

碳管基元序构高性能滤波超级电容器电极研究进展

李 佩^{1,2}, 周子燕^{1,2}, 蒋素侠^{1,2}, 徐潇建^{1,2}, 韩方明^{1,2}, 孟国文^{1,2}

(1. 中国科学院合肥物质科学研究院固体物理研究所, 合肥 230031;

2. 中国科学技术大学研究生院科学岛分院, 合肥 230026)

摘 要: 随着电子产品向轻量化与小型化方向快速发展, 传统铝电解电容器受限于面积/体积比电容低、结构刚性强等固有缺陷, 已难以满足现代电子系统对高性能滤波元件的需求。凭借高比电容、快速充放电特性及优异的循环稳定性, 超级电容器被认为是替代铝电解电容器的潜在方案。然而, 传统电极内部无序多孔结构引发了严重的传质瓶颈。曲折的离子迁移路径极大地限制传质动力学, 导致器件在 120 Hz 频率下陷入“鱼与熊掌不可兼得”的困境——快速响应与高能量密度无法共存, 严重阻碍了其走向实际应用的步伐。近年来, 通过对电极微观结构进行理性设计, 尤其是对组成电极的纳米结构功能基元的空间取向与连接方式实施精确序构调控, 逐渐成为突破上述矛盾的重要策略。基于结构维度与空间组织方式, 本文对碳基滤波电极进行系统梳理, 包括以高堆积密度为特征的二维平面结构、以随机扩散为主导的三维大孔结构, 以及以定向传输为特征的高度垂直取向结构等, 揭示结构有序性在调控电荷输运动力学中的关键作用。在此基础上, 重点综述以碳管为功能基元的三维序构电极体系——三维互连碳管网络, 深入分析其在构建低电阻连续电子传导网络与低曲折度定向离子通道方面的结构优势。围绕“结构序构—离子输运—高频滤波性能”这一内在逻辑, 系统讨论自支撑骨架构筑、基元层级化调控、表界面工程、同轴套构、横向致密化及 Y-形分支设计等结构演化策略, 对离子输运行为及 120 Hz 滤波性能的影响规律。最后, 总结当前研究进展与未来发展趋势, 以期为高性能、小型化电化学滤波器的结构设计 with 理论研究提供参考。

关键词: 功能基元; 碳管; 序构; 滤波超级电容器; 电极结构设计; 电子-离子协同输运

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将交流电(如市电 50/60 Hz)高效转换为稳定的直流电, 是现代电子系统能量管理的核心环节^[1-3]。在此过程中, 滤波电容器作为整流电路的关键元件, 承担着平滑电压脉动、抑制纹波噪声以保护敏感负载的重任, 其性能直接决定了电路的能量转换效率、信号质量与系统稳定性^[4-7]。铝电解电容器凭借成熟的制造工艺、低廉的成本、宽泛的工作电压范围以及快速的频率响应特性, 在滤波应用中长期占据主流地位^[8-10]。然而, 铝电解电容器能量密度较低, 为满足实际滤波需求往往需要较大的器件体积^[11-15]。随着电子器件持续向轻量化、小型化方向发展, 铝电解电容器的这一固有局限日益突出, 已成为制约高端电子系统进一步发展的技术瓶颈^[16-19]。因此, 开发兼具高能量密度与快速频率响应的新型滤波电

容器, 成为当前电力电子与储能领域亟须攻克的重要课题^[20]。

超级电容器是一类介于传统电介质电容器与电池之间的电化学储能器件, 根据储能机制可分为双电层电容器和赝电容器^[21-24]。其中碳基双电层电容器凭借优异的倍率性能、长循环寿命及显著高于铝电解电容器的比电容, 被广泛视为下一代交流滤波电容器的潜在候选者^[25]。理论上, 超级电容器可在同等电容下显著缩小器件体积, 推动电子设备微型化。然而, 要满足市电交流线路滤波的应用需求, 器件须在 120 Hz 下具备极快的频率响应, 即相位角(φ)接近 -90° , 电阻-电容时间常数(τ_{RC})不超过 8.3 ms^[26-28]。遗憾的是, 传统电极(如活性炭、炭黑)普遍存在随机分布且高度曲折的微纳孔隙结

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第一作者: 李 佩(1998—), 女, 博士研究生。

通信作者: 韩方明(1975—), 男, 博士, 研究员;

孟国文(1960—), 男, 博士, 研究员。

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First author: LI Pei (1998—), female, Doctoral candidate.

E-mail: lpei@issp.ac.cn

Correspondent author: HAN Fangming (1975—), male, Ph.D., Professor.

MENG Guowen (1960—), male, Ph.D., Professor.

E-mail: fmhan@issp.ac.cn; gwmeng@issp.ac.cn

构^[1, 29], 这种结构虽有利于提供高比表面积以增大比电容, 却极大地限制了电解质离子的动力学传输, 导致高频下离子阻抗激增, 器件呈现电阻特性而非电容行为, 严重制约了其在滤波领域的应用^[28, 30-34]。

针对上述结构性矛盾, 摒弃传统“随机造孔”模式, 转向电极微观结构的理性设计, 尤其是构建具有高度定向传输通道的“功能基元序构”电极, 已成为突破滤波超级电容器性能瓶颈的关键^[35-37]。所谓“序构”, 是指在微/宏观尺度上对“功能基元”的空间取向及连接方式进行精密调控, 构建连续电子传导网络与低曲折度离子迁移通道, 实现电极内部的电子-离子协同传输, 从而优化传输动力学、并突破容量与频率响应之间的结构性约束^[38-44]。基于此, 本文从结构有序化与传输连续性的视角出发, 系统梳理了碳基滤波电极在结构设计与可控制备方面的研究进展。首先回顾平面二维碳结构、随机三维碳结构以及垂直取向碳结构等典型体系的构型特征与性能表现, 剖析其因本征结构局限(如二维片层的致密堆叠、三维孔道的曲折无序、早期取向结构的连通

性不足)所引发的电子-离子协同传输受阻与频率响应衰减等问题。随后, 重点聚焦以“碳管功能基元+序构”的三维碳管网络电极, 通过梳理其体系化的结构演化路径, 从“结构-输运-性能”关联机制的角度, 解析其对宏观电化学行为的调控规律, 以期为高性能、小型化滤波器件电极的设计提供理论依据。

1 工作机制与性能评估

1.1 超级电容器的滤波机制

在实际电子系统中, 交流电向稳定直流电的转换通常依赖“整流-滤波”两级协同处理(图 1a)。首先, 双向交流电经全桥整流电路后转换为单向脉动直流电。其中, 针对 50/60 Hz 的市电, 全波整流后输出纹波频率翻倍至 100/120 Hz, 因此 120 Hz 下的频率响应行为被公认为评价交流滤波性能的核心指标。由于整流后的脉动直流信号含有显著的周期性纹波, 若直接为敏感电子器件供电, 极易引发信号失真或器件失效, 因此必须引入滤波电容器进行平滑处理, 以获得稳定的直流供电^[4]。

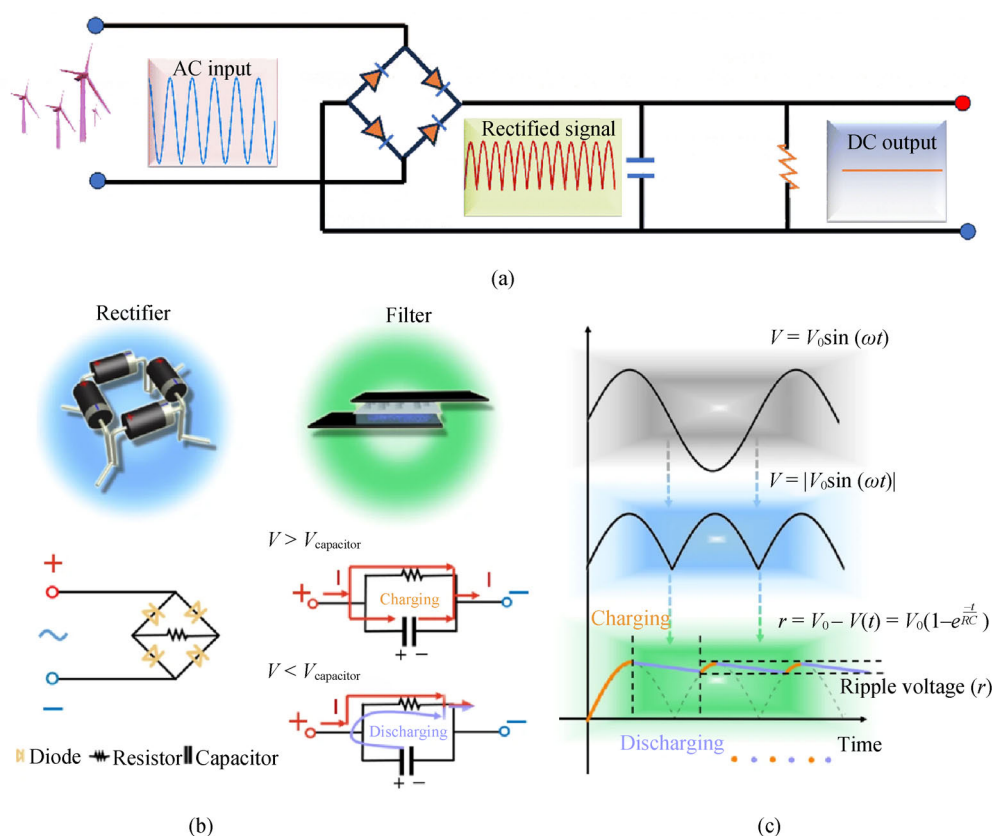


图 1 交流滤波示意图^[2, 24]

