REVIEW

Mobile energy storage technologies for boosting carbon neutrality
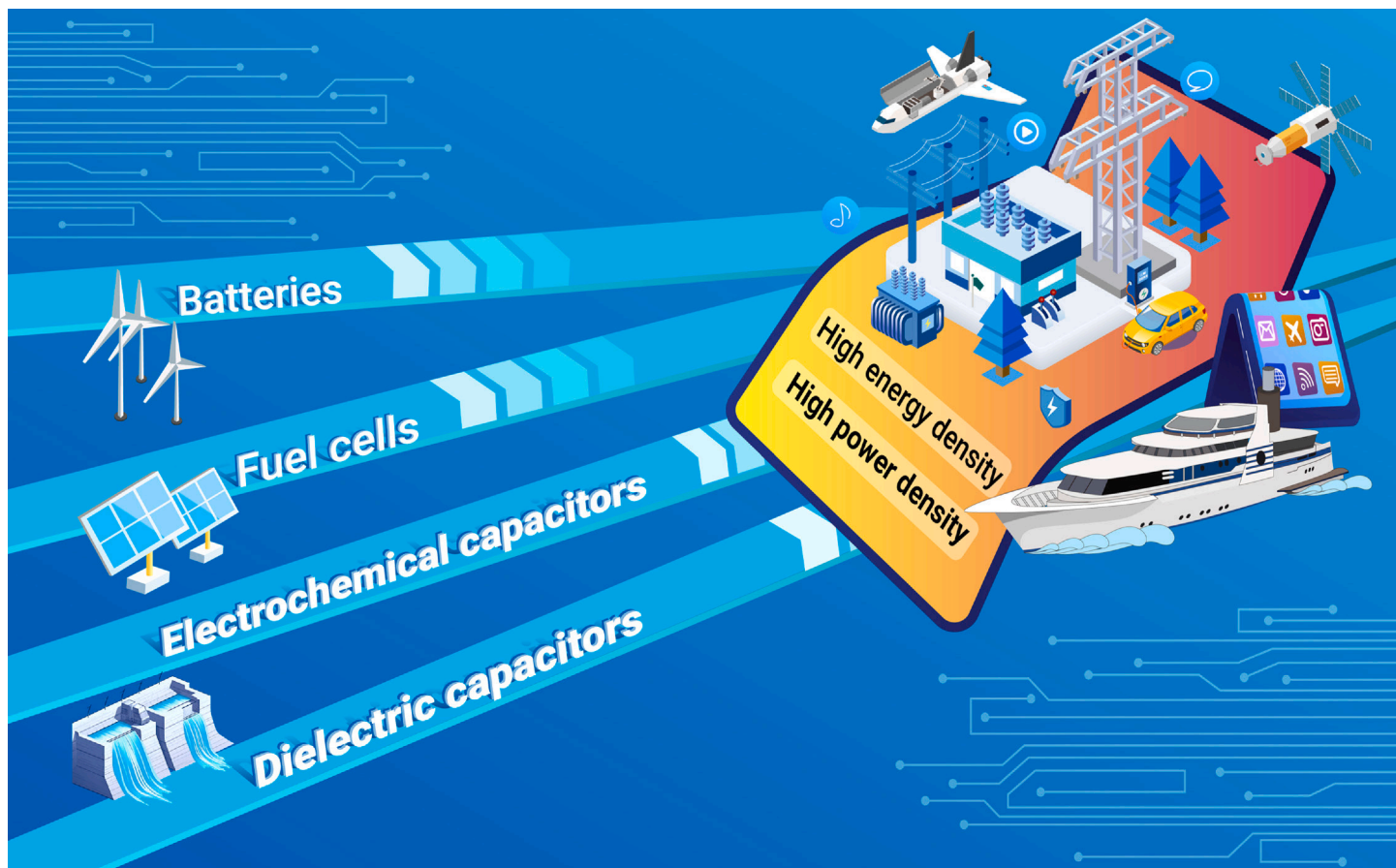
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GRAPHICAL ABSTRACT

PUBLIC SUMMARY

- Mobile energy storage technologies are summarized.

- Opportunities and challenges of mobile energy storage technologies are overviewed.

- Innovative materials, strategies, and technologies are highlighted.

- Development directions in mobile energy storage technologies are envisioned.

Mobile energy storage technologies for boosting carbon neutrality

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Carbon neutrality calls for renewable energies, and the efficient use of renewable energies requires energy storage mediums that enable the storage of excess energy and reuse after spatiotemporal reallocation. Compared with traditional energy storage technologies, mobile energy storage technologies have the merits of low cost and high energy conversion efficiency, can be flexibly located, and cover a large range from miniature to large systems and from high energy density to high power density, although most of them still face challenges or technical bottlenecks. In this review, we provide an overview of the opportunities and challenges of these emerging energy storage technologies (including rechargeable batteries, fuel cells, and electrochemical and dielectric capacitors). Innovative materials, strategies, and technologies are highlighted. Finally, the future directions are envisioned. We hope this review will advance the development of mobile energy storage technologies and boost carbon neutrality.

INTRODUCTION

Energy is one of the driving forces for the progress of human civilization. For a long period, the development of human society has depended on basic energy forms: biomass, solar, water and wind, etc. The first industrial revolution, centered on the innovation of steam engine, popularized the burning of fossil fuels (coal, oil, and gas) for energy.[1] Although the flourishing of the second industrial revolution brought mankind into the electrical era, the generation of electricity was still based mainly on the burning of fossil fuels. Remarkably, consumption of fossil fuels increased dramatically over the past century, about eight times since 1950, and has roughly doubled since 1980,[2] leading to significant economic and environmental issues, including a shortage of resources, massive emissions of greenhouse gases and pollutants, and more frequent extreme weather.[3]

In response to global climate change, the Paris Climate Change Agreement was signed, and 194 parties have joined, with the aim of limiting the average temperature rise in this century to below 2°C (or even less than 1.5°C). In 2020, the government of China also announced at the 75th session of the United Nations General Assembly that China would aim to peak its carbon dioxide emissions by 2030 and strive to achieve its carbon neutrality goal by 2060. However, the process of switching from fossil fuels to low-carbon energy faces huge challenges, and fossil fuels still are the main energy sources at present (Figure 1A). Given the exhaustion crisis of fossil fuels and their environmental hazards, it is urgent to accelerate the development of renewable energy technologies. Although renewable energy technologies such as hydropower, wind, solar, etc., have progressed rapidly, they are hindered by intermittent production due to the alternation of day and night, seasonal variations, and geographical discrepancies, resulting in discontinuous, unstable, and uncontrolled generation of electricity (Figure 1B), which leads to serious impacts on the safety and stability of power grids.[4] Hence, the efficient use of renewable energies requires energy storage mediums that enable the storage of excess energy and reuse after spatiotemporal reallocation.

To date, various energy storage technologies have been developed, including pumped storage hydropower, compressed air, flywheels, batteries, fuel cells, electrochemical capacitors (ECs), traditional capacitors, and so on (Figure 1C).[5] Among them, pumped storage hydropower and compressed air currently dominate global energy storage, but they have disadvantages, such as restriction of location, huge cost, low energy conversion efficiency, inability to transport, and serious damage to the environment.[7] Flywheels and superconducting magnetic energy storage have the merits of high power density but the demerits of high cost for superconducting materials, low energy density, and difficulty moving after they are established. Compared with these energy storage technologies, technologies such as electrochemical and electrical energy storage devices are movable, have the merits of low cost and high energy conversion efficiency, can be flexibly located, and cover a large range, from miniature (implantable and portable devices) to large systems (electric vehicles and even electricity grid systems)[8,9] and from high energy density to high power density (Figure 1D). For example, rechargeable batteries, with high energy conversion efficiency, high energy density, and long cycle life, have been widely used in portable electronics, electric vehicles, and even grid-connected energy storage systems. Fuel cells, especially hydrogen fuel cells, which are being explored as a clean energy solution, have the merits of higher energy densities, providing autonomous power in off-grid locations, including vehicles with long driving ranges and fast refueling. On the other hand, capacitors are known for their high power densities and rapid charge/discharge capabilities, which make them useful in scenarios that require fast energy release and high power output, such as auxiliary starting system, electric braking system for energy recovery, streetcar power systems, hybrid electric vehicles, burst-mode power delivery systems, frequency regulation, grid power buffers, and miniaturized electronics. In particular, dielectric capacitors, with even higher power density, are promising for progressive power devices and pulse power equipment, such as medical equipment and electromagnetic weapons.

In this regard, such mobile energy storage technologies should play a more important role in both industry and our daily lives, although most of them still face challenges or technical bottlenecks. Herein, we provide an overview of the opportunities and challenges surrounding these emerging energy storage technologies (including rechargeable batteries, fuel cells, ECs, and dielectric capacitors). Innovative materials, strategies, and technologies are highlighted. Finally, future directions are envisioned. We hope this review will be beneficial to the further development of such mobile energy storage technologies and boosting carbon neutrality.

