Journal of Semiconductors
(2023) 44, 050201
doi: 10.1088/1674-4926/44/5/050201

RESEARCH HIGHLIGHTS

Micro-nano structural electrode architecture for high power energy storage

Xin Chao[1, ‡], Chengzhan Yan[2, ‡], Huaping Zhao[2], Zhijie Wang[3, †], and Yong Lei[2, †]

[1]Institute of Nanochemistry and Nanobiology, School of Environmental and Chemical Engineering, Shanghai University, Shanghai 200444, China

[2]Fachgebiet Angewandte Nanophysik, Institut für Physik & IMN MacroNano, Technische Universität Ilmenau, Ilmenau, 98693, Germany

[3]Key Laboratory of Semiconductor Materials Science, Beijing Key Laboratory of Low Dimensional Semiconductor Materials and Devices, Institute of Semiconductors, Chinese Academy of Sciences, Beijing 100083, China

Citation: X Chao, C Z Yan, H P Zhao, Z J Wang, and Y Lei, Micro-nano structural electrode architecture for high power energy storage[J]. *J. Semicond.*, 2023, 44(5), 050201. https://doi.org/10.1088/1674-4926/44/5/050201

The necessity and superiorities of micro-nano structural electrodes toward high power: Electrochemical energy storage (EES) technologies have achieved great success in portable electronics and electric vehicles owing to their environmental friendliness and cost effectiveness. With the promotional concepts such as the Internet of Things and ultra-high efficiency self-powered systems in recent years, there are substantial demand for superior EES systems, including but not limited to high-performance, miniaturization and multi-function[1–4]. In a particular EES cell, active materials are carried by electrodes as the basic building blocks of energy storage or release. Material innovation (includes composition, structure, size and morphology) has revealed remarkable energy density, power density and lifespan for associated devices in the lab setting of low mass loading slurry-coating electrodes[5]. However, as the loading increases, the trade-off between energy density and power density becomes arduous. High mass loading electrodes with adequate energy storage sites enhance energy supply but necessitate extended thicknesses. Unfortunately, a thicker slurry-coating electrode may suffer from sluggish kinetics, low active material utilization and poor mechanical stability, leading to inferior power output and short lifetime[6–7]. Therefore, subversive electrode architecture ought to be developed to support high power and long life under the guaranteed high energy density.

It has been realized that local optimization of the active material alone is far from being able to overcome the inherent disadvantages of its randomly individual stacking configurations. Considering the overall electrodes, integrating the micro-nano structures into an ordered conductive network not only maximizes the retention of their inherent advantages but also enables the coordination of performances, especially in providing high power while ensuring sufficient energy supply[8]. Micro-nano structural electrodes can be seen as the longitudinally regular deformations of the planar electrodes along with a larger specific surface area. This undoubtedly enhances the potential to load more active sites and maximize active material utilization. On this premise, high power is also attainable through the facilitation of ion and electron transport. Firstly, the pre-set free space between the micro-nano structures allows the ions in the electrolyte to reach the surface easily, which improves the decisive speed of ion transport in thick slurry-coating electrodes[9]. Besides, ion migration in the solid phase is still fast based on the micro-nano structural units. Secondly, electron transport is effectively facilitated by unobstructed electrical percolation networks. Micro-nano structural electrodes build on material preparation advances without the use of additional additives and binders so that the electron channels are not obstructed by inactive components[3]. As a result, the transport of electrons from the current collector to the active sites is improved as well. Since both ion and electron channels are optimized, the polarization is minimized as much as possible[6]. Up to these points, the reaction efficiency and power will be upgraded. Moreover, the current distribution within micro-nano structural electrodes can also be predicted and regulated by drawing on modeling simulation to ensure uniform current distribution at high power output and reduce the risk of structural collapse.

