable gas layer, the steam could condense on the liquid membrane surfaces. So the non-condensable gas layer is equal to the additional thermal resistance, leading to the poor heat conduction performance.

The steam side pressure drop trend is in accordance with the trend of the heat transfer coefficient. This is caused by the intensified disturbance of the larger steam volume flow rate. When the steam velocity increases, the interaction and the perturbation between condensate water and steam become stronger, which increases the pressure drop. Thus, when there is an unlimited



Fig. 6. Heat transfer coefficient and steam-side pressure variation as the steam volume flow rate is changed.

increase in the inlet velocity, disturbance become more intense. So controlling the flow rate is required, reducing the loss caused by excessive pressure drop.

Increasing the steam volume flow rate could enhance the heat transfer effect, however, in the meantime the pressure drop would increase. So, there exists an optimum steam and air flow rate value, under which the best heat transfer performance of novel plate air heater could be achieved.

#### 3.2. Influence of air volume flow rate

The influences of the air volume flow rate on the heat transfer coefficient and pressure drop values are revealed in Fig. 7.

In contrast with the results of the steam volume flow rate, the pressure drop and heat transfer coefficient values display different trends with that of the increased air volume flow rate. The heat transfer coefficient increases when the air volume flow rate increases. This follows the same principle as the steam side, in that the disturbance intensity increases. That is to say, when a plate on which condensation occurred is sufficiently large or there is a sufficient amount of condensate flow, turbulence may appear in the condensate film. This turbulence results in higher heat-transfer rates. In the condensation process the surface is blanketed by the film, which grows in thickness as it moves down. A temperature gradient exists in the film and the film represents a thermal resistance to heat transfer, which makes the increase of heat transfer coefficient slow down. However, the pressure drop for steamside decreases with the increasing air volume flow rate. The reason is that the increasing air volume flow rate enhances the heat transfer and increases the condensate water volume fraction, which leads to the condensate water outlet velocity increasing and the static pressure decreasing.



**Fig. 7.** Heat transfer coefficient and steam-side pressure variation as the air volume flow rate is changed.

#### 3.3. Optimization criterion

Two well-known dimensionless parameters, Fanning f-friction factor and Colburn j-factor were adopted to describe the heat transfer performance and pressure drop characteristics [27–29].

$$j = \frac{Nu}{RePr^{1/3}} \tag{11}$$

$$f = \frac{2\Delta P D_h}{\rho L u^2} \tag{12}$$

where Reynolds number, Nusselt number and Prandtl number are expressed as follows:

$$Re = \frac{\rho u l}{\mu} \tag{13}$$

$$Nu = \frac{hl}{\lambda} \tag{14}$$

$$Pr = \frac{\mu C_p}{\lambda} \tag{15}$$

The enhancement in heat transfer is accompanied by increases in the pressure drop for the steam side. The performance is presented in terms of the heat transfer coefficient and pressure drop. And to quantify the comprehensive performance, the *JF* factor [30], as the optimization criterion, is defined as follows:

$$JF = \frac{j/j_{ref}}{(f/f_{ref})^{1/3}}$$
(16)

where  $j_{ref}$  and  $f_{ref}$  stands for the reference circular tube heat exchanger with the same inlet cross-sectional area.

The optimization criterion *JF* factor value variations in different air and steam volume flow rates are shown in Fig. 8 and Fig. 9.

As shown, all *JF* factor values range from 1.86 to 2.80 for steam side and from 1.11 to 1.86 for air side, illustrating that the comprehensive performances for the plate air heater with variable channels are better than that for the reference ones. And the optimal points are the volume flow rate 480 m<sup>3</sup> · h<sup>-1</sup> for the air-side and 12 m<sup>3</sup> · h<sup>-1</sup> for the steam-side.

#### 4. Experimental implementation

#### 4.1. Experimental setup

A schematic diagram of the experimental system design of the novel plate air heater is shown in Fig. 10(a). The main sections of experimental apparatus were (1) the test object, (2) the water-loop, (3) the air system, and (4) the measurement equipment.



Fig. 8. Optimization criterion variation as the steam volume flow rate is changed.



Fig. 9. Optimization criterion variation as the air volume flow rate is changed.

The test object consisted of an air heater equipped with 5 plates in a cross-flow arrangement [31,32].

To validate the CFD simulations with accurate and reliable results, precise control systems and sensitive measurement equipment are required. The high sensitive vortex street flowmeter was used to measure the volume flow rate passing through the test object from the air and steam sides. The pressure drop was measured when the local pressure values of the test object inlet and outlet were subtracted. To obtain the precise results, four sensitive pressure sensors with accuracy of  $\pm 0.5$  Pa were used for the steam side and air side. The inlet and outlet temperatures were measured by two pt-100 thermocouples inserted into the flow line. The wall temperature distribution was measured by 12 K-type thermocouples welded onto different positions of the test object's external surfaces [33–36].