RECHARGEABLE BATTERIES

Batteries are electrochemical devices, which have the merits of high energy conversion efficiency (close to 100%). Compared with the ECs, batteries possess high capacity and high energy density. Figure 2A displays a timeline of key discoveries for battery technologies since 1800. The primary battery was invented by Alessandro Volta and widely used as a portable power source.[10] Subsequently, first rechargeable lead-acid batteries were developed, became popular, and are still in use today because of their low cost.[11] Various secondary batteries were developed later, such as nickel-cadmium batteries, nickel-metal hydride batteries, lithium-ion batteries (LIBs), etc.[12] Among them, LIBs have attracted a lot of interest since their commercialization 30 years ago, and they are the current winner for applications in consumer electronics and electric vehicles because of their high energy density (Figure 2B), light weight, and long cycle life.[13,14] Although ongoing research is focused on various novel electrode materials (Figure 2C), the majority of commercial LIBs are still based on graphite anodes and lithium transition metaloxide cathodes (e.g., $LiCoO_2$, $LiNi_xCo_yMn_zO_2$ [NCM], $LiFePO_4$). At present, the state-of-the-art LIBs can reach a specific energy of ~250 Wh kg^{-1} at the cell level and offer a driving range of 300−600 km for electric vehicles.[15,16] The capacity and the driving range are already comparable with traditional oil-fueled automobiles, but they still cannot meet the growing demand for broader applications such as heavy-duty vehicles, electric flights, large vessels, and grid-scale energy storage. Besides, fast charge and discharge (i.e., the power density of LIBs) are often pursued.[17,18] Meanwhile, cyclability, sustainability, safety, and low cost are also important aspects.[19] All-around development of LIBs is sluggish and requires providential permutations and combinations.[20]
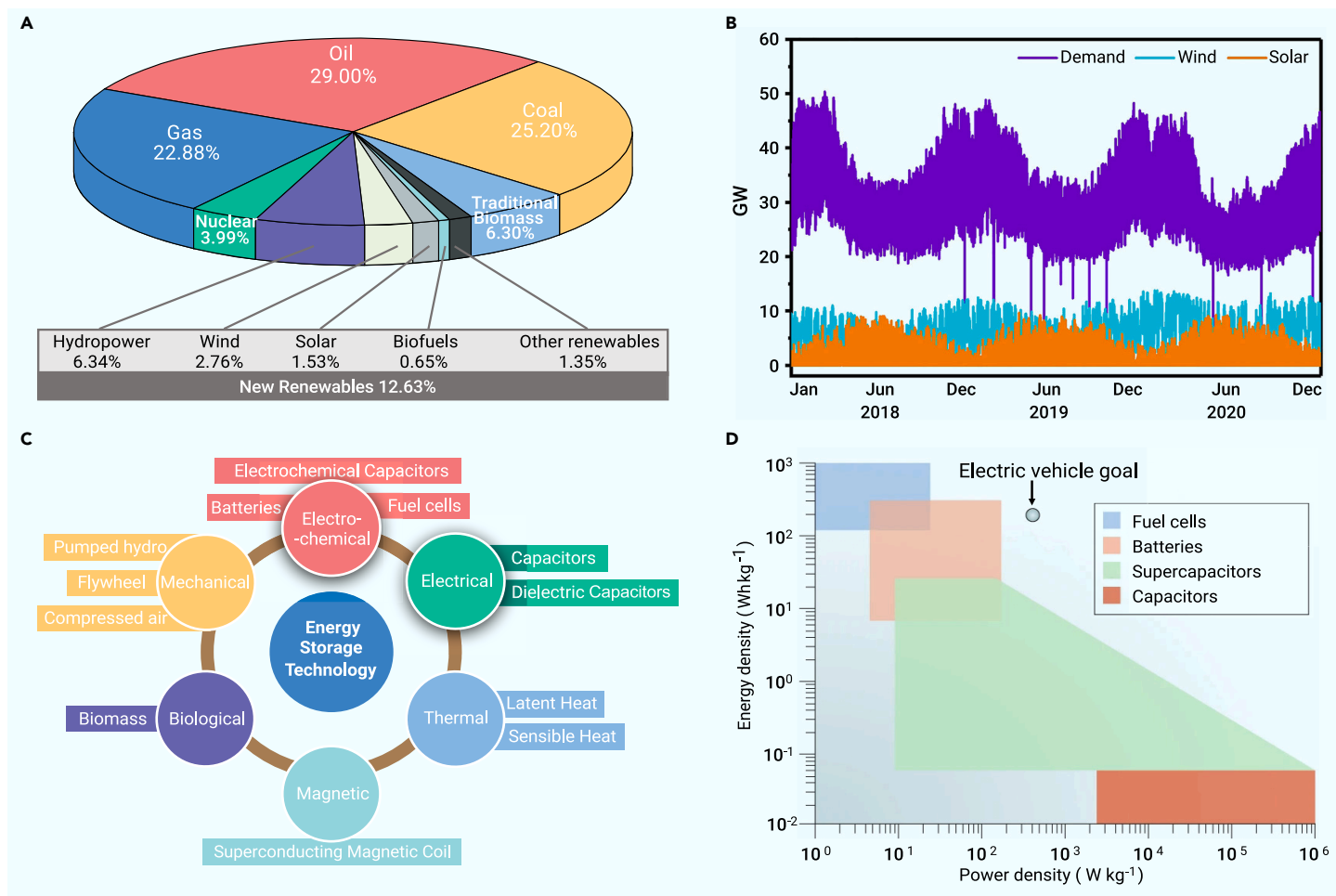
Figure 1. Demand and types of mobile energy storage technologies (A) Global primary energy consumption including traditional biomass, coal, oil, gas, nuclear, hydropower, wind, solar, biofuels, and other renewables in 2021 (data from Our World in Data[2]). (B) Monthly duration of average wind and solar energy in the U.K. from 2018 to 2020. Reprinted with permission from Zhu et al.[4] Copyright 2022, American Chemical Society. (C) Various energy storage technologies. Reproduced with permission from Rahman et al.[5] Copyright 2022, Wiley-VCH. (D) Mobile energy storage technologies. Reprinted with permission from Sun et al.[6] Copyright 2019, Springer Nature.

In the short term, the development of LIBs is highly dependent on current mature commercial systems and can be further improved by optimizing cell-level parameters, for example, decreasing electrode porosity, increasing the content of active materials, reducing inactive materials, increasing the specific capacity (e.g., adopting high-nickel-content NCM or lithium-rich manganese-based cathode materials), enhancing output voltage (choosing cathode materials with high discharge voltages and improving the stability of electrolytes), optimizing the ratio of negative to positive electrodes, and so on (Figure 2D).[21] In addition, the cells in a battery pack system are often connected in parallel to increase capacity and in series to increase output voltage. Thus, the packaging and structural innovation of a battery pack system are also significant to simplify the battery, module, and packaging mode but also enhance energy and power density (Figure 2E).[22] Moreover, cell geometries should be delicately selected to meet space constraints and appearance considerations and balance energy density and other requirements, such as cooling systems for inhibiting potential thermal runaway. Such strategies have been well demonstrated by the innovative "blade battery" from BYD,[23] Cell to Pack (CTP) and Cell to Chassis (CTC) technologies from CATL, and the 4680 structural battery from Tesla.

Another possibility for short-term solutions is to recycle retired batteries to solve the crisis of lithium, cobalt, and nickel resource scarcity, though this would not help enhance performance.[26] Recycling not only provides valuable secondary sources of materials but also prevents environmental pollution caused by rampant disposal of spent LIBs.[27] At present, the main recycling technologies can be divided into four categories: direct recycling by physically disassembling batteries and separating their components, pyrometallurgy, hydrometallurgy, and a combination of pyrometallurgy and hydrometallurgy.[28,29] In addition, some novel ideas also have been proposed, such as using deep eutectic solvents to extract valuable metals from cathodes, adopting a selective anion exchange

method by selectively sulfidizing a target metal from mixed metal oxide cathodes.[30,31] With the continuous efforts on optimizing the recycling process, it can be expected that recycling LIBs could largely alleviate the crisis of resource scarcity in the next few years.[32,33]

In the medium and long terms, replacing traditional commercial electrode materials with novel electrode materials is essential to solve the crisis of exhaustion and the rapid price increases faced by cobalt and nickel[34,35] and is the most efficient choice for pursuing higher performance, as the specific capacity of transition-metal oxide cathodes and graphite anodes is already approaching its theoretical maximum.

On the cathode side, sulfur has become one of the most promising candidates because of its abundance, low cost, and high theoretical capacity (1675 mAh g^{-1}) (Figure 3A). Nevertheless, key challenges (the insulating nature of sulfur and Li_2S, the high solubility of intermediate polysulfides in the electrolyte, sluggish redox kinetics, and volume expansion from sulfur to Li_2S) largely limit the commercialization of sulfur cathodes.[36] Constructing interconnected conductive networks or using conductive porous host materials in sulfur cathodes proved to be able to improve the conductivity of the electrodes, reduce the solubility of the polysulfides, and enhance cyclability (Figure 3B).[37–39] Meanwhile, introducing a functional separator that could block the shuttle of polysulfides, forming insoluble active redox mediators, and implementing all-solid-state battery architectures are also effective strategies to alleviate the dissolution of polysulfides.[40–42] Upon pairing with a lithium anode, Ah-level Li-S pouch cells could be realized with energy densities of 300−600 Wh kg^{-1}, far beyond the current LIBs,[16] and large Li-S battery pack with a high energy density of 330 Wh kg^{-1} were also announced by Dalian Institute of Chemical Physics of the Chinese Academy of Sciences.[43] All these advances will push sulfur cathodes forward to practical applications.[44] Similarly, oxygen (air) cathodes have also attracted much attention because of
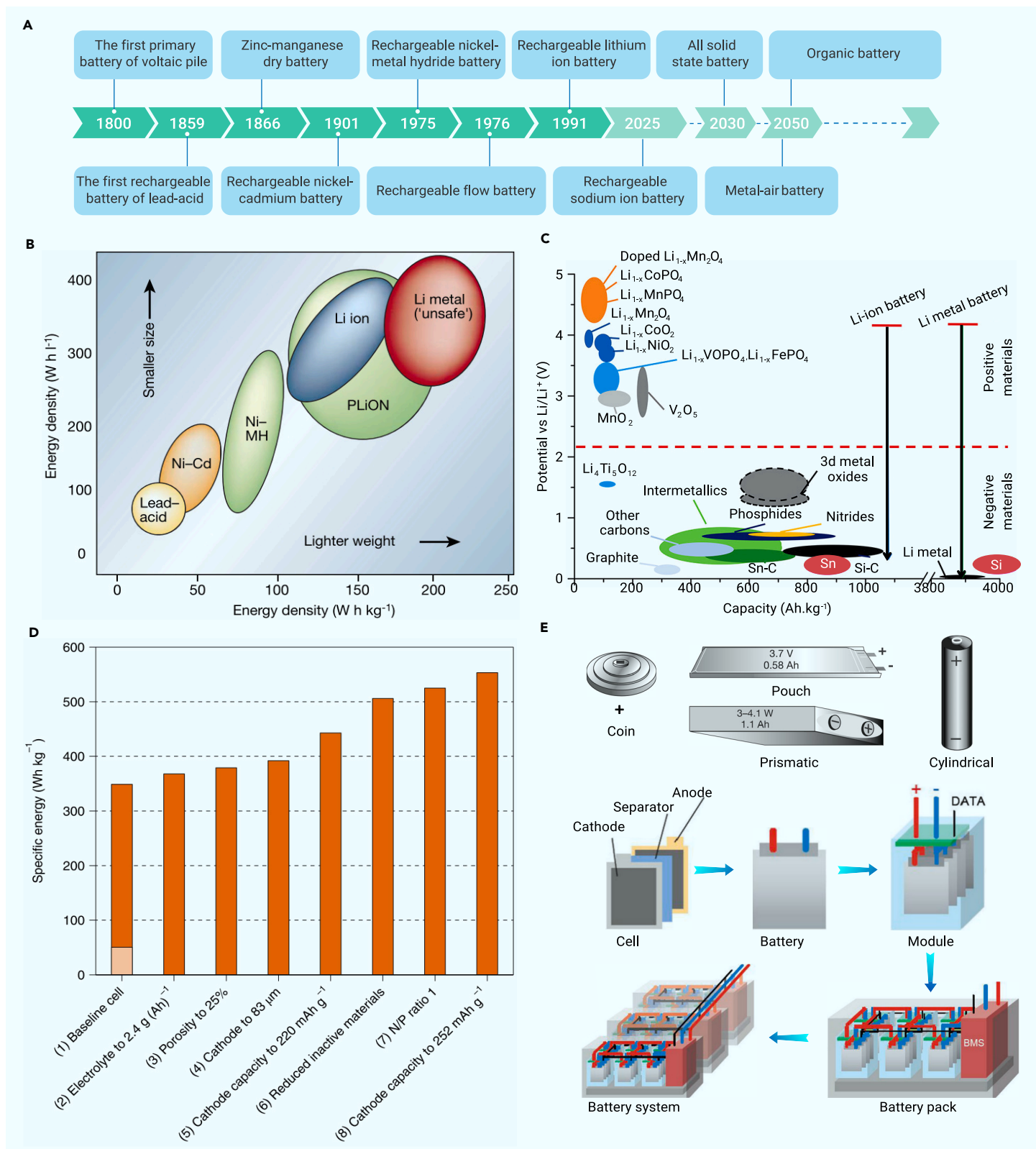
Figure 2. Key developments of battery technologies and possible directions in a short term (A) The timeline of key developments of battery technologies. (B) Comparison of the various battery technologies. Reprinted with permission from Tarascon and Armand.[14] Copyright 2001, Springer Nature. (C) Voltage vs. capacity of some representative electrode materials for LIBs. Reprinted with permission from Lemoine et al.[24] Copyright 2022, American Chemical Society. (D) Calculated cell-level specific energy as a function of cell parameters. Reprinted with permission from Liu et al.[21] Copyright 2019, Springer Nature. (E) Commonly used cell geometries (coin cell, pouch cell, prismatic cell, and cylindrical cell) and schematic diagram of the construction process from cell to battery system. Reproduced with permission from Tarascon and Armand.[14] Copyright 2001, Springer Nature. Reproduced with permission from Ghiji et al.[25] Copyright 2020, MDPI.