Fig. 1 Schematic illustration of the filtering procedure^[2, 24]

滤波电容器的稳压机制本质上源于其快速充放电特性, 即通过对电能快速暂存与释放来抑制电

压波动(图 1b)。当整流输出电压高于电容器两端电压时, 电容器进入充电状态吸收电荷以削弱电压峰

值;反之,当整流输出电压低于电容器两端电压时,电容器释放存储的电荷以补偿电压跌落。通过这种高频次的“充电—放电”周期交替,脉动直流电压的波动幅度被大幅抑制,输出电压趋于平稳(图 1c)。在此过程中,电容器充当“缓冲器”或“蓄水器”,通过“削峰填谷”维持系统电压的稳定^[24, 45]。

上述滤波过程对电容器的动态响应速率与有效储能容量提出了双重严苛要求^[46]。若电压波动频率超过电容器的可响应上限,电容器将难以及时完成电荷存储与释放,表现出电阻的行为,从而削弱甚至丧失滤波效果。此外,电容器的电荷存储量必须足以弥补电压波动产生的能量缺口。如式(1)所示,滤波后的直流输出纹波电压幅值与电容值成反比:

$$r = V_0 - V_t = V_0(1 - e^{-\frac{t}{RC}}) \quad (1)$$

其中: R 为放电回路的等效负载电阻; C 为滤波电容; V_0 为电压峰值; V_t 为瞬时电压。

由此可知,实现高效交流滤波需满足双重条件:一方面,为响应高频交流变化,电容器必须具备极快的时间常数(即低 R 与 C 的协同);另一方面,为抑制纹波幅值,又须具备足够大的有效电容。因此,理想的滤波电容需在快速响应与高精度纹波抑制之间取得平衡。

1.2 性能参数

电化学阻抗谱是表征滤波电容器频率响应特性的核心手段^[47]。该技术通过向电化学体系施加小振幅正弦电位(或电流)扰动,并采集相应的响应电流(或电位)信号,从而获得阻抗随频率变化的谱图信息。凭借其对不同弛豫过程的分辨能力,电化学阻抗谱能够有效区分受电子传导、离子扩散或电荷转移受限的动力学步骤,为电化学响应机制的解析提供定量依据^[24, 48]。

奈奎斯特(Nyquist)图和伯德(Bode)图是最常用的 2 种数据表示方式。其中, Nyquist 图以实部阻抗(Z')为横轴、虚部阻抗(Z'')为纵轴,主要用于解析体系的内部阻抗构成; Bode 图则以频率(f)的对数坐标为横轴、相位角(φ)为纵轴,直观反映体系 φ 随 f 的变化。结合 Nyquist 与 Bode 图的综合分析,可实现对超级电容器等效串联电阻、离子扩散动力学及频率响应极限的系统评估,为开发高性能交流滤波电极奠定表征基础。

Φ 为衡量滤波器性能最直观且最关键的参数,用于表征器件电化学行为相对于理想电容的偏离程度。理想电容器的阻抗仅包含纯电容,其相位角恒

定为 -90° 。而在实际器件中,受等效串联电阻(R_s)的影响, φ 的绝对值会向小于 90° 的方向偏移,其计算公式如式(2)所示^[49](实际多孔电极需按传输线模型处理,更为复杂):

$$\Phi = -\arctan\left(\frac{1}{2\pi f C R_s}\right) \quad (2)$$

其中: f 为工作频率(通常取 120 Hz); C 为滤波电容器的电容值; R_s 为等效串联电阻。 φ 越接近 -90° , 表明器件的电容性行为越显著,对 120 Hz 交流信号的动态响应能力越强。在该状态下,器件能够高效抑制交流纹波,而不会因过大的阻性成分引发显著能量损耗与热效应。

面积比电容(C_A)是评价滤波性能的另一核心参数,表征单位面积电极在目标频率下的电荷存储能力,计算公式如式(3)所示:

$$C_A = -\frac{1}{2\pi f Z'' s} \quad (3)$$

其中: Z'' 为虚部阻抗; s 为电极面积。较高的 C_A 有助于在保证高频响应的同时维持足够的有效电容,从而兼顾滤波效率与器件小型化需求。

实部比电容(C')与虚部比电容(C'')分别反映器件的有效电荷存储能力与能量耗散行为,其计算公式为:

$$C' = -\frac{Z''}{2\pi f |Z|^2 s} \quad (4)$$

$$C'' = \frac{Z'}{2\pi f |Z|^2 s} \quad (5)$$

其中: Z' 表示实部阻抗; $|Z|$ 表示阻抗模值。

电阻-电容时间常数(τ_{RC})的表达式为:

$$\tau_{RC} = -\frac{Z'}{2\pi f Z''} \quad (6)$$

该参数反映了电荷(电子与离子)迁移过程所受到的动力学限制, τ_{RC} 越小,表明器件的电荷输运动力学越快,倍率性能越优。

弛豫时间常数(τ_0)被定义为电容器释放超过 50% 能量所需的最短时间,其表达式如式(7)所示:

$$\tau_0 = \frac{1}{f_0} \quad (7)$$

其中: f_0 为虚部电容 C'' 达到最大值时对应的频率。 τ_0 越小,表明器件的能量释放速率越快,频率响应性能越优。

此外,耗散因子(DF)用于表征电容器在高频条件下的能量损耗特性,其定义如式(8)所示:

$$DF = \frac{|P|}{|Q|} = \frac{C''}{C'} \quad (8)$$

其中: $|P|$ 以及 $|Q|$ 分别代表无功功率和有功功率。较低的 DF 值意味着更低的能量损耗。

体积能量密度(E)和体积功率密度(P), 分别用于评价滤波电容器的能量存储能力与功率输出特性, 其计算公式如式(9)和式(10)所示:

$$E = \frac{CV^2}{2} \quad (9)$$

$$P = \frac{E}{\Delta t} \quad (10)$$

其中: C 为体积比电容; V 为电势窗口; Δt 为放电时间。

综上所述, 相位角、时间常数及面积比电容等关键参数的本质, 均指向电极体系中电子传输阻抗与离子扩散动力学的协同匹配问题。如何在保证高电荷存储能力的同时, 构建低曲折度、短离子扩散距离且高度连续导电的双通道运输结构, 已成为滤波超级电容器电极设计的核心科学问题。

2 碳基滤波超级电容器电极的结构演化与动力学瓶颈

在超级电容器研究体系中, 碳基材料凭借优异的电子导电性、高度可调的孔隙结构以及良好的电化学稳定性受到广泛关注^[50-51]。随着应用场景由传统的“高功率储能”向“交流滤波”领域拓展, 电极的设计逻辑亦由“容量驱动”转向“动力学驱动”^[52]。交

流滤波过程要求电极在 120 Hz 甚至更高频率下实现近乎理想的电容行为, 这对离子的迁移效率提出了严苛要求。研究者试图通过电极维度的精密调控与空间结构的优化, 打破传统双电层电荷存储在时间尺度上的限制。

本部分将系统梳理碳基滤波超级电容器电极从“平面二维限制”到“随机三维贯通”, 再到“垂直取向引导”的结构演化历程, 深入剖析各阶段在降低离子传输路径曲折度方面的突破及其残存的动力学瓶颈, 进而揭示从“几何取向”走向“基元序构”的必然性。

2.1 平面二维结构

平面二维结构主要指由炭黑、碳纳米管、石墨烯及其复合组分通过高密度堆叠形成的薄膜体系^[31, 53-57]。此类设计旨在利用碳材料本征的高电子迁移率构建高效电子传导网络, 并依托高堆积密度在有限体积内实现高电荷存储。然而, 该类结构表现出极强的传输各向异性, 虽然面内导电性能优异, 但面外方向由于缺乏连续的离子传输通道, 导致离子迁移阻力随电极厚度增加而迅速上升。

为破解上述瓶颈, 研究者主要采取“减薄物理厚度”与“构筑跨层通道”两条技术路径, 旨在通过极短的扩散路径补偿面外输运的迟滞性, 或利用面内穿孔打破层间阻塞。例如, Zhang 等^[53]采用原位碳热还原策略制备了具有均匀且密集纳米孔的超薄自支撑石墨烯纳米网薄膜(如图 2a 所示)。通过在层间构筑离子迁移“捷径”, 显著缩短了纵向扩散路径, 使其