Recent progresses of micro-nano structural thin-film electrodes: Ordered arrays of micro-nano structures can be assembled on planar current collectors to design micro-nano structural thin-film (MNTF) electrodes. MNTF electrodes deliver large specific surface areas, fast ion and electron transport channels, and interactive electrolyte permeation networks[10]. The refinement of micro-nano manufacturing technology enables the integration of a variety of micro- and nano-structures such as lithography, templates, pulsed laser deposition (PLD), electrodeposition and so on[11, 12]. Typically, silicon can be designed on the microscopic scale as highly ordered arrays or patterns of rods, tubes, pores and so on. Hallot's group constructed silicon microtubes three-dimensional (3D) scaffolds (area enhancement factor (AEF) of 50–60) with different thicknesses (100–250 nm) coating of spinel-type $LiMn_2O_4$ (LMO) films by atomic layer deposition (ALD) for lithium ion storage (Fig. 1(a))[13]. Among them, the specific areal capacity of 100 nm thick LMO film coating silicon microtubes scaffold is close to 180 μA·h/cm^2 (Fig. 1(b)), which is one to two orders of magnitude higher than that of 3D electrodes fabricated by all ALD deposition techniques while maintaining suffi-

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Xin Chao and Chengzhan Yan contributed equally to this work and should be considered as co-first authors.

Correspondence to: Z J Wang, wangzj@semi.ac.cn; Y Lei, yong.lei@tu-ilmenau.de

Received 11 JANUARY 2023; Revised 1 FEBRUARY 2023.