their high theoretical capacity (1,675 mAh g^{-1} for two-electron transfer from O_2 to Li_2O_2[45,46] and 3,350 mAh g^{-1} for four-electron transfer from O_2 to Li_2O[47,48]) and rich resources. In most reports, the O_2 reduction involves only two-electron transfer rather than a four-electron reaction (Figure 3C).[49,50] Impressively, by us-

ing a molten nitrate electrolyte and solid ceramic membrane to elevate the operated temperature, Xia et al.[47] reported a Li-O_2 battery that could reversibly form Li_2O in a stepwise four-electron redox process, which dramatically improved the energy density (Figure 3D). It was believed that according to the Gibbs-Helmholtz
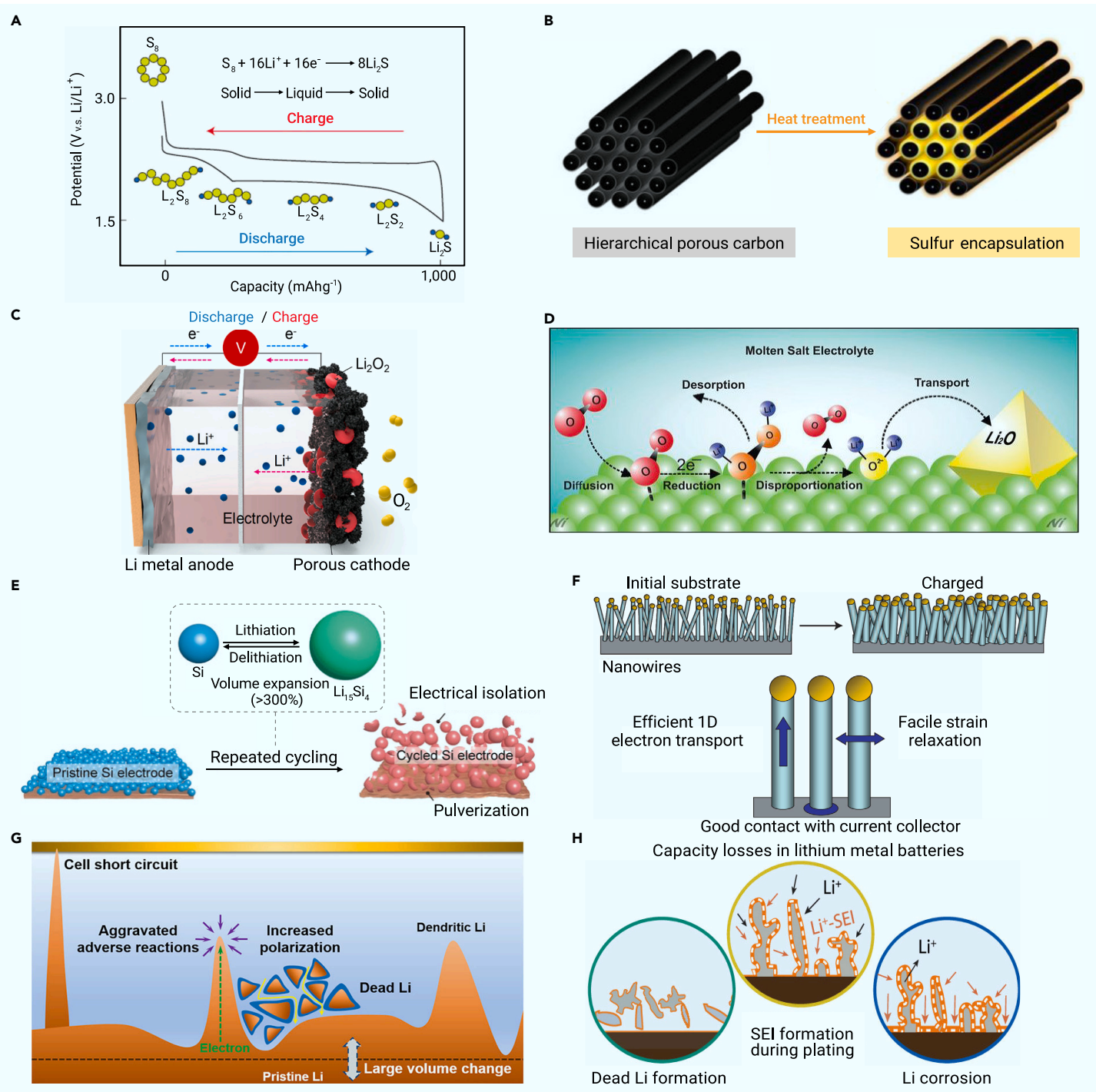
Figure 3. Possible choices for a medium and long term development of batteries (A) Voltage profiles of a Li-S battery. Reproduced with permission from Bruce et al.[50] Copyright 2012, Springer Nature. (B) A schematic diagram of the sulfur (yellow) confined in the interconnected pore structure of mesoporous carbon. Reproduced with permission from Ji et al.[37] Copyright 2009, Springer Nature. (C) Schematic presentation of typical Li-O$_2$ battery with products of Li$_2$O$_2$. Reprinted with permission from Kwak et al.[49] Copyright 2020, American Chemical Society. (D) Schematic illustration of the pathway of the oxygen reduction reaction from Li to Li$_2$O. Reprinted with permission from Xia et al.[47] Copyright 2018, American Association for the Advancement of Science (AAAS). (E) Schematic illustration of the disadvantages of Si anodes. Reproduced with permission from Chae et al.[74] Copyright 2020, Wiley-VCH. (F) The design of Si nanowire anode to solve the problems of Si anodes. Reproduced with permission from Chan et al.[54] Copyright 2008, Springer Nature. (G) Scheme of a dilemma for Li metal anode in rechargeable batteries. Reprinted with permission from Cheng et al.[75] Copyright 2017, American Chemical Society. (H) Analysis of capacity losses (SEI Li$^+$, unreacted metallic Li^0, Li corrosion) in lithium metal batteries by a series of analytical methods. Reprinted with permission from Gunnarsdóttir et al.[76] Copyright 2020, American Chemical Society.

equation, at high temperatures, Li$_2$O was more thermodynamically stable. More recently, Kondori et al.[48] reported a composite polymer electrolyte based on Li$_{10}$GeP$_2$S$_{12}$ nanoparticles and modified polyethylene oxide matrix to achieve a reversible four-electron reaction at room temperature. They proposed that a local air/O$_2$-deficient environment was important, which together with the sufficient supply of Li cations and electrons led to the further reduction of Li$_2$O$_2$ to Li$_2$O. The obtained Li-O$_2$ cells showed excellent cycling stability (1,000 cycles) with a

low polarization gap and are expected to provide a high specific energy of >1,000 Wh kg^{-1}, far beyond current LIBs. Although still in their infancy, these innovative technologies will probably promote a revolution in batteries.[12]