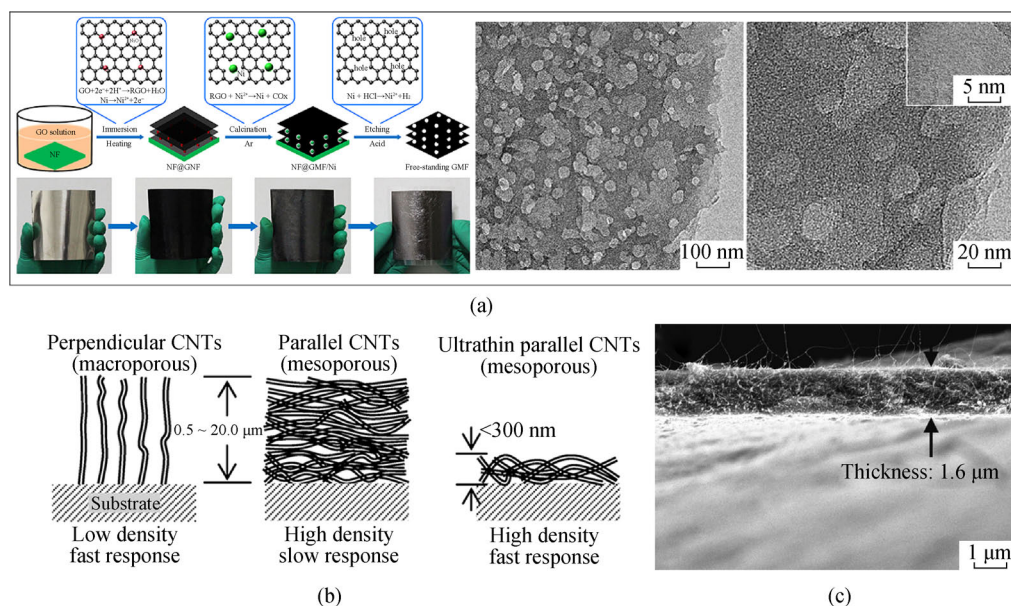


图 2 (a)自支撑石墨烯纳米网膜^[53]; (b)具有不同取向和厚度的碳纳米管膜^[56]; (c)单壁-多壁碳纳米管膜^[57]

Fig. 2 (a) Free-standing graphene nanomesh films^[53]; (a) Carbon nanotube films with different orientations and thicknesses^[56]; (b) Single-walled carbon nanotubes/multi-walled carbon nanotubes film^[57]

在 120 Hz 下实现了 $7.6 \text{ F}\cdot\text{cm}^{-3}$ 的高体积比电容和 -82.3° 的相位角, 性能显著优于传统无孔石墨烯薄膜电极。类似地, Yoo 等^[56]通过真空抽滤制备了水平取向的超薄碳纳米管膜(如图 2b 所示), 利用其介孔网络与极短的纵向孔隙长度, 在 120 Hz 下实现了

-82.2° 的相位角和 $282 \text{ }\mu\text{F}\cdot\text{cm}^{-2}$ 的面积比电容。进一步地, Rangom 等^[57]通过强化超薄单壁碳纳米管薄膜与集流体的界面耦合(如图 2c 所示), 将面积比电容提升至 $601 \text{ }\mu\text{F}\cdot\text{cm}^{-2}$ 。表 1 汇总了典型平面二维结构电极的关键性能参数。

表 1 典型平面二维结构电极的关键性能参数对比

Table 1 Comparison of key performance parameters for typical two-dimensional planar electrodes

Electrode materials	Phase angle at 120 Hz/(°)	f_{-45}/Hz	C_A at 120 Hz/($\text{mF}\cdot\text{cm}^{-2}$)	$E_A@120 \text{ Hz}/(\text{mF}\cdot\text{V}^2\cdot\text{cm}^{-2})$	Reference
Free-standing graphene nanomesh films	-82.3	6211	0.31	0.10	[53]
Ultrathin carbon nanotube films	-82.2		0.28	0.88	[56]
Single-walled carbon nanotubes/multi-walled carbon nanotubes film	-81.0	1425	0.60	0.25	[57]

尽管通过减薄或穿孔在一定程度上改善了离子可及性, 但二维平面构型的本征结构局限仍未根除。受制于有限的离子渗透深度, 电极通常需维持在纳米至亚微米级的超薄状态以确保高频响应。一旦通过增加负载来提升面积比电容, 水平堆叠引发的层间阻塞将产生大量的动力学“死角”, 导致深层活性位点在高频下难以被有效利用。这种“容量-速率”的矛盾, 制约了其在高性能、小型化滤波器件中的应用。

2.2 随机三维大孔结构

为克服平面二维电极面积比电容有限、厚度难以拓展的瓶颈, 研究者在“面内致密堆积”策略之外, 提出“体相空间构筑”的设计思路, 通过构建贯通的三维互穿网络, 发展出空间连续的电极结构^[2, 58-66]。该策略利用三维空间的延展性, 旨在削弱致密薄膜对离子传输的几何约束, 为电解质提供多维度扩散通道, 从而在“高载量负载”与“快速频率响应”之间探索新的性能平衡。

利用自组装技术构建连续的电子/离子双通路, 被认为是实现高性能滤波电极的重要途径。例如, Sheng 等^[59]利用电化学还原氧化石墨烯制备的三维互穿结构电极(如图 3a 所示), 通过骨架间的开放孔隙, 在显著增加电解质可及比表面积的同时, 形成了连续的电子传导网络和低曲折度的离子扩散通道, 使器件在 120 Hz 下实现了 -84° 的相位角和 $283 \text{ }\mu\text{F}\cdot\text{cm}^{-2}$ 的面积比电容。为进一步缓解高负载下的频率响应衰减问题, 研究者将活性材料集成于三维导电骨架中, 发展出兼顾负载量与动力学的复合结构。Islam 等^[63]采用等离子体增强化学气相沉积技术, 在碳纤维微纤维片基底上垂直生长边缘取向多层石墨烯(如图 3b 所示), 构建出具有定向传输通道的三维网络。该电极在水系电解液中表现出卓越的动力学特性, 其 -45° 相位角对应的特征频率高达

13.8 kHz, 且 120 Hz 下面积比电容达到 $1.07 \text{ mF}\cdot\text{cm}^{-2}$ 。此外, Li 等^[65]通过 CH_4/H_2 等离子体处理 ZIF-67 金属有机框架, 在碳化过程中原位生长边缘取向石墨烯与碳纳米管, 形成以碳化颗粒为骨架, 石墨烯片层包覆、碳纳米管穿插其中的三维分级互连结构(如图 3c 和图 3d 所示)。该体系在 6 mol/L KOH 电解液中实现了 120 Hz 下约 $1.02 \text{ mF}\cdot\text{cm}^{-2}$ 的面积比电容和 -80.0° 的相位角, 进一步验证了分级连通三维结构在高频滤波电极设计中的有效性。表 2 汇总了典型随机三维大孔结构电极的关键性能参数。

尽管随机三维大孔电极在空间利用率与界面动力学方面具有显著优势, 但其结构随机性引发的内在矛盾依然突出。随机体系中孔道的高曲折度和微观无序性, 使离子难以在毫秒时间尺度内完成对深层孔隙的均一化响应。这种动力学不一致性意味着仅靠维度扩展仍难以触及滤波超级电容器的性能极限。因此, 构建取向一致、孔道垂直有序的特征结构, 成为推动滤波电极性能提升的必然选择。

2.3 垂直取向结构

针对平面结构各向异性传输受限以及随机大孔结构高曲折度引发的动力学瓶颈, 构建垂直于集流体方向的取向阵列结构(如垂直石墨烯、垂直碳纳米管)逐渐成为实现高效定向运输的突破路径^[5, 14, 67-82]。该设计的本质在于在电极厚度方向构筑开放且低曲折度的连续传输通道, 将离子的“随机扩散”升级为“定向迁移”, 从而在物理尺度上显著缩短离子响应时间、并降低离子扩散阻抗。

Miller 等^[67]率先利用射频等离子体增强化学气相沉积技术, 在镍基底上直接生长垂直取向石墨烯纳米片(如图 4a 所示), 通过优化离子传输路径显著降低了电阻电容时间常数, 使电容器在 120 Hz 下表现出接近理想的交流滤波特性(-82° 的相位角)。Xu