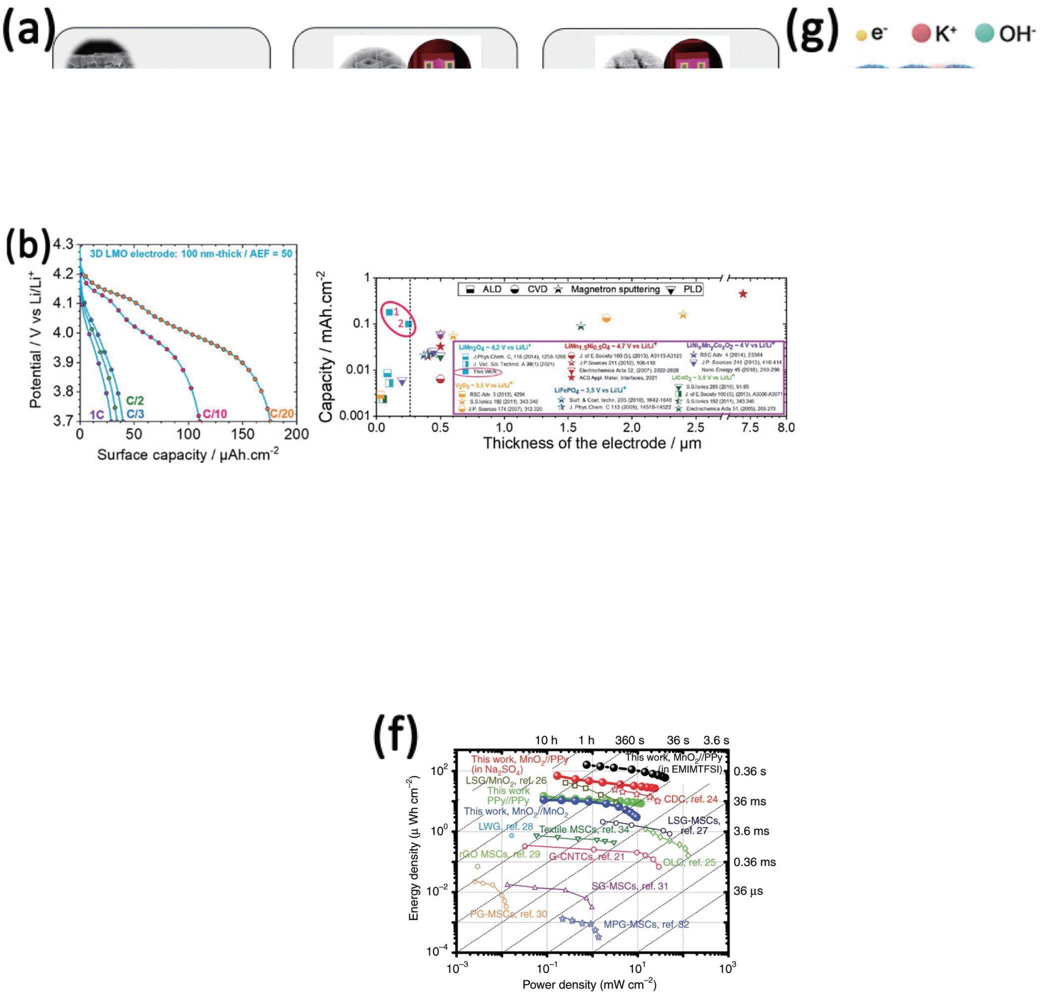

Fig. 1. (Color online) (a) Three main steps to deposit LMO films on planar and three-dimensional substrates. (b) Galvanostatic discharge plot of a 3D LMO electrode (100 nm thick/AEF = 50) at different C-rates. (c) Performance of the 3D LMO electrodes made by atomic layer deposition regarding state-of-the-art values of thin film electrodes deposited by magnetron sputtering, pulsed laser deposition (PLD), chemical vapor deposition (CVD), and ALD methods. Reproduced with permission[13], Copyright 2022, Wiley-VCH. (d) Illustration of the HAN fabrication process. (e) Areal energy and power densities of HAN@SnO_2@MnO_2//HAN@SnO_2@PPy asymmetric MSCs measured at different current densities. (f) Performance comparison of MSCs based on HAN nanoelectrodes with some reported MSCs. Reproduced with permission[14], Copyright 2020, Nature Publishing Group. (g) The working principle of Ni microelectrode. (h) Ragone plots of the Ni–Zn microbattery. Reproduced with permission[15], Copyright 2021, Wiley-VCH. (i) Comparison of a usual SnO_2 NW electrode (left-hand) with a new design of micropatterned NW electrode (right-hand). Reproduced with permission[16], Copyright 2022, American Chemical Society.

cient power density (Fig. 1(c)). Decreasing the scale of the structure from micron to nanoscale can further increase the specific surface area of the current collector to provide more loading sites within the same height. An ultrathin and rigid honeycomb alumina nanoscaffold (HAN) (Fig. 1(d)) was designed by Lei's group for micro-supercapacitor (MSC)[14]. The vertically aligned HAN with ultra-high cell density and ultrathin nanoscaffold has a surface area