On the anode side, silicon, with abundant resources and an ultrahigh theoretical capacity of 4,200 mAh g^{-1} that is far beyond the 372 mAh g^{-1} of traditional graphite, is regarded as a promising choice for LIBs.[51] But the huge volume variation of Si ($\approx$400%) upon Li$^+$ insertion/extraction causes severe pulverization
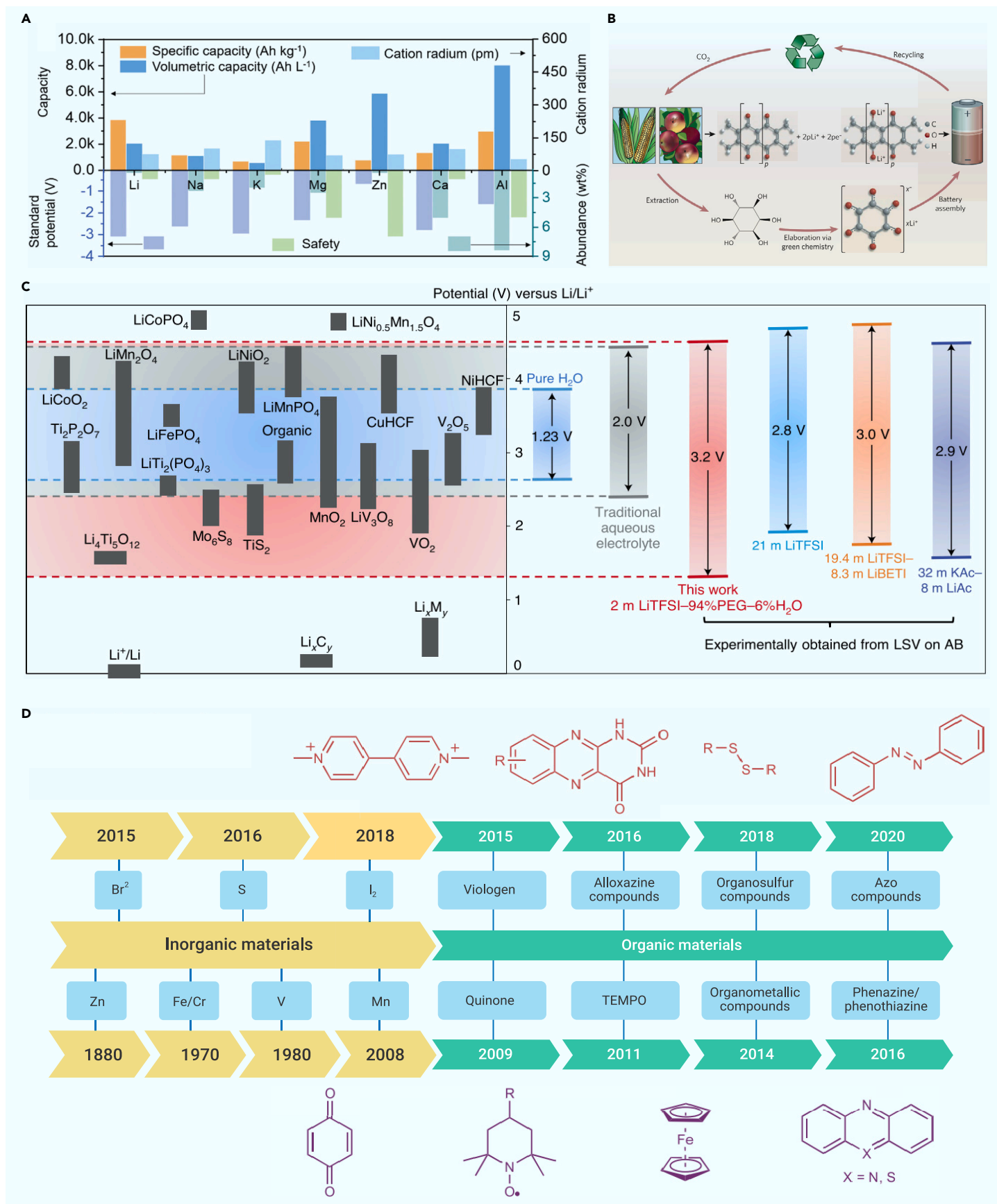
Figure 4. Future batteries for large-scale applications (A) Capacity, cation radium, standard potential, abundance, and safety of various metal anodes. Reprinted with permission from Chen et al.[86] Copyright 2022, Wiley-VCH. (B) The sustainable organic batteries based on organic electrode materials from biomass. Reprinted with permission from Tarascon and Armand.[14] Copyright 2008, Springer Nature. (C) Comparison of the measured electrolyte stability window of various aqueous electrolytes and potential electrode materials for aqueous batteries. Reprinted with permission from Xie et al.[122] Copyright 2020, Springer Nature. (D) Timeline of important inorganic and organic redox-active materials in redox-flow batteries. Reprinted with permission from Zhang et al.[121] Copyright 2022, Springer Nature.
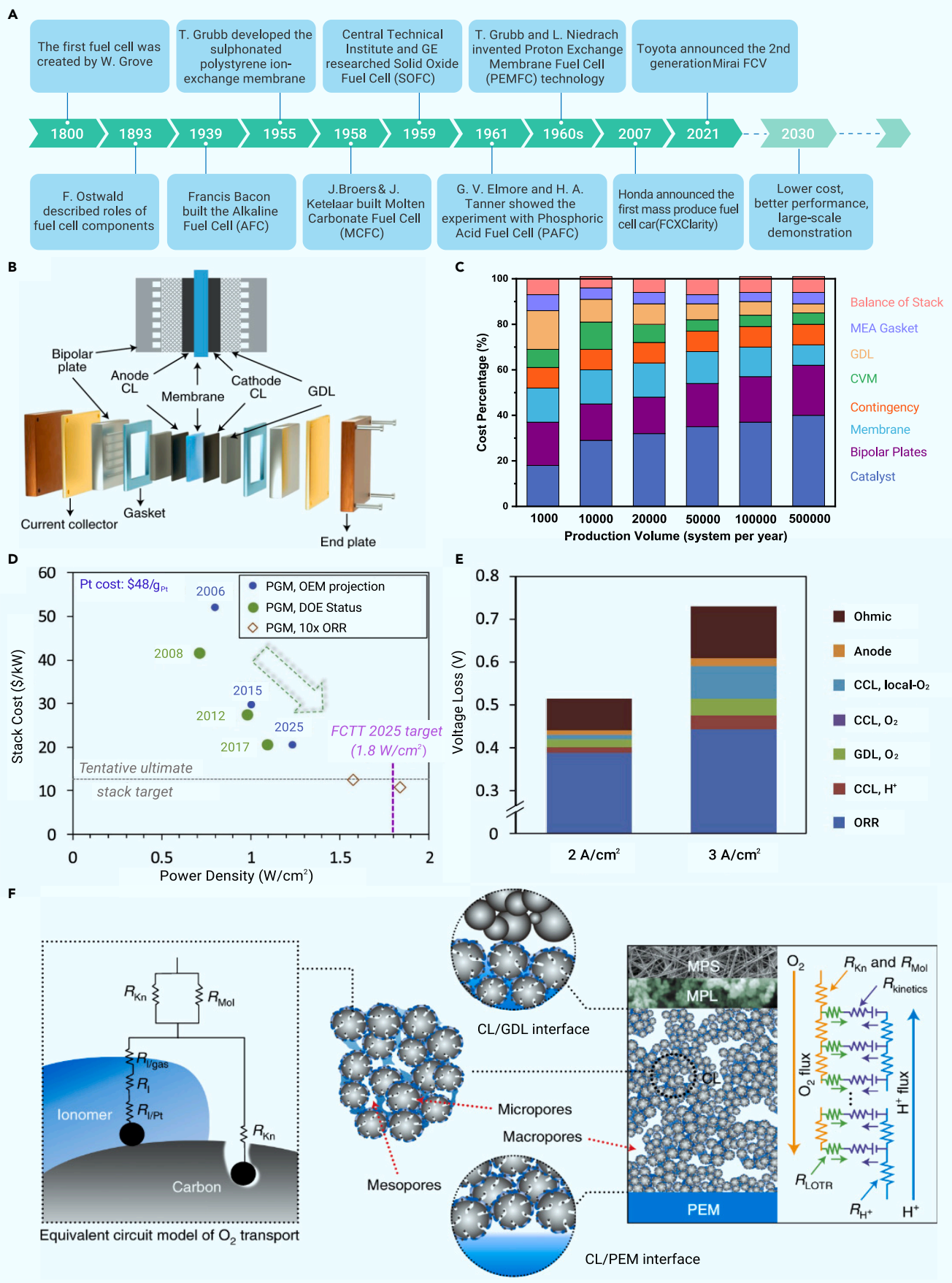
Figure 5. Key developments, targets, and technical difficulties of fuel cell technologies (A) The key developments of fuel cell technologies. (B) Configuration of a single fuel cell. Reprinted with permission from Fan et al.[134] Copyright 2021, Springer Nature. (C) The DOE's 2022 cost breakdown of a fuel cell stack for typical light-duty-vehicle (LDV) applications.[132] (D) Progress in stack cost reduction and power density improvement assuming $48/g_{Pt}. Reprinted with permission from Gittleman et al.[127] Copyright 2019, Elsevier. (E) Voltage loss contributions at 2 and 3 A cm^{-2}. (F) Sources of mass transport resistance inside the MEA of a PEMFC. Reprinted with permission from Fan et al.[134] Copyright 2021, Springer Nature.

and structural collapse of active materials, unrestrained growth of the solid electrolyte interface, loss of contact between the active materials and the current collector, and poor cycle stability (Figure 3E).[52] A series of strategies were proposed to solve these issues, such as controlling particle size, mixing with other active materials or additives, spatial confinement, interfacial modulation, and exploiting novel binders and electrolytes.[53] Among these technologies, silicon nanowires (Figure 3F) and the hierarchical bottom-up assembly of Si and conducting carbon nanoparticles were developed by Chan et al.[54] and Magasinski et al.,[55] respectively, supporting the establishment of Amprius Technologies, Inc., and Sila Nanotechnologies, Inc., for practical applications of Si anodes.[56] After decades of development, Si/C complex electrodes are now on the verge of large-scale commercial success. Among all the candidates for anodes, Li metal is regarded as the "holy grail" to break the energy-density bottleneck of current LIBs, which has the lowest standard electrochemical redox potential (-3.04 V vs. standard hydrogen electrode [SHE]) and an the highest theoretical specific capacity ($3,860$ mAh g^{-1}).[57] Li anodes were widely adopted before the commercialization of LIBs.[15,58] However, Li-dendrite growth, low Coulombic efficiency, poor cycle life, and potential safety hazards associated with Li metal anodes severely hinder their commercialization (Figure 3G).[59] Around 2010, the pursuit of high-energy-density devices triggered reviving research on Li metal batteries.[60] Advanced techniques were developed to deepen the understanding of the nature of Li anodes, including the structure of deposited Li metal, the atomic structures of solid electrolyte interphase (SEI) layers, the formation of LiH,[61,62] and the quantification of dead metallic Li^0 and chemically formed Li^+ in the SEI (Figure 3H).[63] Innovative strategies hence have been proposed, such as using solid-state electrolyte,[64] electrolyte engineering,[65,66] using a three-dimensional (3D) host,[67] separator modification,[68] and artificial SEI engineering.[69] Impressively, benefiting from the excellent mechanical strength, inflammability, and dense structure of solid-state electrolytes, all-solid-state batteries can suppress Li dendrite penetration and solve the safety issues of lithium metal anodes.[70] However, suitable solid-state electrolyte with high overall performance is challenging, often showing low ionic conductivity, a narrow electrochemical window, instability, or low compatibility with electrode materials, among other issues. In addition, the presence of inevitable cracks in solid-state electrolytes during battery assembly or long-time cycling could not eliminate lithium dendrite penetration. Recently, a solid-state electrolyte with a multilayer structure, in which a less stable electrolyte with high ionic conductivity between more-stable solid electrolytes for improving the compatibility with electrodes, achieved outstanding capacity retention after 10,000 cycles at a 20 C rate and an ultrahigh specific energy of 631.1 Wh kg^{-1}, using $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ as cathode.[71] Noted that, for all-solid-state batteries, the anode is not limited to Li, and the cathode also has more options.[40,72,73] The elimination of liquid electrolyte and separator can further enhance energy density, and hence all-solid-state batteries have been regarded as among the medium-term alternatives to current commercial LIBs (Figure 2A).

Because of the increasing demand of mobile energy storage devices and a shortage of lithium resources,[77] the replacement of lithium with more sustainable materials has become urgent. The abundances of sodium, potassium, magnesium, calcium, and aluminum in the earth's crust are much higher than that of lithium (Figure 4A).[78,79] Among them, sodium and potassium, as the same main group elements of Li, have the approximate electrode potential (-2.71 V vs. SHE for Na/Na^+, -2.93 V vs. SHE for K/K^+) and similar electrochemical performance as Li, which makes it possible to directly use similar materials, systems, and processes of current commercial LIBs.[80] However, the large size of Na^+ (1.02 Å) and K^+ (1.38 Å) compared with Li^+ (0.76 Å) will lead to faded capacity, sluggish reaction kinetics, poor cyclability of sodium-ion batteries (SIBs) and potassium-ion batteries (PIBs).[81] Therefore, SIBs and PIBs are considered as low-cost alternatives for large-scale applications in the near future, which should be a complementary of LIBs and could compete with lead-acid batteries.[82] Impressively, the SIBs with obvious cost advantages was announced by HiNa Battery Technology Co., Ltd, using sodium-copper-iron-manganese oxide and anthracite-based soft carbon as positive and negative electrode materials, respectively, and reaching an energy density of 145 Wh kg^{-1} and a cycle life of about 4,500 times.[83] CATL also released its first-generation SIBs by using a Prussian white ($Na_2Mn[Fe(CN)_6]$) cathode and a hard carbon anode in 2021, and the product possessed an energy density of 160 Wh kg^{-1} and excellent fast charging and low-temperature capabilities.[84] Choosing suitable electrode materials and optimizing the other components in the cells would be able to reaching the energy density over 200 Wh kg^{-1}.[85] Apart from alkali metal-ion batteries, rechargeable multivalent metal ion (Zn^{2+}, Mg^{2+}, Ca^{2+}, Al^{3+}) batteries have also attracted widespread attention.[86] Compared with high reactive alkali metals, Zn and Al can be handled in air, which significantly reduces the fabrication cost and enhances safety.[87,88] More important, multivalent metal-ion batteries (MMIBs) enable multi-electron transfer, leading to high (both specific and volumetric) capacity and energy density. However, it is worth noting that the development of MMIBs is still in its infancy.[89] The main reason is that the high charge density of multivalent metal ions leads to strong polarization of the chemical environment, slow reaction kinetics, strong solvation, low capacity, and poor cyclability. The ambiguous storage mechanism needs to be further elucidated, and the slow desolvation process at the interface as well as sluggish solid-state diffusion remain two major obstacles for their transition from research to commercialization.[90]