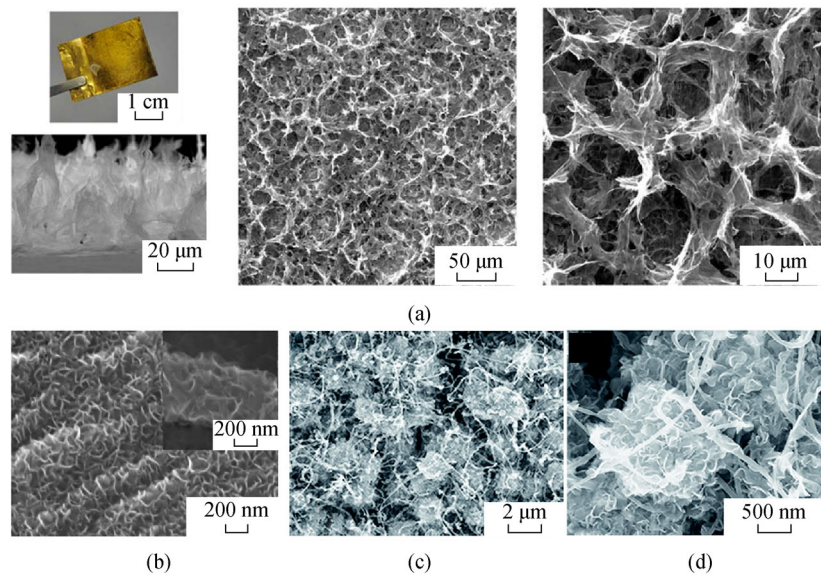


图 3 (a)3D 互穿结构的电化学还原氧化石墨烯^[59]；(b)垂直生长在纤维素微纤维片上的石墨烯纳米片^[63]；(c, d)ZIF-67 衍生的边缘取向石墨烯团簇与碳纳米管的复合材料^[65]

Fig. 3 (a) 3D interpenetrating electrochemical reduction of graphene oxide^[59]; (b) Edge-oriented graphene nanosheets perpendicularly grown around carbonized cellulose microfibril sheets^[63]; (c, d) ZIF-67-derived edge-oriented graphene clusters coupled with carbon nanotubes^[65]

表 2 典型随机三维大孔结构电极的关键性能参数对比

Table 2 Comparison of key performance parameters for typical random three-dimensional macroporous electrodes					
Electrode materials	Phase angle at 120 Hz/(°)	f_{-45}/Hz	C_A at 120 Hz/(mF·cm ⁻²)	$E_A@120\text{ Hz}/(\text{mF}\cdot\text{V}^2\cdot\text{cm}^{-2})$	Reference
3D interpenetrating electrochemical reduction of graphene oxide	-84.0	4200	0.40	0.13	[59]
Edge-oriented graphene nanosheets perpendicularly grown around carbonized cellulose microfibril sheets	-80.5	13 800	1.07	0.34	[63]
ZIF-67-derived edge-oriented graphene clusters coupled with carbon nanotubes	-80.0	1700	1.02	0.41	[65]

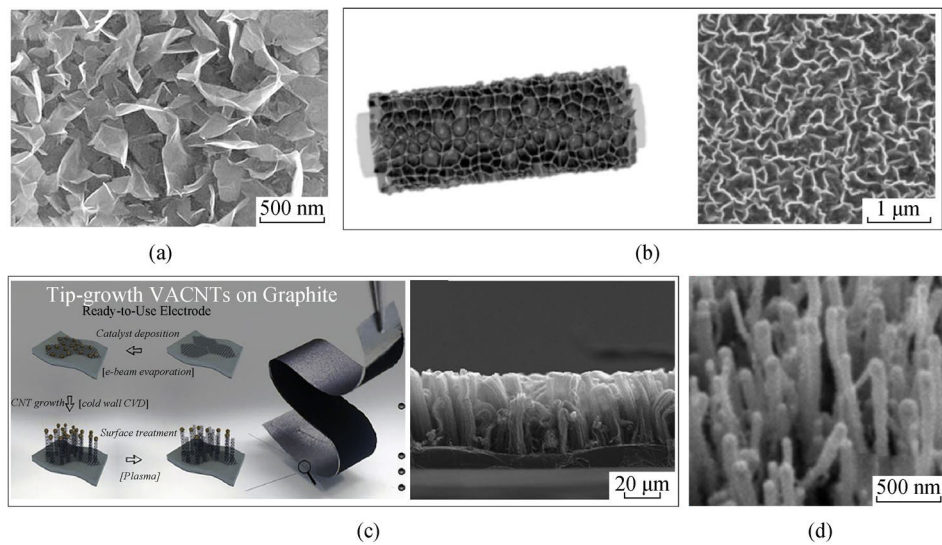


图 4 (a)垂直取向的石墨烯纳米片电极^[67]；(b)生长在纤维基底的垂直取向石墨烯^[76]；(c)石墨/垂直排列碳纳米管^[79]；(d)氮掺杂垂直排列碳纳米结构^[5]

Fig. 4 (a) Vertically oriented graphene nanosheet electrode^[67]; (b) Vertical graphene grown on the fiber substrate ^[76]; (c) Graphitic ordered mesoporous carbons and carbon nanotubes^[64]; (d) Nitrogen-doped vertically aligned carbon nanostructures^[5]

等^[76]在金属纤维电极上直接生长垂直取向石墨烯(如图 4b 所示), 得益于高度有序的垂直通道, 该纤维电极构筑的电容器展现出出色的滤波能力。类似地, Li 等^[79]在高度取向的石墨薄膜表面直接生长垂直排列碳纳米管(如图 4c 所示), 形成共价键合的石墨/碳纳米管结构。该设计在降低界面接触电阻的同时, 有效规避了金属的腐蚀, 在 6 mol/L KOH 电解液中实现了 120 Hz 下 -84.8° 的相位角和 $1.38 \text{ mF}\cdot\text{cm}^{-2}$ 的面积比电容。

此外, 为突破单一碳材料在电容密度方面的固有局限, 研究者在保持取向特征的基础上, 通

过异质原子掺杂或与其他碳材料复合策略引入额外储能位点。Santhosh 等^[5]对直接生长在镍箔集流体上的垂直排列碳纳米管原位氮掺杂(如图 4d 所示), 通过改善界面润湿性并引入赝电容贡献, 在 100 Hz 下获得约 $1145 \mu\text{F}$ 的电容和接近 -80° 的相位角。Premathilake 等^[81]则通过在垂直取向石墨烯纳米片电极表面及片层间隙中引入导电炭黑, 在 120 Hz 下实现了 $2.30 \text{ mF}\cdot\text{cm}^{-2}$ 的面积比电容, 且相位角仍保持在 -80° , 证明了协同优化策略的有效性。表 3 汇总了典型垂直取向电极的关键性能参数。

表 3 典型垂直取向电极的关键性能参数对比

Table 3 Comparison of key performance parameters for typical vertically oriented electrodes

Electrode materials	Phase angle at 120 Hz/ $^\circ$	f_{-45}/Hz	C_A at 120 Hz/ $(\text{mF}\cdot\text{cm}^{-2})$	E_A @120 Hz/ $(\text{mF}\cdot\text{V}^2\cdot\text{cm}^{-2})$	Reference
Vertically oriented graphene nanosheet electrode	-82.0	15 000	0.09	0.01	[67]
Graphite / carbon nanotube hybrid material	-84.8	1980	1.38	0.44	[79]
Vertically aligned carbon nanostructures (100 Hz)	-80.0	6000	1.15	0.57	[5]
Carbon-black-coated, vertically-oriented graphene	-80.0	1000	2.30		[81]

尽管垂直取向结构有效降低了纵向离子阻力, 实现了滤波性能的提升, 但其固有的“维度缺失”仍存在动力学与力学局限。单一纵向阵列在基元间缺乏充分的横向连通, 不仅制约了电子在平面方向的快速传导, 也削弱了电极整体的结构稳定性。随着电极厚度与负载增加, 孤立的纵向基元易发生局部聚集、塌陷或堆叠致密化, 导致有效活性面积下降, 并伴随离子传输阻力的显著增加, 动力学“死角”再次出现。这种现象表明, 仅依赖单向取向调控难以实现传输维度与结构稳定性的同步优化。

3 功能基元序构: 三维互连碳管网格电极的演进

针对前述平面二维电极的本征结构受限、随机三维大孔体系离子的传输曲折度高, 以及垂直阵列结构缺乏横向连通所导致的动力学瓶颈, 如何在结构层面实现“超快电荷响应”与“高面积比电容”之间的有效解耦, 已成为高性能滤波电极构建的关键。基于此, 电极的设计逻辑亟须由单纯的“几何空间堆叠”转向“功能基元序构”——即以碳基功能基元为基础, 通过对其空间取向与连接方式的精确调控, 构建一种既能继承垂直取向结构定向离子输运优势, 又兼具强横向电子耦合与多维结构稳定性的“序构化互连网络”。