enhancement factor of 240 with no aspect ratio limitation, allowing for effective ion transport and large electroactive surface area in a limited footprint of the microelectrode. The robust HAN was first coated with tin oxide (SnO_2) and manganese oxide (MnO_2) or polypyrrole (PPy) was then electrodeposited to generate HAN@SnO_2@MnO_2 or HAN@SnO_2@PPy electrodes, respectively. The peak energy density of the as-prepared sandwich-type asymmetric MSCs is about 4 times higher than that of carbide-derived carbon (CDC) based MSCs, but has a similar peak power density (Fig. 1(e)), which is one of the best performances of advanced MSCs (Fig. 1(f)). The incorporation of micro-nano structures is also beneficial to improve the electrochemical reaction kinetics under aqueous and solid state electrolytes. Recently, Zhu *et al.* constructed an aqueous Ni–Zn microbattery (MB) with ultrahigh rate capability and durability by in situ reconstruction of epitaxial phase nanoporous Ni (Fig. 1(g))[15]. The surface reconstruction of interconnected nanoporous Ni enables sufficient conductivity and reaction sites, and the as-prepared Ni–Zn MB has a high power density of 320.17 mW/cm^2 and a maximum energy load of 0.26 mW·h/cm^2 (Fig. 1(h)), achieving a breakthrough in the performance of aqueous MB. For years, solid-state electrolytes are considered to be one of the effective strategies to solve battery safety, but their poor contact with the electrode interface often leads to low rate performance in addition to their own low conductivity. In order to increase the contact interface between the electrode and the solid electrolyte, Kim *et al.* developed imaged SnO_2 nanowire arrays on the stainless steel