Innovative findings in material science and deep understanding of charge storage mechanisms make these metal-ion batteries promising for energy storage. However, a common challenge faced by these batteries is the traditional inorganic electrode materials (OEMs), which have difficulty tolerating the large sizes of alkali metal ions beyond Li (Na^+ and K^+) and the high charge density of multivalent cations (Zn^{2+}, Mg^{2+}, Ca^{2+}, and Al^{3+}).[90] In this regard, organic batteries and OEMs are considered a long-term option for battery technology. Different from inorganic electrode materials, OEMs possess weak intermolecular interactions[91] and relative flexibility, which make it feasible to store metal ions with large size or multivalence with high capacity, good rate capability, and cyclability.[86] In addition, because of their structural designability, high abundance, and sustainability, possibly from natural green resources, organic batteries could be regarded as "green batteries" (Figure 4B).[12,92,93] Flexible batteries are also possible.[94] Despite the advantages, there are critical issues that need to be addressed before practical applications of OEMs, such as their poor electrical conductivity, possible dissolution in electrolytes, and low redox potential (and/or low capacity) of organic cathodes.[95] In the past decade, various innovative strategies have been reported to solve these challenges and enhance the electrochemical performance of OEMs. For example, it has been proposed that extending the conjugated system could enhance electrical conductivity and rate performance,[96] which has been widely adopted to design novel OEMs. Redox potentials can be tailored through molecular design and electron-withdrawing groups, and p-type reactions could provide high voltage output.[97] Polymerization is effective to inhibit/eliminate the dissolution of active materials,[80,98] while small molecules are of priority to reduce the cost. Wang and others have highlighted the manipulation of intermolecular interactions to inhibit/eliminate the dissolution of small molecules and enhance the charge transfer and subsequent electrochemical performance of organic batteries.[99–102] Another drawback of organic materials is their low density, which will limit the energy density of organic batteries. Organic-inorganic hybrid materials could combine the merits of both organic and inorganic materials and are useful to solve such challenge. Wang et al. promoted the application of conjugated coordination polymers (CCPs) for high-performance batteries,[103–106] which show high conductivity and have become an intriguing type of OEMs. All of these innovative materials and strategies will substantially accelerate the practical applications of organic batteries.[107]

To lower cost and solve the safety issue of batteries, particularly for large-scale applications, one attractive strategy is to use aqueous electrolytes.[108,109] The main challenges of aqueous electrolytes are the narrow electrochemical window (≈ 1.23 V) of water (giving rise to the low voltage and energy density) and the high freezing point of aqueous electrolytes (leading to salt precipitation, volume expansion of the electrolytes, and associated deformation of the batteries, low ion conductivity, and poor kinetics under low temperature).[110] To date, innovative strategies have been developed to expand the electrochemical window of water and protect electrodes (Figure 4C).[111,112] Great efforts were made to mitigate the hydrogen bonds between water molecules, thus inhibiting the formation of ordered water molecules and reducing the freezing point of aqueous electrolytes.[113–115] Strategies include using "water-in-salt" electrolytes,[110] adding organic additives as co-solvents,[116] and using hydrogels as electrolytes.[117] For large-scale energy storage, particularly at the power-grid level, aqueous redox-flow batteries are deemed one of the most suitable technologies, though the energy density cannot compete with that of the aforementioned batteries.[118] Currently, several vanadium- and zinc-based flow batteries have been demonstrated and even moved into the commercialization stage.[119,120] In consideration of further reducing the cost and enhancing energy density, organic materials

have been considered as electroactive species for flow batteries (Figure 4D).[121] With the continuous pursuit of better flow batteries, flow battery systems including hybrid redox-flow batteries, semi-solid (or slurry) redox-flow batteries, and solar redox-flow batteries have also emerged in recent years.[121] On the whole, redox-flow batteries also have a bright tomorrow for future applications, although it should be noted that for mobile applications, flow batteries may be only suitable for low-energy-density scenarios such as low-speed electric vehicles and household energy storage cabinets.

In summary, with the worldwide popularity of electric vehicles and the growing demand for broader applications of mobile energy storage devices, the development of rechargeable batteries has reached a new level. Batteries can be designed in a large range of sizes, from miniature to large systems, and hence are attractive for various application scenarios. With the blooming of innovative materials, strategies, and technologies, great improvements have been made in both academic and industrial fields in terms of energy and power density, safety, long cycle life, cost reduction, pollution control, and so on. Further efforts in transforming advanced batteries from the lab to industry are still needed. We believe that further breakthroughs in key materials and technologies will promote the market share of rechargeable batteries, thus contributing to boosting the goal of carbon neutrality.

FUEL CELLS

Similar to batteries, fuel cells can convert chemical energy of fuel (H_2, methanol, etc.) and oxidant (O_2) to electric energy through electrochemical reactions.[123] Yet unlike batteries, they do not deplete or need recharging and can provide continuous output current during their service period. Since William Grove first invented the fuel cell in 1839, different fuel cells have been developed with various forms of fuels and working conditions, such as proton exchange membrane fuel cells (PEMFCs), alkaline fuel cells (AFCs), phosphate fuel cells (PAFCs), molten carbonate fuel cells (MCFCs), and solid oxide fuel cell (SOFCs) (Figure 5A).[124] Among these devices, PEMFCs with hydrogen and oxygen as the fuels represent one of the most promising and attractive technologies for a wide variety of mobile and stationary power applications because of their high energy density, low working temperature, high energy conversion efficiency, and low pollution.[125,126] For example, the 2nd-generation Mirai fuel cell vehicle released by Toyota in 2021 can provide a maximum stack output power of 128 kW with a volumetric power density of 4.4 kW L^{-1} and can reach a typical cruising range of 850 km. Nonetheless, there are still many problems that need to be solved during the design of practical fuel cell systems (FCSs).[127,128]

The continuous optimization of FCS metrics is a complex project combining material science and technical engineering.[129] More specifically, fuel cell performance-related metrics include mass activity (MA) of the catalyst, current density at a specific voltage (e.g., 0.7 V), rated power density, energy conversion efficiency, etc. Durability metrics include MA/voltage loss after a certain period of life test or potential cycling.[130,131] Among these indicators, cost issue is the main bottleneck for the large-scale applications. Typically, a single fuel cell consists of a membrane electrode assembly (MEA; containing a catalyst, a membrane, and a gas diffusion layer [GDL]), a gasket, a bipolar plate (BP), a current collector, and an end plate (Figure 5B). According to a U.S. Department of Energy (DOE) 2022 cost breakdown of a fuel cell stack for typical light-duty-vehicle (LDV) applications (Figure 5C), the cost of the MEA can reach more than 50% of the total system,[132] which is attributed mainly to the use of noble metal Pt-based catalysts (more than U.S. $1,000 per ounce) and the proton exchange membrane (PEM) (Nafion; more than U.S. $200 per m^2). In particular, with the increase of annual production volume, the cost of catalyst would become a larger fraction of the stack cost (40% at 0.5 M systems per year).[127] According to the cost target of the DOE ($30 kW$_{net}^{-1}$), the cost for the stack alone should be less than $12.6 kW$_{net}^{-1}$, with a power density target of 1.8 W cm^{-2}, which poses a great challenge for the improvement of the fuel cell performance with a lower cost (Figure 5D).[133]

The optimization of the MEA is considered first, as the complicated electrochemical reactions (anode: hydrogen oxidation reaction [HOR]; cathode: oxygen reduction reaction [ORR]) and mass transfer process (O_2, protons, electrons, and water) within the catalyst layer (CL) are the key factors determining the performance of a practical cell.[134] ORR kinetics, ohmic resistance, and mass transfer resistance mainly dominate the voltage loss at 2 and 3 A cm^{-2} (Figure 5E).[135] Among them, the sluggish ORR kinetics account for most of the voltage loss, because of the strong adsorption of oxygenated intermediates (e.g., *O and

*OH) on pure Pt catalysts. Hammer and Nørskov[136] developed the d-band center theory to guide catalyst design, which suggests that a proper downward shift of the d-band center could achieve a weakened adsorption of oxygen intermediates. On this basis, some catalysts far exceeding the activity of pure Pt catalysts have been successively developed by reducing particle size,[137] alloying with transition metals,[138] designing unique morphology,[139] and constructing composite structures[140] (Figure 6A). Among them, PtM alloys (where M = Fe, Co, Ni, etc.) are the most investigated and commercially promising catalysts. However, Pt-based alloy catalysts suffer from severe degradation in PEMFCs because of the fast dissolution of transition metals under strong acid (pH < 1) and high potential (>0.8 V). In this regard, several approaches have been developed to slow the degradation of Pt alloy catalysts, such as realizing disorder-order phase transformation,[141] construction of protection layer,[142] etc. Among them, structurally ordered Pt-based intermetallic alloys have been considered as promising cathode catalysts with balanced ORR activity and durability. The strengthened interatomic bonding in the ordered intermetallic alloy helps stabilize the metal component, and the optimized surface strain toward Pt-skin accounts for the enhanced ORR activity (Figure 6B).[143,144] Yang et al.[145] developed an S-doped carbon to stably synthesize small Pt-based intermetallic alloy nanoparticles (PtCo, PtNi, etc.), which exhibited excellent activity in the practical fuel cell. Because of the huge rarity of Pt, non-platinum-group metal (PGM) catalysts, especially M-N_x/C (metal-nitrogen-carbon) catalysts, have been a research hotspot in the past decade. By tuning atomically dispersed transition metal centers (e.g., Fe,[146,147] Co[148,149]) and their coordination structures (typically with 4 pyridinic or pyrrolic nitrogen atoms and other non-metal atoms, e.g., B, O, S, P),[150,151] the intrinsic ORR activity and stability of M-N_x/C catalysts can be efficiently promoted.[152–155] Liu et al.[156] reported a highly durable and active Fe-N_4/C catalyst using heat treatment and a subsequent chemical vapor deposition (CVD) process, which exhibited excellent activity and durability in the fuel cell test. Nonetheless, in terms of activity and especially stability, state-of-the-art non-PGM catalysts still fall short of currently practical requirements. This is due primarily to the detachment of metal atoms under electrochemical environment, structure collapse by the Fenton reaction and radical attack, carbon corrosion, etc.[154] Thus, Pt is currently the only option for the commercialization of fuel cells. The efficient recovery of Pt resources from spent catalysts appears to be a more practical and immediate method.[157]