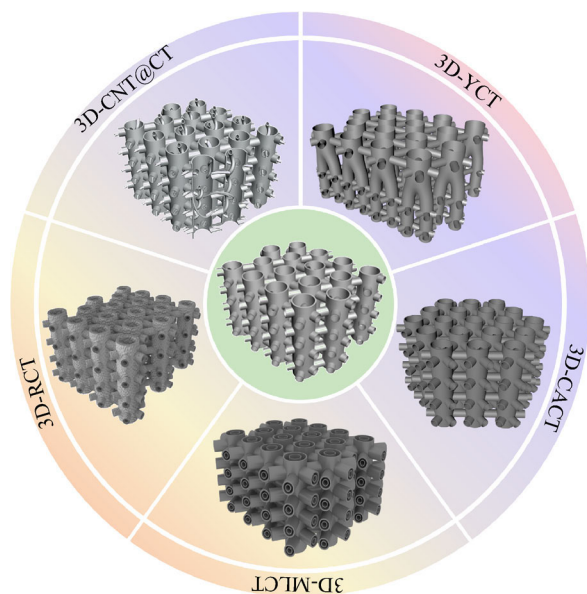
在上述设计理念驱动下, 基于“类脚手架”构型

的三维互连碳管网格膜电极应运而生。该体系突破了传统结构中基元孤立排列的局限, 通过在纵向阵列中引入横向互连, 实现了电子与离子在三维空间内的双连续传输, 完成了从“单向传输”向“多维协同动力学”的性能跃迁。这种互连架构在显著降低离子迁移路径曲折度的同时, 最大化了平面方向的电子导通效率, 且最大程度地保留了高负载量下的电化学活性界面, 为实现高频响应与高面电容的协同优化提供了结构支撑。

本部分将紧扣“基元序构”这一逻辑主线, 系统梳理三维互连碳管网格电极的结构演进逻辑: 从三维网格膜(3D-CT)的开创性构筑出发, 深入探讨基元内嵌碳纳米管(3D-CNT@CT)、基元表面粗糙化修饰(3D-RCT)、“套娃”式结构嵌套(3D-MLCT)、纵向阵列的横向致密化(3D-CACT)以及 Y-形分支序构(3D-YCT)等策略, 解析基元-空间构筑方式对动力学性能的协同调控机制(图 5)。此外, 表 4 汇总了 3D-CT 及其结构变体的关键电化学性能参数, 为定量评估“基元序构”对滤波电极传输性能的调控规律提供了关键数据支撑。

3.1 3D-CT 的开创性构筑

Han 等^[83]首次将“碳管”作为“基本功能单元”, 采用“自下而上”的序构策略, 将 3D-CT 引入滤波超级电容器领域。该研究以三维互连多孔阳极氧化铝为模板, 采用化学气相沉积法在模板孔道内限域沉

图 5 3D-CT^[83]及其结构演化^[83-86]Fig. 5 3D-CT^[83] and its structural variants^[83-86]表 4 3D-CT 及其结构变体的关键性能参数对比^[83-86]Table 4 A comparison of key performance parameters of 3D-CT and its structural variants^[83-86]

Electrode materials	Thickness/ μm	Phase angle at 120 Hz/($^{\circ}$)	f_{-45}/Hz	C_A at 120 Hz/($\text{mF}\cdot\text{cm}^{-2}$)	E_A @120 Hz/($\text{mF}\cdot\text{V}^2\cdot\text{cm}^{-2}$)	Reference
3D-CT	10	-81.5	2610	1.04	0.52	[83]
3D-CNT@CT	10	-81.0	2360	1.81	0.91	[83]
3D-RCT	12	-80.5	1120	2.81	1.41	[83]
3D-TLCT	12	-80.1	951	3.08	1.54	[84]
3D-CACT	12	-80.2	957	3.23	1.62	[85]
3D-YCT	15	-80.0		3.60	1.80	[86]

积碳层,随后去除氧化铝模板,获得类“脚手架”结构的 3D-CT(图 6a~图 6c)。该结构在传统垂直碳管阵列的基础上引入横向碳管,实现了纵向与横向互连的空间协同,构建出贯通三维空间的有序传输网络。高度取向的纵向孔道为电解质离子提供低曲折度、短扩散路径的迁移通道,确保离子在厚度方向上快速渗透;同时,横向碳管通过化学键连接垂直碳管,形成一体化的连续电子传导网络,使电子与离子传输路径在空间维度上实现协同贯通。此外,开放互连的三维自支撑骨架不仅赋予电极良好的结构稳定性,也保证了在较高负载条件下仍可维持内部孔道的连续开放性,避免因结构塌陷或局部堵塞导致频率响应的衰减。电化学测试结果进一步验证了该序构策略的有效性。如图 6d~图 6g 所示,该电极在 120 Hz 下实现了 -81.5° 的相位角和 $1.04 \text{ mF}\cdot\text{cm}^{-2}$ 的面积比电容, Nyquist 曲线呈现较小的等效串联电阻和几乎平行于虚轴的特征,表明其具备较理想的

电容行为。器件串联后仍保持稳定的交流滤波能力,说明该结构在实际电路环境中具有良好的电化学一致性与推广潜力。

然而,该体系仍存在结构尺度上的内在约束。较大的碳管管径与相对宽松的管间间距虽然有利于离子快速迁移,却在一定程度上降低了单位面积内的有效界面面积,使可参与快速电荷积累的活性位点数量受限,储能界面利用率仍有提升空间。这种“传输通畅性”与“界面密度”之间的结构权衡关系,成为后续围绕“碳管基元”开展精细化“序构”调控研究的出发点,也为进一步优化容量—频率响应关系提供了设计方向。

3.2 碳管基元层级化拓展序构

针对单一碳管基元储能界面不足的问题,研究思路由“骨架构建”进一步转向“基元内部空间利用”,发展出 3D-CNT@CT^[83]。该策略通过将三维互连多孔阳极氧化铝模板预浸于 NiSO_4 溶液中,在孔壁上

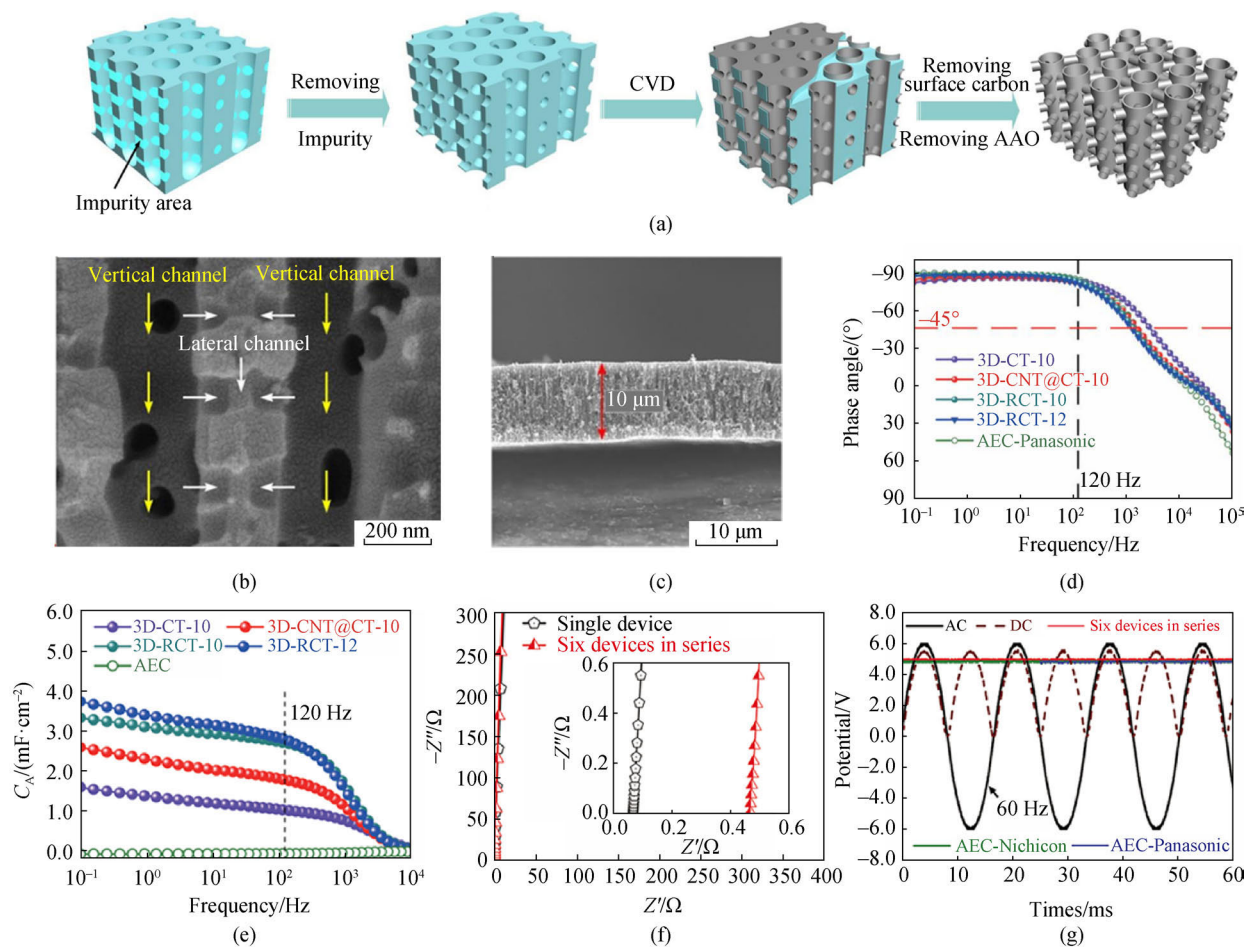


图6 3D-CT的结构构筑与电化学性能表征^[83]。(a)3D-CT的制备流程示意图；(b)三维互连多孔阳极氧化铝模板的扫描电子显微镜照片；(c)3D-CT的扫描电子显微镜照片；(d, e)3D-CT及3D-CNT@CT和3D-RCT变体的Bode图(d)和面积比电容随频率变化曲线(e)；(f)单个器件及六个器件串联后的Nyquist图；(g)六个器件串联组装后的交流线路滤波性能测试结果