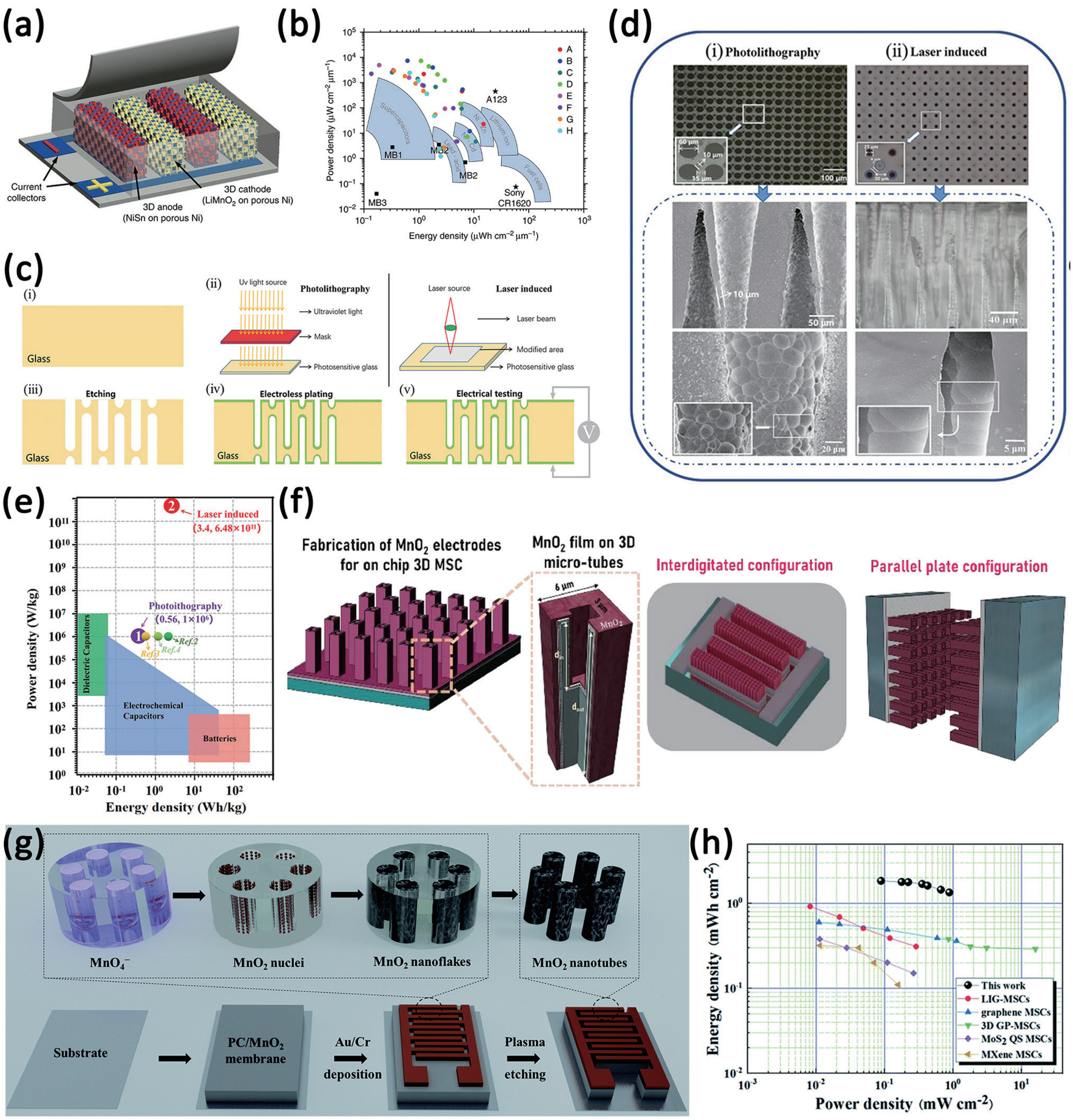

Fig. 2. (Color online) (a) Schematic showing Li-ion MBs with interdigital patterns of 3D bicontinuous nanoporous NiSn anodes and LiMnO$_2$ cathodes. (b) Ragone plot of microbattery cells and conventional power technologies. Reproduced with permission[21], Copyright 2013, Nature Publishing Group. (c) The process sequence to prepare 3D interdigital electrodes dielectric capacitor. (d) SEM and optical microscope of 3D interdigital electrodes dielectric capacitor prepare by photolithography and laser induced. (e) Ragone plot showing high-energy, high-power densities of the 3D interdigital electrodes dielectric capacitor. Reproduced with permission[22], Copyright 2022, Wiley-VCH. (f) Schematic representation of MnO$_2$-coated Si microtube electrodes for on-chip MSCs with interdigitated and parallel plate configurations. Reproduced with permission[23], Copyright 2021, Elsevier. (g) Schematic illustration of the fabrication of 3D MnO$_2$ NTA-based flexible MSCs. (h) Ragone plot comparing the 3D MnO$_2$ NTA-based MSCs with other 2D and 3D MSCs. Reproduced with permission[24], Copyright 2022, Royal Society of Chemistry.

through photolithography and controlled the array formation by modifying different area and spacing of the SnO$_2$ nanowires patterns (Fig. 1(i))[16]. Tailored designs in the microchannels formed between the patterns improve the interfacial bonding between the electrolyte and electrodes, enhancing the capacity of solid-state lithium-ion batteries (LIBs) at high charge-discharge rates and long cycles.

Recent progresses of micro-nano structural interdigitated electrodes: Different from the sandwich-type device assembly of thin-film electrodes, the interdigitated electrodes arrange the cathodes and anodes in a finger or comb shape on the same plane and remove the traditional separator[17–19]. The size and spacing of the electrode units can reach the micron level, and the micro-spacing enables the convenient and rapid transmission of electrolyte ions in narrow gaps to provide ultra-high power density. In order to further increase the exposed sites on the electrode surface, the micro-nano structure arrays can be combined with the interdigitated arrangement to realize EES devices with high energy density and power density[20]. To ensure high power density without sacrificing energy density, a strategy to simultaneously optimize ion and electron transport in interdigitated electrodes is proposed. Pikul and his colleagues developed highly conductive bicontinuous nanostructures using porous templates[21].