Local mass transport (O_2, H^+) resistance is another main reason causing voltage loss, which comes primarily from the ionomer/gas interface resistance and the ionomer/Pt interface resistance (Figure 5F).[158,159] Improvement of mass transport at interface can be achieved through modulating or designing ionomer structure.[160] Through decreasing the length of perfluorosulfonic acid (PFSA) ionomers, ionomer coverage and proton conductivity can be improved.[161,162] Also, a lower value of equivalent weight (EW) can enable lower local mass transport resistance.[163,164] New ionomer structures are expected to provide new perspectives for reducing mass transfer resistance. Recently, Zhang et al.[165] showed that adding a sulfonated covalent organic framework (COF) to Nafion could effectively promote oxygen permeation and improve the activity on the basis of Pt by up to 60%. It is noteworthy that the catalyst/ionomer interface is also closely linked to the coverage and the distribution of ionomer over carbon and Pt.[162]

The structure of carbon support is another critical issue that could reduce local mass transport resistance (Figures 5E and 5F).[162,170] Through enhancing the interaction between Pt and carbon support (e.g., using an N-modified carbon support), surface Pt can be well anchored against migration and aggregation, and the exposed surface area of Pt nanoparticles can be increased because of the uniform distribution of ionomer (Figure 6C).[167,171] In addition, special attention should be paid to the grand challenge of carbon corrosion during the long-term service of fuel cells.[172,173] Many different carbon supports (e.g., graphited carbon) have been developed to retard carbon corrosion. Especially, developing non-carbon supports such as metal oxides may be a promising direction because of their excellent corrosion resistance and strong metal-support interaction.[174]

As the one of the core components of FCSs, the current challenges for PEMs lie mainly in durability and cost.[162,175] The failure of membrane is believed to be the results of a combined chemical and mechanical effects, including gas crossover, radical attack, ion pollution, humidity change, etc.[176,177] For chemical degradation, radical scavenger (e.g., CeO_2 and derivatives) has been added to the PEM or CL to reduce the effect of radical (formation of H_2O_2) and protect the membrane (Figure 6D).[168,178] As for mechanical degradation, changes in
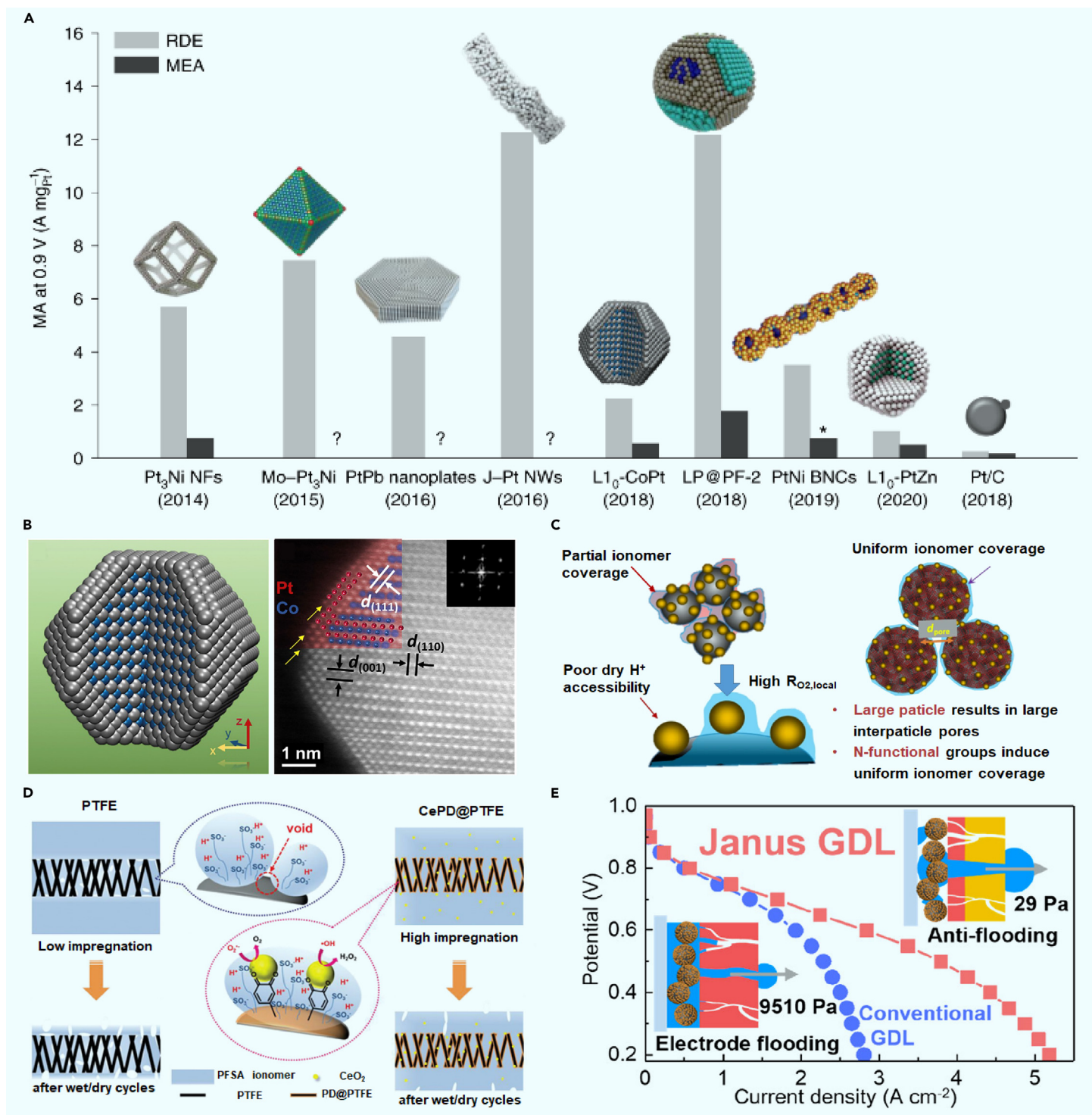
Figure 6. Possible directions of device optimization for the high-performance PEMFCs (A) Performance of state-of-the-art catalysts evaluated in RDE and MEA. Reprinted with permission from Fan et al.[134] Copyright 2021, Springer Nature. (B) Intermetallic L1₀-CoPt ORR catalyst with ordered core and Pt-skin shell. Reprinted with permission from Li et al.[166] Copyright 2019, Elsevier. (C) Schematic illustration of the advantages of carbon support with well-distributed N-functional group. Reprinted with permission from Islam et al.[167] Copyright 2022, Springer Nature. (D) Reaction mechanism of MEAs with CeO₂ as a radical scavenger. Reprinted with permission from Yoon et al.[168] Copyright 2019, Wiley-VCH. (E) Schematic diagram of the antiflooding mechanism of a Janus GDL. Reprinted with permission from Wen et al.[169] Copyright 2022, American Chemical Society.

temperature, pressure, and humidity conditions can induce the obvious mechanical stress of membrane during fuel cell operation, leading to the formation of perforation or tearing of membrane.[177] On the basis of the comprehension to these two mechanisms, many efforts have been made, such as developing nano-composite membranes to enhance the membrane strength and reduce cost.[179]

Other PEMFC components, such as the GDL and BP, are critical to gas transport and water management, which determine high current density performance. Currently, much interest has been focused on tuning the microstructure

parameters of carbon-based GDLs. A Janus GDL with spontaneously directional water transport capability is prepared by building a proper wettability gradient and large-sized pore distribution in the GDL and achieves a high peak power density in a H₂-air fuel cell (Figure 6E).[169] On the other hand, the BP accounts for more than 60% of the weight and about 20% of the total cost of the PEMFC stack, and the flow-field configuration in the BP also affects the water management.[180] So far, many different flow channel configurations and layouts have been deduced to achieve favorable air-flow distributions and water concentration profiles through optimized simulation and other means.[181]
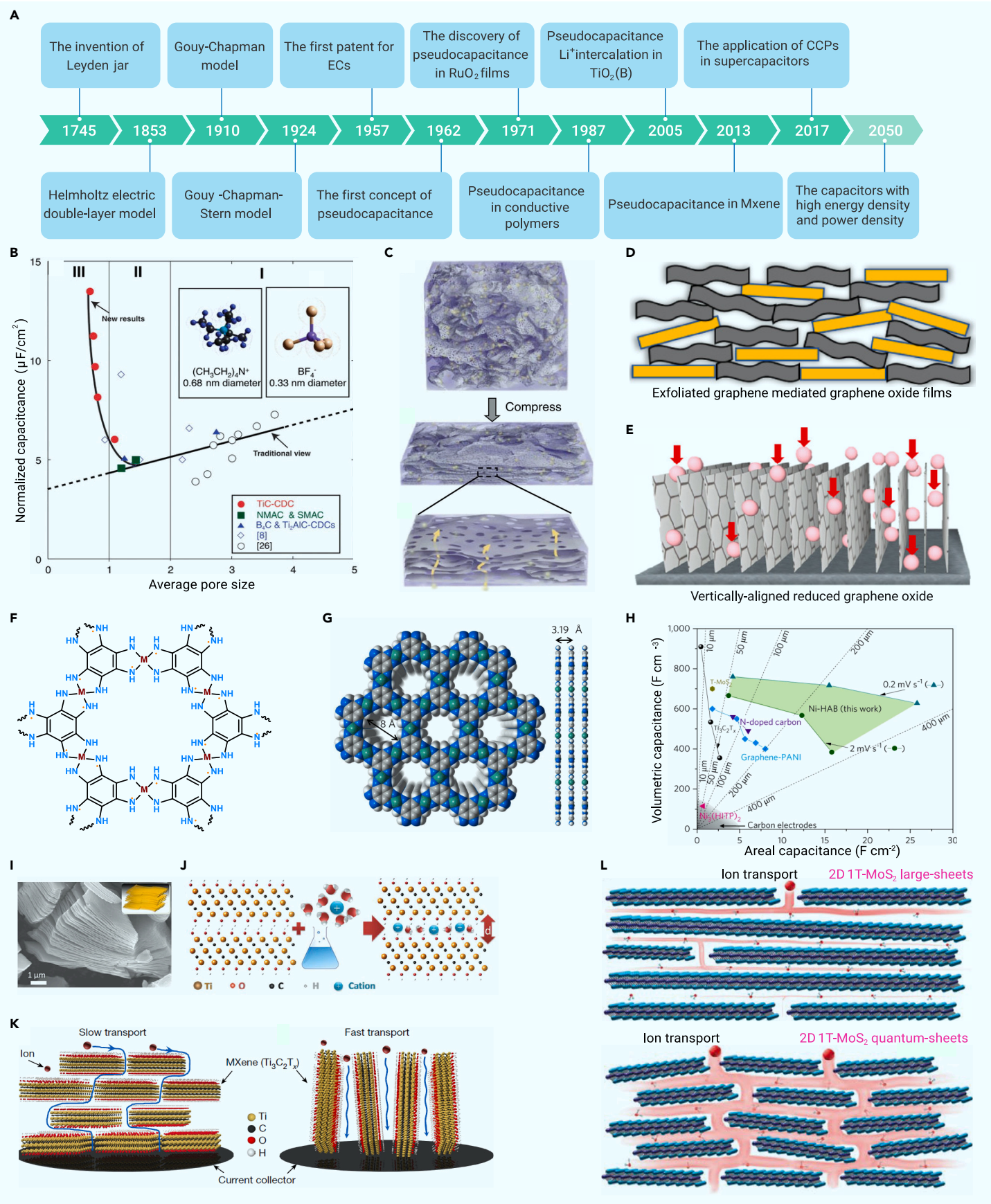
Figure 7. Key developments of electrochemical capacitors and current research hotspot (A) Historic timeline for the development of ECs. (B) Plot of specific capacitance normalized by average pore size for various carbon electrodes. Reprinted with permission from Chmiola et al.[195] Copyright 2006, AAAS. (C) Schematic diagram of compressed films of the holey graphene framework. Reprinted with permission from Xu et al.[203] Copyright 2014, Springer Nature. (D) Schematic illustration of the exfoliated graphene mediated graphene oxide films. Reprinted with permission from Li et al.[205] Copyright 2020, Springer Nature. (E) Schematic illustration of the diffusion of electrolyte ions into the vertically aligned reduced