Fig. 6 Structural construction and electrochemical performance of the 3D-CT^[83]. (a) Schematic illustration of the fabrication process of the 3D-CT; (b) Scanning electron microscopy (SEM) image of the three-dimensional interconnected porous anodic aluminum oxide template; (c) SEM image of the 3D-CT; (d, e) Bode plots (d) and areal capacitance as a function of frequency (e) of the pristine 3D-CT and its structural variants, including 3D-CNT@CT and 3D-RCT architectures; (f) Nyquist plots of a single device and six devices connected in series; (g) AC line-filtering performance of six devices connected in series

引入均匀分布的Ni催化位点,从而实现小直径碳纳米管的原位生长;同时依托模板限域构筑外层大直径碳管,形成碳管内填充碳纳米管的3D-CNT@CT结构(图7a~图7c)。其中,外层大直径碳管提供低曲折度的离子传输主通道,保证高频条件下电解质的快速迁移;内嵌的小直径碳纳米管则显著提升单位面积内的双电层活性界面,增加可参与快速吸附/脱附过程的有效活性位点。由于内部细管沿纵向分布,并未显著引入额外的横向阻滞或孔道堵塞,因此宏观离子传输路径长度与结构曲折度基本保持不变。

如图6d所示,该结构在120 Hz下仍保持接近理想电容的行为,说明内部层级化序构未显著增加阻抗成分。此外,面积比电容随频率变化曲线显示,

在120 Hz下的面积比电容由原始结构的 $1.04 \text{ mF}\cdot\text{cm}^{-2}$ 提升至 $1.81 \text{ mF}\cdot\text{cm}^{-2}$ (图6e),表明单位面积内有效储能界面得到实质性拓展。以上结果表明该层级化序构在保持电子与离子双连续传输框架的前提下,有效提高了界面电荷积累密度,实现了对“界面数量”与“传输通畅性”之间耦合关系的部分解耦。由此可见,对碳管基元进行内部层级化序构以扩展有效储能面积,是提升碳管基滤波电极容量性能而不显著牺牲高频响应的重要途径之一。

3.3 基元表面粗糙化序构

除内部空间开发外,研究者亦从基元表界面工程角度出发,发展出3D-RCT^[83]。通过利用 KMnO_4 处理初始的三维互连碳管网格膜,在碳管管壁上构

筑丰富的纳米孔隙与边缘缺陷(图 7d), 可有效提升在高频下电解质离子可及的电化学活性面积。不同于以内部填充或管内生长为代表的体相调控策略, 该策略主要作用于碳管基元的表层区域, 通过化学键连接的一体化导电骨架得以完整保持, 从而在有

效扩展电化学活性位点的同时, 确保了电子传导路径的连续性。该策略使 120 Hz 下的面积比电容提升至 $2.81 \text{ mF} \cdot \text{cm}^{-2}$, 且相位角达到 -80.5° (图 6d~图 6e), 验证了基于表面序构的“活性位点增加-输运路径保持”协同策略对提升高频储能性能的关键作用。

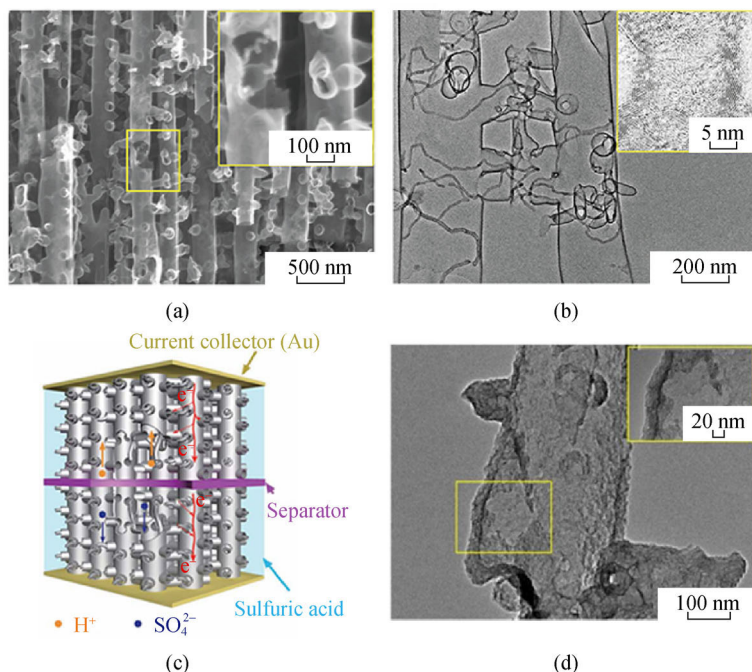


图 7 结构改性三维互连碳管网格膜的形貌及器件构型^[83]。(a~c)3D-CNT@CT 的扫描电子显微镜照片(a, b)和基于该结构的超级电容器组装示意图(c); (d)3D-RCT 的扫描电子显微镜照片

Fig. 7 Morphology and device configuration of structurally modified three-dimensional interconnected carbon tube grid^[83]. (a~c) SEM images of the 3D-CNT@CT (a, b) and schematic illustration of the supercapacitor assembled based on this architecture (c); (d) SEM image of the 3D-RCT

3.4 基元“套娃”式叠构

为满足更高面积比电容的需求, 研究进一步突破单层碳管结构的限制, 发展出 3D-MLCT^[84]。该策略借鉴“俄罗斯套娃”理念, 在三维多孔阳极氧化铝模板限域下, 通过多步化学气相沉积碳层与原子层沉积牺牲层交替进行, 构筑多层取向一致的碳管网络, 并最终获得了单层(3D-3LCT)、双层(3D-DLCT)及三层(3D-TLCT)有序同轴碳管结构。扫描电子显微镜照片(图 8a 和图 8b)清晰地显示了多层碳管在径向方向上规则嵌套且保持轴向高度取向, 未出现明显塌陷或错位; 氮气吸附-脱附等温线(图 8c)表明该结构在提升碳管密度的同时仍保留连续开放的孔道体系, 为电解质离子渗透提供保障。该多层同轴套娃结构并非简单增加界面, 而是在保持纵向取向一致性与横向电子网络连续性的前提下, 实现径向方向上的有序扩展, 提高单位面积内碳管基元数量及有效储能界面密度。

电化学测试结果进一步验证了这一结构优势。随着同轴层数增加, 电极面积比电容持续提升, 其中, 三层结构在 120 Hz 下实现约 $3.08 \text{ mF} \cdot \text{cm}^{-2}$ 的面积比电容, 且相位角保持在 -80° 以下, 较单层结构显著提高, 亦较此前层级填充结构的面积比电容($1.81 \text{ mF} \cdot \text{cm}^{-2}$)提升超过 70%。这一结果表明, 通过径向有序叠构扩展碳管基元数量, 可以在不显著牺牲频率响应的前提下实现容量的进一步跃升。

3.5 垂直碳管基元横向致密化序构

除在径向对碳管基元实施有序叠构外, 研究视角进一步延伸至垂直碳管基元在横向空间中的致密化排布。Chen 等^[85]通过系统调控阳极氧化电压、电解液组成及温度, 实现了对三维多孔阳极氧化铝模板参数的精确调节, 成功将垂直孔孔径与孔间距分别控制在 70~250 nm 与 110~450 nm 之间。如图 8d 所示, 随着碳管管径和管间距的减小, 单位体积内