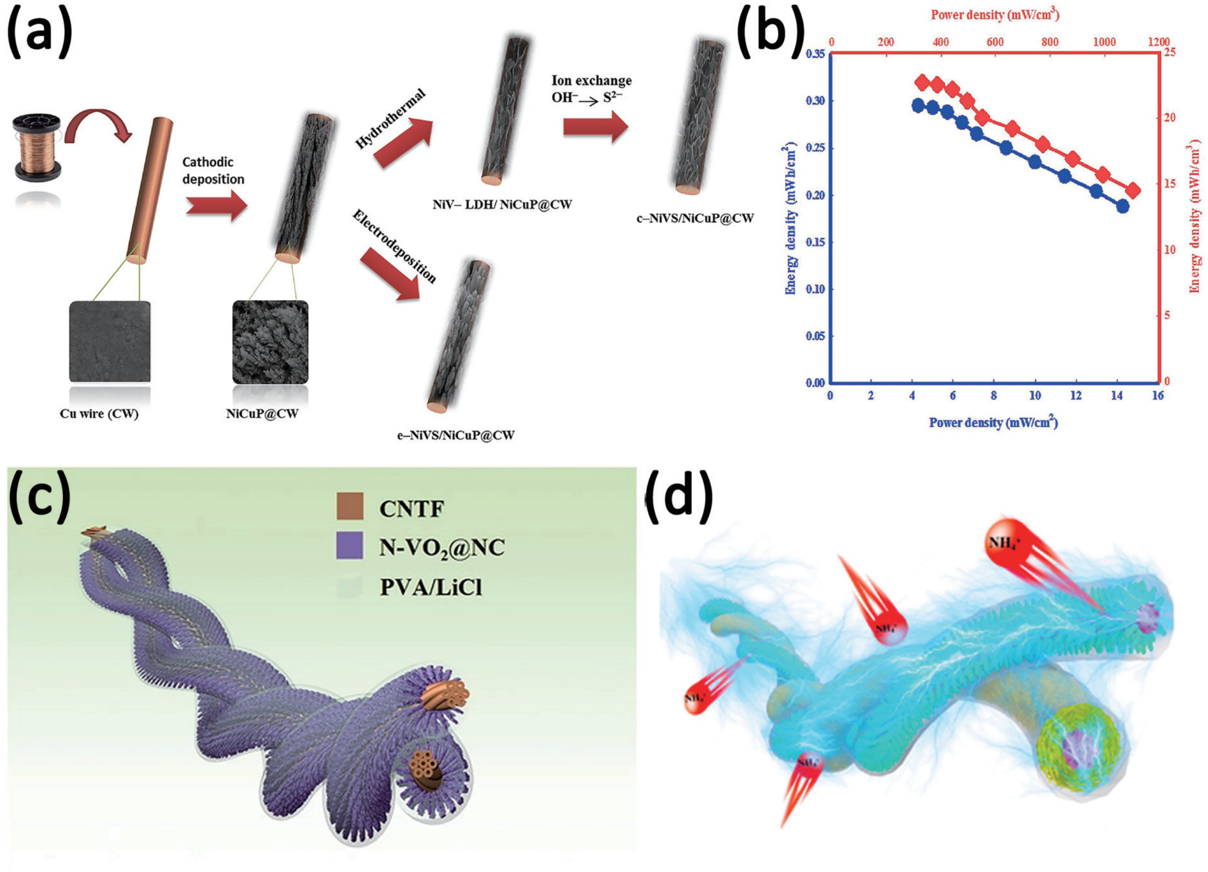

Fig. 3. (Color online) (a) Schematic illustration for preparation of c-NiVS/NiCuP and e-NiVS/NiCuP on Cu wire. (b) Areal and volumetric Ragone plot of the asymmetric device. Reproduced with permission[29], Copyright 2020, Elsevier. (c) Schematic diagram of the FNSC. Reproduced with permission[30], Copyright 2022, Wiley-VCH. (d) Schematic diagram of fiber type supercapacitors. Reproduced with permission[31], Copyright 2022, American Chemical Society.

The NiSn anode and the lithiated LMO cathode were conformally coated on the interdigitated highly porous Ni scaffold (Fig. 2(a)), respectively and as-obtained MB have a power density as high as 7.4 $mW/cm^2/\mu m$, which is 2000 times higher than other similar MBs (Fig. 2(b)). In particular, some specific technique realizes the transformation of two-dimensional thin film electrode to interdigitated electrode simultaneously with micro-nanostructures. For example, a novel micro-nano structural interdigitated electrode (Fig. 2(c)) with high depth-to-diameter ratio porous (diameter and depth of pores are 25 and 490 μm, respectively) were fabricated on photosensitive glass substrates by laser induction (Fig. 2(d))[22]. Then, the Ni electrodes were grown on the inner wall of porous glass by electroless plating thus transforming the planar interdigitated structure into a spatial interdigitated structure with higher specific surface area (422 times the specific capacitance of the planar structure). Due to the laser-induced smooth hole inner wall and pore shape electrode, an energy density of 3.4 W·h/kg and a power density of 6.48×10^{11} W/kg were achieved (Fig. 2(e)). Moreover, it is possible to combine micron Si structure with interdigitated electrodes as technology advances. A Si microtube with an AEF of 47 was used to construct the three-dimensional structure electrode of MSC. Bounor *et al.* used ALD to sequentially deposit Al_2O_3 layers (50 nm) and Pt layers (50 nm) on Si microtubes[23]. Then a 580-nm-thick nanostructured MnO_2 film was electroplated on the $Si/Al_2O_3/Pt$ interdigitated microelectrodes by pulsed electrodeposition to maximize energy density while maintaining a high power density (> 1 mW/cm^2) (Fig. 2(f)). Recently, a porous MnO_2 nanotube arrays (NTAs) assembled by intersecting nanoflakes was applied in on-chip MSC devices by a poly-dimethylsiloxane (PDMS)-assisted transfer method (Fig. 2(g))[24]. Due to the sufficient space between porous nanotubes, the accessible surface of ions is greatly increased while shortening the diffusion path of ions. The porous nanotube structure not only shortens the diffusion path of ions but also greatly increases the accessible surface of ions, so as to greatly improve the utilization rate of MnO_2. The associated MSCs achieve high specific area and volume energy densities (1.9 $\mu W{\cdot}h/cm^2$ and 2.38 $mW{\cdot}h/cm^3$) (Fig. 2(h)).