The uncertainties of the experimental data [37–39] are based on the method proposed by Wheeler AJ [40]. After calculation, the uncertainty values of the total heat transfer and the overall heat transfer coefficient, the pressure drop were about 2.21%, 1.78% and 5.46% respectively.

#### 4.2. Model validation

To validate the CFD results, the experimental approach was carried out using the same geometry as presented in the numerical simulations and were operated at the same air inlet flow rate while the steam flow was  $12 \text{ m}^3 \cdot \text{h}^{-1}$ , due to the difficulties in controlling the steam flow rate in the experimental operation. The numerical results and the experimental results for the novel plate air heater are shown in Fig. 11 and Fig. 12.

As depicted in Fig. 11 and Fig. 12, the obtained numerical results are compatible with the experimental results. As illustrated, the deviation between the simulation and experiment is about 6.21% and 5.44% for pressure drop and heat transfer coefficient respectively with the range of air flow rate from  $360 \text{ m}^3 \cdot \text{h}^{-1}$  to  $480 \text{ m}^3 \cdot \text{h}^{-1}$ . The pressure drop of the simulation is about 6.21% lower than that of the experimental data. Probably, the thickness of channel and cover plates in the real test section make the pressure drop of the experimental study higher than the obtained numerical one. Therefore, we can conclude that the present numerical approach was reliable in predicting the heat transfer and hydraulic performance for the novel plate air heater.

#### 4.3. Example of applied study

The most commonly-used air heater type in practical application is the finned tube, but because of the disadvantages associated with the finned tube air heaters in two feed mills of Shandong province, substitution of the original one with the presented novel





(b)

Fig. 10. Experimental set-up of air-steam heat exchanger system of new plate air heater.



Fig. 11. Comparison between the simulated and experimental values of pressure variation.

plate air heater would be advantageous. Fig. 13 displays the physical model of the finned-tube air heater and its geometric parameters. Within a number of engineering practices, the novel plate air



Fig. 12. Comparison between the simulated and experimental values of heat transfer coefficient.

heaters show advantages in enhancing heat transfer efficiency, reducing production costs and steam consumption. The comparison results between the two air heaters under the same conditions



Fig. 13. Physical model of the finned-tube air heater and its geometric parameters.

#### Table 3

Comparison between the experimental values of air heater.

Parameters	Novel plate air heater	Finned tube air heater
Volume flow rate $(m^3 \cdot h^{-1})$	19,000	19,000
Velocity $(m \cdot s^{-1})$	5	3
Pressure drop for air side (Pa)	440	728
Air outlet temperature (°C)	80	80
Overall heat transfer coefficient $(W(m^2 \cdot K)^{-1})$	70-90	25-40
Heat transfer area (m <sup>3</sup> )	20.56	60
Steam inlet temperature (°C)	200	200
Steam consumption (t)	6–7	8-9
Steam outlet temperature and status	50–60 °C	>95 °C
	Condensate water	Condensate water and steam

of 5.5 kW fan and 19,000  $m^3 \cdot h^{-1}$  air volume flow rate are presented in Table 3.

The results indicate that the novel plate air heater could save 20-35% of steam compared to the finned-tube heat exchanger. Additionally, the heat transfer area is 1/3 of that of the finned-tube air heater while the heat transfer coefficient is approximately 2-3 times that of the finned-tube type.

#### 5. Conclusions

In this paper, a novel plate air heater was proposed for consideration by academic researchers and engineering designers. A CFD study was used to analyze the steam and air volume flow rate affecting the performance of the novel plate air heater. An experimental system was built to verify the accuracy of the modeling approach and simulation results. The results were compared based on the heat transfer coefficient and the drop in pressure. The following conclusions were obtained according to the results:

- (1) The experimental results showed that the main effect on the overall heat transfer coefficient was the air-side thermal resistance. The steam volume flow rate was not closely related to the overall heat transfer coefficient, because the steam-side heat transfer coefficient was larger than the air-side coefficient.
- (2) The modeling of the air heater with the air-side and steamside channels separated was tried and tested in the CFD simulation and the results proved very satisfactory.

- (3) Consistency was found between the results of the numerical simulation and experimental testing for a wide range of flow conditions-6.21% difference for the steam-side pressure drop and 5.44% for the heat transfer coefficient.
- (4) The numerical simulation showed that the increased steam volume flow rate enhanced the heat transfer effect, but at the same time increased the drop in pressure. Further, an increase in the air volume flow rate increased the heat transfer coefficient but decreased the pressure drop. An optimal index was adopted to decide the best volume flow rate 480 m<sup>3</sup> h<sup>-1</sup> for the air-side and 12 m<sup>3</sup> h<sup>-1</sup> for the steamside in which conditions of the heat transfer coefficient were approximately 70 W (m<sup>2</sup> K)<sup>-1</sup>.
- (5) The novel plate air heater had advantages compared to the finned-tube air heater, such as the enhanced heat transfer efficiency, lower production cost and steam consumption in an engineering application.

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