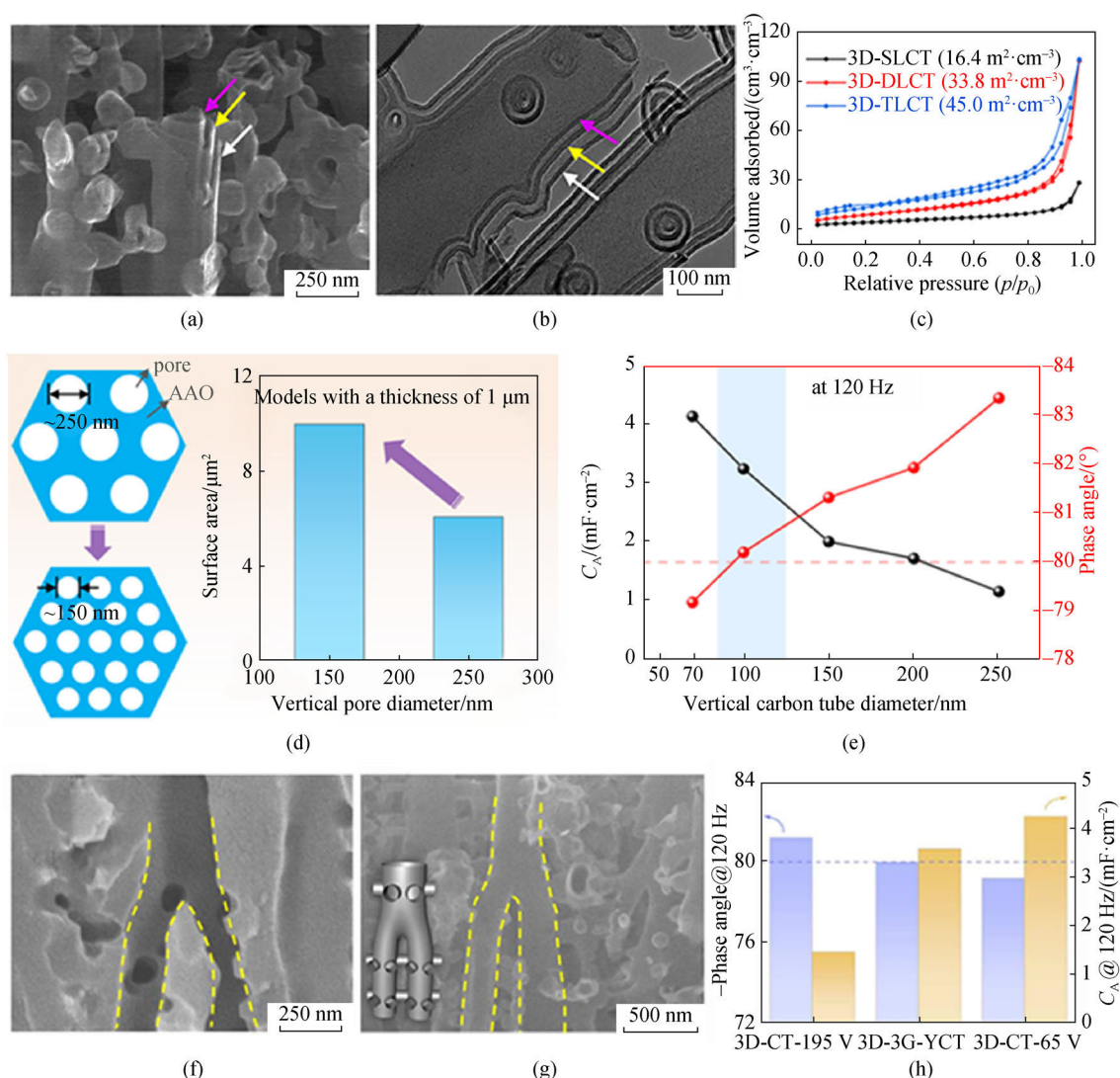


图8 不同结构调控策略下三维互连碳管网格膜的形貌与电化学性能。(a-c)^[84]3D-MLCT的扫描电子显微镜照片(a, b)和氮气吸附-脱附等温线(c); (d, e)^[85]3D-CACT的表面积图(d)和面积比电容与相位角随碳管管径变化曲线(e); (f-h)^[86]3D-YCT的扫描电子显微镜照片(f, g)和120 Hz下面积比电容及相位角(h)

Fig. 8 Morphology and electrochemical performance of three-dimensional interconnected carbon tube grids under different structural regulation strategies. (a-c)^[84] SEM images of the 3D-MLCT (a, b) and the corresponding nitrogen adsorption-desorption isotherms (c); (d, e)^[85] Specific surface area of the 3D-CACT (d) and the areal capacitance and phase angle as a function of carbon tube diameter (e); (f-h)^[86] SEM images of the 3D-YCT (f, g) and the areal capacitance and phase angle at 120 Hz (h)

的碳管基元数量显著增加,使得电极在120 Hz下的面积比电容升至 $3.23 \text{ mF} \cdot \text{cm}^{-2}$ 。

然而,致密化排布在强化储能密度的同时,也对其高频动力学性能产生了一定影响。如图8e所示,虽然面积比电容随管径减小而持续增大,但同时相位角也呈现出一定的偏移。当孔径尺度进入亚百纳米范畴时,离子在高频条件下的快速进入与嵌出过程逐渐受到空间限域的制约,导致传输阻抗上升。尽管该体系在120 Hz下仍保持了 -80.2° 的优异相位角,表现出极佳的滤波潜力,但其性能演化曲

线呈现出单纯依赖横向致密化的本征瓶颈:即储能位点的增加以牺牲部分离子迁移效率为代价。这一发现不仅揭示了“容量-动力学”在限域空间内的竞争机制,也为后续引入仿生Y-形分支结构以兼顾快速离子传输与高密度电荷存储提供了关键的实验依据。

3.6 Y-形分支基元结构

为突破结构“致密化”与“传输受阻”的矛盾,研究者制备了模拟生物运输系统的Y-形分支结构^[86]。针对小孔径碳管传质受阻的瓶颈,Y-形分支结构构

建了一套层级化的孔道体系。研究者利用阳极氧化铝孔道分支的形成规律——即当阳极氧化电压降低至初始电压的 $1/\sqrt{2}$ 时发生Y分叉的特性,通过精准调控阶梯电压与后续的湿化学刻蚀技术,成功构筑了具有横向贯通孔道的三维Y-形分支多孔阳极氧化铝模板(图8f)。如图8g所示,基于该模板生成的碳管网格膜展现出“Y”形分支特征(3D-YCT)。在该设计中,较大直径的主干碳管作为“一级动力学高速公路”,确保电解质离子能够低阻快速地迁移至电极内部,而由主干衍生出的多级小直径分支碳管,则在维持低曲折度的同时,提供高的有效储能界面。

电化学测试结果验证了这种仿生结构的优越性。如图8h所示,相比于单一的大孔径或致密小孔径结构,Y-形分支结构在维持优异频率响应的同时,显著提升了电荷存储能力。该3D-YCT在120 Hz下不仅保持了约 -80° 的相位角,更获得了该系列研究中最高水平的 $3.60\text{ mF}\cdot\text{cm}^{-2}$ 面积比电容。这种仿生结构完美平衡了储能界面密度与离子运输效率之间的矛盾,既规避了横向致密化导致的离子“拥堵”现象,又突破了单一取向结构容量不足的限制,为电力电子系统的进一步微型化提供了极具竞争力的结构范式。

4 结论与展望

超级电容器作为下一代轻量化、微型化电子电路中极具潜力的滤波元件,其性能突破始终聚焦于“超快频率响应”与“高面积比电容”的协同优化。纵观滤波超级电容器电极的发展历程,其核心逻辑已由单纯的材料组成调控,演进为以微观结构理性设计为导向的“序构”范式。本文基于结构维度与空间组织方式,对碳基滤波电极进行系统梳理,包括二维平面结构、三维随机大孔结构以及高度垂直取向结构,揭示了结构有序性在调控电荷输运动力学中的关键作用;并重点指出,以“碳管”为功能基元“序构”的三维互连网格电极,是破解“容量密度-频率响应”本征矛盾的有效途径。

从构建一体化电子/离子传输网络出发,到利用碳管内填细碳纳米管与表面粗糙化实现基元层级化调控,再到通过同轴多层套构与横向致密化进一步提升空间利用率,以及最终的仿生Y-形分支设计实现多级连通优化。这一系列结构演变清晰表明,滤波超级电容器性能的持续提升并非单纯依赖比表面积无限增大,而是源于离子迁移通畅性(低曲折度通道)与界面电荷存储能力(高有效活性表面)之间的

最优平衡。这种基于碳管基元的“序构化互连网络”,不仅拓展了容量与频率响应的性能边界,也为电荷的超快动力学输运提供了稳定可复制的结构范式,有力推动了超级电容器由传统储能向高频滤波应用的领域跨越。

尽管功能基元序构策略已展现出显著潜力,但其向深化理论与产业化应用的转化仍面临多重挑战。首先,在理论表征维度,亟须建立一套量化描述电极“序构度”的普适性参数体系,以实现不同结构之间的可比性并指导理性设计。具体而言,未来应引入曲折因子(反映离子迁移路径偏离理想直线程度)、连通性参数(表征横向连接特征,如横向连接密度)、取向角分布(描述垂直排列一致性),以及空间分布参数(反映结构均匀性)等几何参数,并结合电化学阻抗谱及多物理场仿真模拟,构建其与相位角、截止频率及特征时间常数之间的定量关联关系。这种从“几何参数”到“动力学响应”的函数化表达,将推动电极设计由“经验寻优”向“数字化定制”跨越。

其次,在输运机制解析方面,建立电子传导与离子扩散贡献的解耦评估模型至关重要。例如,可通过“结构恒定—本征电导变量”的实验策略,在保持三维结构与负载一致的前提下调控材料结晶性,以区分电子传导对频率响应的贡献。此外,借助基于真实结构的有限元模拟,通过分别调控电子电导率(σ_e)与离子扩散系数(D_{ion})开展参数敏感性分析,从而建立“结构-传输参数-性能”之间的因果关系,为序构结构的定向优化提供理论依据。