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In a predictable prospect, the development of hydrogen energy is crucial to solve the serious environmental pollution and achieve the goal of carbon neutrality, with the commercialization of high-performance PEM fuel cells as a necessary prerequisite. In the face of possible wide application scenarios, durability has become an urgent problem, making innovations in materials and systems at all levels are necessary to improve the overall performance of fuel cell as much as possible.

ELECTROCHEMICAL CAPACITORS

Although batteries and fuel cells have the advantages of high energy density, they suffer from sluggish kinetics and irreversible variation of electrode materials, leading to low power density and limited lifetime.[182,183] In sharp contrast, ECs, which can store charges through electrochemical adsorption/desorption of ions at the electrode/electrolyte interface or surface faradic redox reactions,[182,184] show much faster reaction kinetics, enabling high power density (being operated in seconds or minutes) and ultralong cyclic stability (Figure 1D).[185,186] The prototype EC, the Leyden jar, was invented in 1745, which predates the invention of the battery (Figure 7A). Different from storage in bulk in batteries, surface storage in ECs leads to much lower energy density, although state-of-the-art energy density is already several orders of magnitude higher than that of traditional dielectric capacitors.[187] Therefore, ECs could meet demands in rapid-response or space-limited applications, such as auxiliary starting systems, electric braking systems for energy recovery, streetcar power systems, hybrid electric vehicles, burst-mode power delivery systems, frequency regulation, grid power buffers, and miniaturized electronics.[9,188,189] Understandably, the main route for ECs is to enhance energy density while maintaining their power density and cycling life.

Among the two types of ECs, electrochemical double layer capacitors (EDLCs) have been commercialized, and the charge storage mechanism has been well understood. As early as in 1853, Helmholtz reported the electric double layer model (Figure 7A), which was further developed and refined by Gouy, Chapman, and Stern et al.[184,190] According to this model, the double layer capacitance is proportional to the specific surface area and reverse proportional to the effective thickness of the double layer. This led to the first EDLC with porous carbon electrodes, invented by General Electric,[191] and a surge in research on various porous carbon materials with high specific surface areas.[192–194] However, later it was found that the capacitance is not always linearly increased with specific surface area.[195] The anomalous increase in capacitance in sub-nanopores (<1 nm) of porous carbide-derived carbons (CDCs; Figure 7B) revealed that not only the specific surface area but also the carbon nanostructure, pore size, and ionic size played significant roles in the capacitance.[195–197] It also demonstrated that the partial desolvation of ions resulted in accessibility of ions to sub-nanopores and thus increased charge storage, which differed from the Helmholtz model on the understanding of charge storage.[195,197–201] It can be understood that at high current densities, the mesopores (2 nm < pore size < 50 nm) and macropores (>50 nm) become the main locations for charge storage because of a serious jam of the micropores (<2 nm).[202] Therefore, ordered hierarchical microporous/mesoporous carbon materials (Figure 7C) are promising for achieving high specific capacitance and rate performance.[199,203,204] A series of strategies, such as using exfoliated graphene mediated graphene oxide films (Figure 7D),[205] adopting vertically oriented electrodes (Figure 7E),[206,207] and compressing carbon electrodes by capillarity,[193,202] were used to shorten the paths of ion transport and adjust the interlayer gap distance while ensuring high packing density.[208] In addition, the introduction of surface functional groups and heteroatom doping are also used to ameliorate the electrochemical performance of carbon materials (Figures 7D and 7E).[209] Benefiting from these optimized carbon-based electrodes, these as-fabricated supercapacitors deliver high energy density, which is far higher than that delivered by commercial supercapacitors (5–8 Wh L^{-1}) and nearly approaches the value for typical lead-acid batteries (50–90 Wh L^{-1}).[192,203,205]

Pseudocapacitors, another kind of ECs, involve Faradaic redox reactions at or near the surface of electrodes and hence show high capacitance and energy den-

sity compared with EDLCs.[215] Similar to the bridge between the surface-controlled EDLCs and the bulk Faradaic reactions in batteries,[216] pseudocapacitors blur the boundaries between ECs and batteries. The pioneer scientists[217] disclosed three faradaic mechanisms—underpotential deposition (adsorption pseudocapacitance), surface redox pseudocapacitance, and intercalation pseudocapacitance—and concluded that the latter two were the main mechanisms (Figure 7A). The materials for pseudocapacitors include transition metal oxides, hydroxides, and conducting polymers.[188] The metal oxides have high conductivity and stability, and economic and environmentally friendly metal oxides (e.g., MnO_2 instead of expensive RuO_2) are a current research hotspot (Figure 7A).[218,219] Nanocrystallization could also produce pseudocapacitive behaviors.[216,217] Conducting polymers offer pseudocapacitance, through redox reactions that involve either oxidation with concurrent storage of anions (p-doping) or reduction with cation storage (n-doping).[220] The capacitance of conductive polymers is governed mainly by their reversibly accessible doping level (typically 0.3–0.5 per monomer unit).[221] The challenge is that the conducting polymers are highly conductive only under high doping, and bulk conducting polymers always suffer from swelling and shrinking during the storage of ions, resulting in poor cycling stability. Hence, nanostructured polymers[222] or composite electrodes (combined with carbon materials or metal oxides/hydroxides) are often chosen.

Besides the traditional materials, the recent progress in material sciences brings various candidates for pseudocapacitors, including COFs,[223,224] CCPs,[225] transition-metal carbides, and nitrides (MXenes),[212,226] transition-metal dichalcogenides[227] and artificial mixed conductors,[228] etc. Among them, CCPs with π-d conjugation can effectively facilitate the delocalization of charges (Figure 7F) and form continuous electron transfer pathways.[103] Thereinto, two-dimensional (2D) CCPs also possess high porosity, large specific surface area, and plentiful redox-active sites, which endow 2D CCPs with good rate capability, large capacitance, and high power and energy densities.[229] For example, the hexa-aminobenzene (HAB)-based 2D CCPs (Cu-HAB and Ni-HAB; Figures 7F and 7G)[211] exhibit clear reversible redox behaviors, along with large gravimetric capacitances of 215 F g^{-1} for Cu-HAB and 420 F g^{-1} for Ni-HAB, respectively, outperforming many previously reported materials, including porous carbons (Figure 7H). The discovery of MXenes in 2011 also opened up an exciting field because of their high electrical conductivity, superior hydrophilicity, rich surface chemistry, and 2D layered structure (Figure 7I).[212,230] A variety of cations, including Li$^+$, Na$^+$, K$^+$, NH$_4^+$, Mg^{2+}, and Al^{3+}, can be readily intercalated into the interlayer (Figure 7J).[212,230] The unique properties have positioned MXenes as promising materials for supercapacitors.[212] The engineering of electrode architectures with such innovative materials could further enhance the performance. For example, vertical aligned MXene could avoid the restacking and enhance the ions transport,[213] contributing to high energy and power densities (Figure 7K). On the other hand, quantum sheets are capable of achieving rapid ion transport (Figure 7L), thus improving the gravimetric capacitance.[214] Such 2D structure also endows them as miniaturized electrodes for on-chip electronics.[9]

The high energy density of batteries and the high power density of supercapacitors stimulated hybrid supercapacitors by combining a battery-type electrode with a capacitive electrode in the same cell.[231] Within the hybrid systems, the cells showed improved energy and power densities.[232] Hybrid supercapacitors based on an AC//graphite system have been commercialized by JM Energy, and a device called Ultimo displayed volumetric energy density of 20 Wh L^{-1}, twice as high as that of present commercial EDLCs.[232] Additionally, a system of AC//Ni(OH)$_2$ hybrid ECs has been successfully applied in trolley buses by the Aowei Technology Co., Ltd. (Shanghai, China).[188] However, it should be noted that a trade-off exists between energy density and power density in hybrid cells. In most cases, battery-type electrodes lead to an increase in energy density at the cost of cyclability and powder density. This is undoubtedly the main drawback of hybrid devices, likely forming a mediocre electrochemical cell.

Besides material innovation for developing novel electrode materials with high capacitance, another efficient method to enhance the energy density is to