Recent progresses of micro-nano structural fiber electrodes: Flexible fiber electrodes emergence meets the demand for wearable electronic devices in the fields of medical care, sports, infotainment and so on. They coupled with flexible electrolytes form a monolithic one-dimensional structure and further assembled into devices in parallel, twisted and coaxial structures. Moreover, the fiber electrodes with sufficient energy/power density and molding flexibility are possible to compound with other functional electronic devices to form multifunctional integrated flexible electronic devices[25–28]. NiVS/NiCuP nanostructures (Fig. 3(a)) were fabricated on copper wires as fiber electrodes for high-performance MSC[29]. For the first time, porous 3D NiCuP layers were prepared on Cu fibers by electrodeposition. Then NiVS electroactive material was covered on the porous three-dimensional NiCuP layer by hydrothermal method, followed by sulfuration treatment. The proposed electrode was able to maintain an energy density of 188 μW·h/cm^2 even at a high power density of 14.3 mW/cm^2 (Fig. 3(b)), which is significantly higher than other reported fiber SCs. However, fiber electrodes generally suffer from low specific capacitance, slow ion diffusion and poor electrical conductivity, whereas the introduction of micro-nano structures may yield superior energy density and power density. Usually, the array of micro-nano structures is loaded on the surface of the flexible conductive current collector by vertical arrangement. Furthermore, micro-nano heterostructural composites are also feasible. To this end, Guo *et al.* used a combination of doping and micro-nano structural electrode construction (Fig. 3(c)) to prepare nitrogen-doped vanadium dioxide/nitrogen-doped carbon (N-VO$_2$@NC@CNTF) fiber electrodes by growing VO$_x$ on PPy nanowires of carbon nanotube fibers (CNTF)[30]. The nitrogen-doped PPy-derived carbon is not only able to act as a conductive scaffold for enhancing the ion transport process and conductivity, but also to regulate the loading of the VO$_2$. On the basis of the synergistic effect of doping and conducting network, all-solid-state fiber-shaped nonpolarity supercapacitors (FNSC) assembled from LiCl/PVA gel electrolyte and N-VO$_2$@NC@CNTF fiber electrodes achieved an energy density of 51.8 mW·h/cm^3 at a power density of 1000 mW/cm^3. Meanwhile, the fiber-shaped aqueous zinc-ion batteries (FAZIB) assembled from N-VO$_2$@NC@CNTF cathode, Zn@CNTF anode and CMC/ZnSO$_4$ gel electrolyte obtained an excellent energy density of 313.13 mW·h/cm^3 at a power density of 142 mW/cm^3. Besides, a multifunctional core-shell heterostructure electrode was formed by anchoring MoS$_2$ nanosheets on TiN nanowires grown on CNTF (MoS$_2$@TiN/CNTF) (Fig. 3(d))[31]. The uniformly distributed MoS$_2$ nanosheets are used as the shell to provide abundant electrochemical active sites, and the neatly arranged TiN nanowires are used as the conductive core. The flexible fiber-shaped ammonium-ion asymmetric supercapacitors (FAASCs) assembled with MoS$_2$@TiN/CNTF anode, MnO$_2$/CNTF cathode and NH$_4$Cl-PVA gel electrolyte twist achieved an energy density of 195.1 μW·h/cm^2, even a high specific volume energy density of 144.4 μW·h/cm^2 is available at a high power density of 20 mW/cm^2.