在结构设计层面,未来研究应聚焦于厚电极与高负载条件下的“动力学增益维持”。针对高质量负载引发的动力学“屏蔽”,需通过构筑多维互连网络、优化跨尺度孔径分布等手段,确保电子/离子在宏观电极深层依然具备极低的传输阻抗,实现面积比电容与频率响应的深度解耦与同步提升。

最后,在工程化应用维度,大面积、高均匀序构电极的可规模化制备是产业化的核心瓶颈。未来需发展更具普适性与工艺容错性的制备技术,以确保厘米级实验样品向工业级电极体系的平稳过渡。此外,还需进一步解决封装工艺与电路匹配问题,包括降低电极/集流体界面电阻、抑制高频条件下的寄生效应,以及优化器件在目标频率(如120 Hz及更高频率)下的阻抗特性。同时,三维多孔结构在长期循环、电热耦合及机械变形条件下的稳定性,以及多器件集成过程中的连接可靠性,也将对其实际

应用产生重要影响。

总体而言, 功能基元序构不仅是结构优化的工具, 更是连接材料设计、器件性能与应用场景之间的系统性研究范式。随着多尺度结构调控能力的提升以及原位表征与理论模拟的持续发展, 滤波超级电容器电极的研究将由经验驱动逐步迈向机制驱动与定量设计, 并在电子系统小型化与高集成化进程中展现出更为广阔的应用前景。

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Research Progress on Carbon-Tube Units Ordering Electrodes for High-Performance Line-Filtering Supercapacitors

LI Pei^{1,2}, ZHOU Ziyang^{1,2}, JIANG Suxia^{1,2}, XU Xiaojian^{1,2}, HAN Fangming^{1,2}, MENG Guowen^{1,2}

(1. Institute of Solid State Physics, Hefei Institutes of Physical Science, Chinese Academy of Sciences, Hefei 230031, China;

2. Science Island Branch, Graduate School of USTC, Hefei 230026, China)

Extended Abstract

With the rapid advancement of electronics toward lighter and more miniaturized designs, conventional aluminum electrolytic

capacitors, constrained by intrinsically low areal/volumetric capacitance and mechanically rigid architectures, struggle to meet the demands for high-performance filtering in modern electronic circuits. Supercapacitors are regarded as promising alternatives to aluminum electrolytic capacitors due to their high specific capacitance, rapid charge-discharge capability, and excellent cycling stability. However, the intrinsically disordered porous architecture of conventional electrodes gives rise to severe mass-transport limitations. The highly tortuous ion migration pathways severely restrict transport kinetics, leading to a fundamental trade-off at 120 Hz, where fast frequency response and high energy density cannot be simultaneously achieved, thereby significantly impeding their practical application. In recent years, the rational design of electrode microstructures, particularly the precise ordering control of the spatial orientation and interconnection of nanoscale functional building units, emerges as an effective strategy to address this contradiction. This review represents carbon-based filtering electrodes based on structural dimensionality and spatial organization, encompassing two-dimensional planar architectures characterized by high packing density, three-dimensional macroporous structures dominated by random diffusion, and highly vertically aligned configurations featuring directional transport. The critical role of structural ordering in regulating charge-transport kinetics is thereby elucidated. On this basis, a special emphasis is placed on three-dimensional ordered electrode systems constructed from carbon tubes as functional units, *i.e.*, three-dimensional interconnected carbon tube grids. Their structural advantages in constructing low-resistance, continuous electronic conduction networks and low-tortuosity, directional ion transport channels are analyzed. For the intrinsic logic of “structural ordering–ion transport–high-frequency filtering performance”, this review discusses the effects of various structural evolution strategies (*i.e.*, self-supporting scaffold construction, hierarchical regulation within building units, surface/interface engineering, concentric nesting, lateral densification, and Y-branch design) on the ion transport behavior and filtering performance at 120 Hz. In addition, the existing research progress and future development trends are also summarized to provide theoretical insights and structural design guidelines for high-performance, miniaturized electrochemical filtering devices.

Summary and Prospects As a highly promising filtering component for next-generation lightweight and miniaturized electronic circuits, supercapacitors have pursued performance breakthroughs centered on the synergistic optimization of ultrafast frequency response and high areal capacitance. Reviewing the evolution of electrode design for filtering supercapacitors reveals a paradigm shift from mere compositional engineering toward an ordering-driven paradigm guided by rational microstructural design. In this review, carbon-based filtering electrodes are systematically categorized according to structural dimensionality and spatial organization, including two-dimensional planar architectures, three-dimensional randomly macroporous frameworks, and highly vertically aligned configurations, thereby elucidating a pivotal role of structural ordering in governing charge-transport kinetics. In particular, three-dimensional ordered grid electrodes constructed from carbon tubes as functional building units are identified as an effective strategy to resolve the intrinsic trade-off between capacitance density and frequency response.

From the construction of an integrated electron/ion transport networks, the systematic structural evolution clearly demonstrates that the continuous improvement of filtering supercapacitor performance does not arise merely from the unbounded enlargement of specific surface area, but rather from achieving an optimal balance between ion transport accessibility (*i.e.*, low-tortuosity pathways) and interfacial charge-storage density (*i.e.*, highly active surfaces). Such carbon-tube-based, ordered interconnected networks extend the performance limits in terms of capacitance and frequency response, and provide a robust and reproducible structural framework for ultrafast charge transport, thereby effectively propelling the transition of supercapacitors from conventional energy-storage devices toward high-frequency filtering applications.

Despite the remarkable potential of functional unit ordering strategies, some substantial challenges remain in translating this concept toward deeper theoretical understanding and practical implementation. From a theoretical perspective, there is an urgent need to establish a universal, quantitative descriptor system for assessing the “degree of ordering” in electrode architectures. Specifically, key geometric descriptors should include tortuosity (*i.e.*, quantifying deviation of ion transport pathways from ideal straight channels), connectivity parameters (*i.e.*, lateral junction density), orientation distributions (*i.e.*, reflecting alignment along the current collector normal), and spatial uniformity metrics. The quantitative relationships can be established between structural parameters and kinetic metrics such as phase angle, cutoff frequency, and characteristic time constants *via* integrating these descriptors with electrochemical impedance spectroscopy and multiphysics simulations. Such a framework linking geometric descriptors to dynamic electrochemical responses will enable a transition from empirical optimization toward digitally guided electrode design.

In terms of transport mechanism analysis, it is particularly important to develop methods capable of decoupling the respective contributions of electronic conduction and ionic diffusion. For instance, an experimental strategy involving “fixed structure-variable conductivity” can be employed, whereby the crystallinity of the material is selectively tuned, while maintaining identical three-dimensional architectures and mass loading, thereby isolating the influence of electronic transport on the frequency response. In parallel, finite element simulations based on realistic structural models with independent variation of electronic conductivity (σ_e) and ionic diffusivity (D_{ion}) can be used to perform sensitivity analyses and establish causal relationships among structure, transport parameters, and electrochemical performance. These methods provide a critical theoretical guidance for the targeted optimization of ordered electrode architectures.

At the structural design level, future efforts should focus on maintaining kinetic advantages under thick-electrode and high-mass-loading conditions. To mitigate the transport limitations caused *via* increased loading, strategies such as constructing multidimensionally interconnected networks and optimizing hierarchical pore size distributions are required to ensure ultralow transport resistance for both electrons and ions, even in the deep regions of electrodes. This is essential for achieving simultaneous

enhancement of areal capacitance and frequency response.

From an engineering perspective, the scalable fabrication of large-area, highly uniform ordered electrodes remains a key bottleneck in practical applications. Future developments should aim at synthesis strategies with improved generality and process tolerance to enable a smooth transition from centimeter-scale laboratory samples to industrial-scale electrode systems. In addition, some challenges relating to device packaging and circuit integration must be addressed, including minimizing the resistance at electrode/current collector interface, suppressing parasitic effects under high-frequency operation, and optimizing impedance characteristics at target frequencies (*e.g.*, 120 Hz and above). Furthermore, the structural stability of three-dimensional porous architectures under long-term cycling, electro-thermal coupling, and mechanical deformation, as well as the reliability of multi-device integration, will critically determine their feasibility in applications.

Overall, building-unit ordering represents not merely a structural optimization strategy, but a unifying paradigm that bridges materials design, device performance, and application scenarios. With continued advancement of multiscale structural regulation capabilities, *in-situ* characterization techniques, and theoretical modeling tools, a research on filtering supercapacitor electrodes is expected to evolve from empirically driven exploration toward mechanism-informed and quantitatively guided design, ultimately enabling a broad application in highly integrated and miniaturized electronic systems.

Keywords functional units; carbon tubes; ordered structures; filtering supercapacitors; electrode architecture design; electron–ion synergistic transport