Conclusion and prospects: With regard to electrode structural investigations, finite element simulation modeling is another important method to optimize electrode or device design and clarify structure-performance relationships[32–34]. Some of the micro-nano structural electrodes have been modeled to simulate reaction degree of the electrode units and the ion concentration between electrodes under different charge/discharge states to predict the optimal structure parameters. However, experimental and simulation combined studies are still lacking. One reason may be that some high performance materials are difficult to match with special micro-nano structure preparation processes. In conclusion, the introduction of micro-nano structures into the electrode architecture of thin film electrodes, interdigital electrodes and fiber electrodes is an effective strategy to ensure the retention of sufficient energy density at high power density. Nevertheless, the fabrication of micro-nano structural electrodes is rather demanding, and their reproducibility and reliability still need to be studied[35–38].

Acknowledgements

The authors acknowledge support from the National Natural Science Foundation of China (22076116), German Research Foundation (DFG: LE 2249/15-1) and the Sino-German Center for Research Promotion (GZ1579). Chengzhan Yan would like to acknowledge the China Scholarship Council for the financial support.

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Xin Chao got her B.S. degree from Henan University of Technology in 2021. Now she is a M.S. student at Shanghai University. Her research interests focus on the construction and functionalization of nanomaterials for energy storage devices.

Chenghuan Yan received his M.S. degree in chemistry from Wenzhou University in 2020. He is currently a PhD student under the supervision of Prof. Yong Lei at the Technical University of Ilmenau in Germany. His research interests focus on designing and synthesizing nanomaterials for electrochemical energy storage.

Huaping Zhao obtained his PhD in Materials Science from the State Key Laboratory of Crystal Materials of Shandong University in 2007. Following two years of postdoctoral research at the Institute of Chemistry (Chinese Academy of Sciences, 2007–2009), he was employed as a scientist by the University of Muenster from 2009 to 2011. Since 2012, he has been a senior scientist (permanent) in Prof. Yong Lei's group at the Technical University of Ilmenau, Germany. His current research focus is the design and fabrication of functional nanostructures for energy storage and conversion.

Zhijie Wang received his B.S. degree in 2004 from Zhejiang University and Ph.D. degree in 2009 from the Institute of Semiconductors, Chinese Academy of Sciences. After four years of postdoc research in the University of Wyoming and the University of Michigan, he worked as a senior scientist and a junior group leader at the Ilmenau University of Technology (Germany) in the 3D Nanostructuring Group of Prof. Yong Lei since 2013. He is currently a professor in the Institute of Semiconductors, Chinese Academy of Sciences. His research interest includes nanomaterials, nano-devices, energy-related sciences, surface science and photoelectron chemistry.

Yong Lei is Professor and the Head of Group (Chair) of Applied Nano-physics at Technical University of Ilmenau in Germany. He started to work in Germany as an Alexander von Humboldt Fellow at Karlsruhe Institute of Technology in 2003. From 2006 he worked at University of Muenster as a group leader and Junior Professor. In 2011, he joined the Technical University of Ilmenau as a Chair Professor. His research focuses on template-based nanostructuring, energy conversion and storage devices, and optoelectronic applications of functional nanostructures. He received a few prestigious funding in Europe and German including two European Research Council Grants. Prof. Lei is Advisor Board Member or Associate Editor of a few journals such as *Advanced Energy Materials*, *Energy & Environmental Materials*, *InfoMat* and *Carbon Energy*.