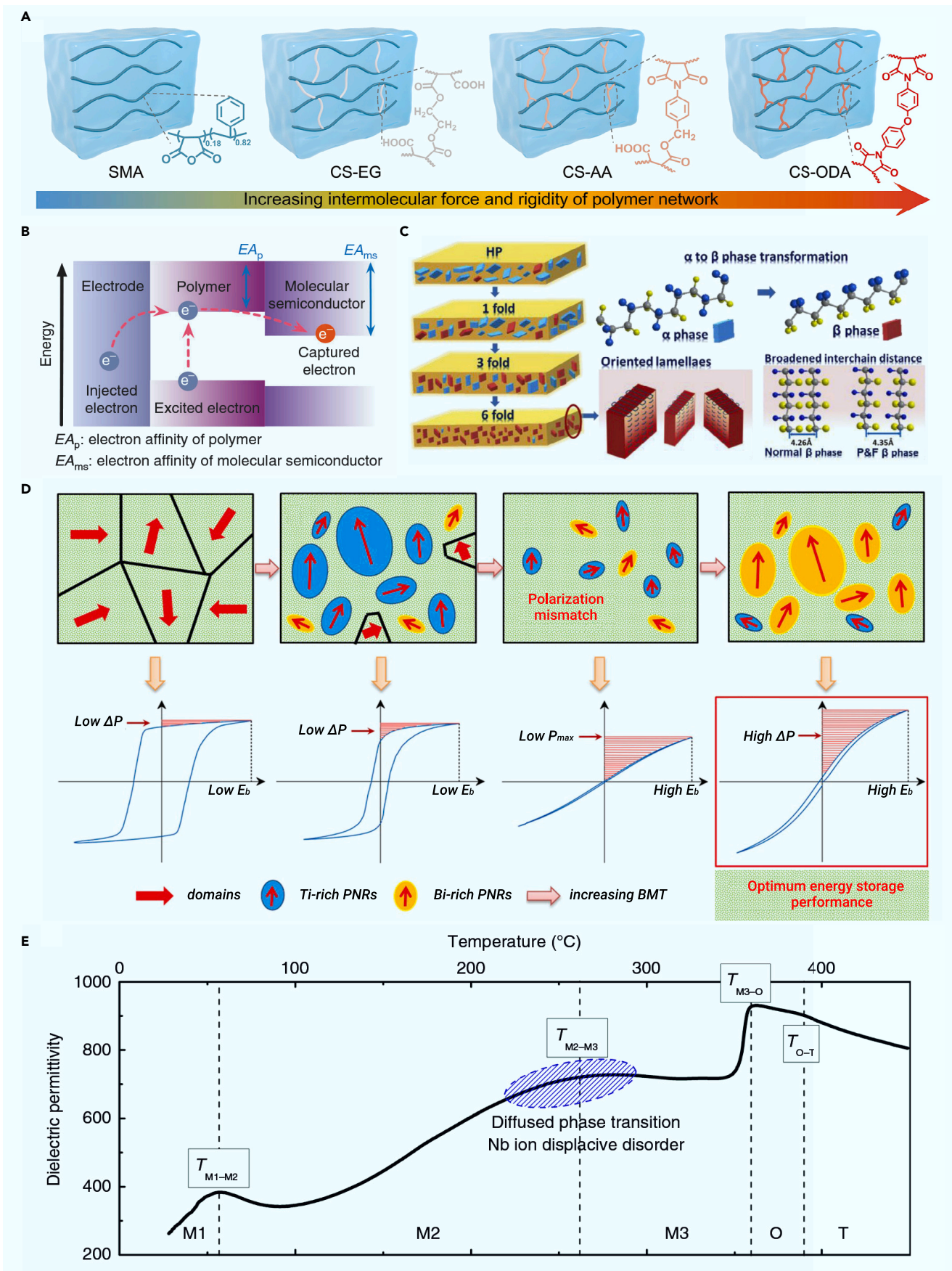**Figure 8. The typical methods for optimizing the performance of dielectric capacitors** (A) Schematic and structures of poly(styrene-co-maleic anhydride) (SMA) and the crosslinked polymers. Reprinted with permission from Pan et al.[251] Copyright 2022, Wiley-VCH. (B) Band diagram showing the possible charge transfer in the all-organic composite. Reprinted with

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increase the voltage.[233] Various strategies have been exploited, including adjusting the pH value, optimizing the surface of electrodes, adopting a "water-in-salt" electrolyte, or using organic solvents or ionic liquids as electrolytes.[110,234] However, the cost, safety, and ionic conductivity problems of organic electrolyte and ionic liquids need to be carefully considered, which may also sacrifice the power density and energy density.

There is no doubt that for either batteries or ECs, energy density and power density are both essential. The pursuit of both energy density and power density of electrochemical cells has blurred the boundary between batteries and supercapacitors. However, as pointed out by Deng Xiaoping, "It doesn't matter if a cat is black or white, so long as it catches mice." A bright development is that the first international standard for supercapacitor electrodes of both EDLCs and pseudocapacitors (International Electrotechnical Commission Technical Specification [IEC TS] 62565-5-2, "Nanomanufacturing—Material Specifications—Part 5-2: Nano-Enabled Electrodes of Electrochemical Capacitor—Blank Detail Specification") has been established and officially released recently under the auspices of the Shanxi Institute of Coal Chemistry of the Chinese Academy of Sciences.[235] The standard will unify the terminology, standardize the production process and establish product specifications, promote technical exchange and cooperation among related industries, and provide standard support for eliminating trade barriers. With the development of both "black and white cats," electrochemical cells will take more important roles in energy storage market.

DIELECTRIC CAPACITORS

Dielectric capacitors charged and discharged by electric-field-induced dielectric polarization and depolarization possess high power density ($\sim 10^4$–10^7 W/kg) (Figure 1D) because of their fast charging-discharging rates (approximately nanoseconds or microseconds)[236,237] and are promising for modern power devices and pulse power equipment such as electric/hybrid energy vehicles, consumer electronics, medical equipment, smart grids, electromagnetic weapons, and renewable energy systems. The core components of dielectric capacitors are the dielectrics that determine the energy storage performance. Recently, the demand for miniaturized and integrated devices has brought about a dramatic increase in research on dielectrics. The exploration of high-energy-storage dielectric materials focuses mainly on polymers,[238,239] ceramics,[240,241] and their composites.[242,243] Organic polymers have favorable processing properties and a high breakdown electric field (E_b) but relatively low dielectric constant (ε_r) and are susceptible to environmental factors such as temperature and humidity. Inorganic oxide ceramics have high ε_r and high-temperature resistance but are prone to cracking and breakdown because of mechanical stress. Polymer-ceramic composites combine the advantages of polymers and ceramics but have a complex composite process and relatively high cost. Therefore, future development directions mainly include (1) developing new materials with high dielectric properties, high temperature stability, large E_b, and durability; (2) exploring new manufacturing processes to optimize the interface properties and reduce cost; and (3) designing new device structures, such as staggered arrangements and multilayer structures.

On the basis of the different dielectric responses to applied electric fields, the dielectric materials can be divided into two categories: linear dielectrics (LDs) and nonlinear dielectrics.[244] LDs that exhibit linear polarization response to the applied electric field always demonstrate minimal energy loss (W_{loss}),[245] making them significant advantages in terms of E_b and energy efficiency (η). However, for LD polymers, the low energy density of LDs due to the relatively low ε_r severely hindered the practical applications.[246,247] Until now, biaxially oriented polypropylene (BOPP) films are one of the primary commercial dielectrics, with a market share of approximately 50%.[238,248] By introducing polar groups such as -OH and -NH$_2$ or grafting with maleic anhydride, the ε_r of polypropylene (PP) can be significantly increased.[249] Highly thermal stable polyimides (PI) have been developed for high-performance dielectric capacitors, such as Kapton (developed by DuPont) and UPILEX (developed by UBE).[250] Recently, by optimizing the molecular structures and controlling the trap-state energy level of polymer dielectric networks to inhibit high-temperature conduction loss, an ultrahigh recoverable energy density (W_{rec}) of more than 7 J cm^{-3} with $\eta > 90\%$ was achieved in poly(-styrene-co-maleic anhydride) (SMA) at 150°C (Figure 8A).[251] Incorporating appropriate polymers as organic fillers is also an effective way to optimize energy storage performance. Polymers blended with high-electron-affinity molecular semiconductors exhibit both high W_{rec} (3.0 J cm^{-3}) and high η (90%) up to 200°C (Figure 8B).[252] For LD ceramics, such as CaZrO$_3$ and CaTiO$_3$ ceramics, suitable dopants/oxide additives, solid solution, structure regulation (such as core-shell structure, layered structure), and sintering process are the main methods to optimize the energy storage performance.[253–255] However, the increase in ε_r often accompanied by the increase in dielectric loss and induced thermal management problems and electric-field-induced stress, leading to early failure and low reliability of capacitors.[256] It has been proposed that maximum energy density can be achieved in intermediate rather than high-ε_r materials, as they exhibit larger E_b.[257] Therefore, the development of new high-performance LDs should solve the primary problems of (1) the strong coupling between ε_r and E_b (from a scientific point of view) and (2) cycling life (from an engineering application point of view).

As for nonlinear dielectrics, their dipole rearrangement, domain inversion, and domain wall movement determine a nonlinear polarization response of ε_r to an applied electric field. Compared with ferroelectrics (FEs) with large maximum polarization (P_{max}), a moderate E_b, but low W_{rec} and η caused by high remnant polarization (P_r), relaxor FEs (RFEs) have higher P_{max} and lower P_r. Besides, they also have high dielectric properties in a wide temperature range because of their distinctive macro/micro-structure and diffuse phase transition. Polyvinylidene fluoride (PVDF), a typical FE polymer, has high ε_r due to its inherently strong polar C-F bonds and dipolar spontaneous polarization.[258] To reduce its high dielectric loss caused by the large FE domains during charging and discharging processes, several chemical or physical routes have been used to modify PVDF and its copolymers, such as electron irradiation, the introduction of a third larger monomer, and the grafting of poly(ethyl methacrylate) (PEMA). Such strategies can reduce the domain size of FE and realize the phase transition from FE to RFE.[259,260] In addition, more suitable for large-scale production of physical blending is also an effective approach through averaging to significantly reduce the dielectric loss of PVDF-based dielectric capacitors. But it also leads to significant decreases in ε_r and E_b because of the formation of conductive channels via the 3-3 structure of the blends in the polymer. The internal lattice stress engineering of PVDF is reported to induce relaxor-like FE behavior (Figure 8C).[261]

Realizing RFEs through a chemical modification to smash FE domains is also applicable in ceramics (Figure 8D),[262,263] such as many potential lead-based RFE ceramics, including PbZrO$_3$-SrTiO$_3$ (PZ-ST),[264] Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$-PbTiO$_3$ (PMN-PT),[265] and La-doped Pb(Zr,Ti)O$_3$.[266] But considering the toxicity of lead-based compounds, lead-free ceramics, such as BaTiO$_3$-based dielectric ceramics, have attracted a lot of attention. Most BaTiO$_3$-Bi(Me$_1$,Me$_2$)O$_3$-based (where Me$_1$ = Li$^+$, Mg^{2+}, Zn^{2+}, Al^{3+}, etc., and Me$_2$ = Zr^{4+}, Ti^{4+}, Nb^{5+}, etc.) RFEs have unique properties, including high insulation resistance even at high temperatures and a relatively stable high ε_r in a wide temperature range. Through A-site (e.g., Bi ions with a lone-pair electron 6s^2 configuration) and B-site doping (e.g., different valence and ionic radius from Ti to perturb long-range polar coupling) can increase P_{max} and minimize P_r, respectively. Doping and forming binary/ternary solid solutions to decouple the long-range polar without sacrificing average ionic polarizability can induce the relaxor behavior and improve the E_b of (Na,Bi)TiO$_3$-based ceramics.[267]

The large P_{max} and low P_r of antiferroelectrics (AFEs) due to the anti-parallel dipoles at low electric fields and the electric-field-induced reversible FE phase at high electric fields make AFEs a major candidate for energy storage capacitors. The key for high-performance AFEs is to improve the switching electric field (E_{AFE-FE} and E_{FE-AFE}), reduce hysteresis ($\Delta E = E_{AFE-FE} - E_{FE-AFE}$), and increase P_{max}. Various approaches have been adopted, such as chemical modification,[268] different fabrication methods,[269,270] and macro-scale and micro-scale mechanical constraints.[271] For example, to obtain stable PbZrO$_3$-based AFE ceramics, various dopants, such as Ba, La, Y, Sr, and Sm in the Pb sites and Sn, Ti, and Nb in the Zr sites, have been used.[272–274] Also, some new lead-free materials have exhibited AFE behavior, such as silver niobate (AgNbO$_3$ [AN]), sodium
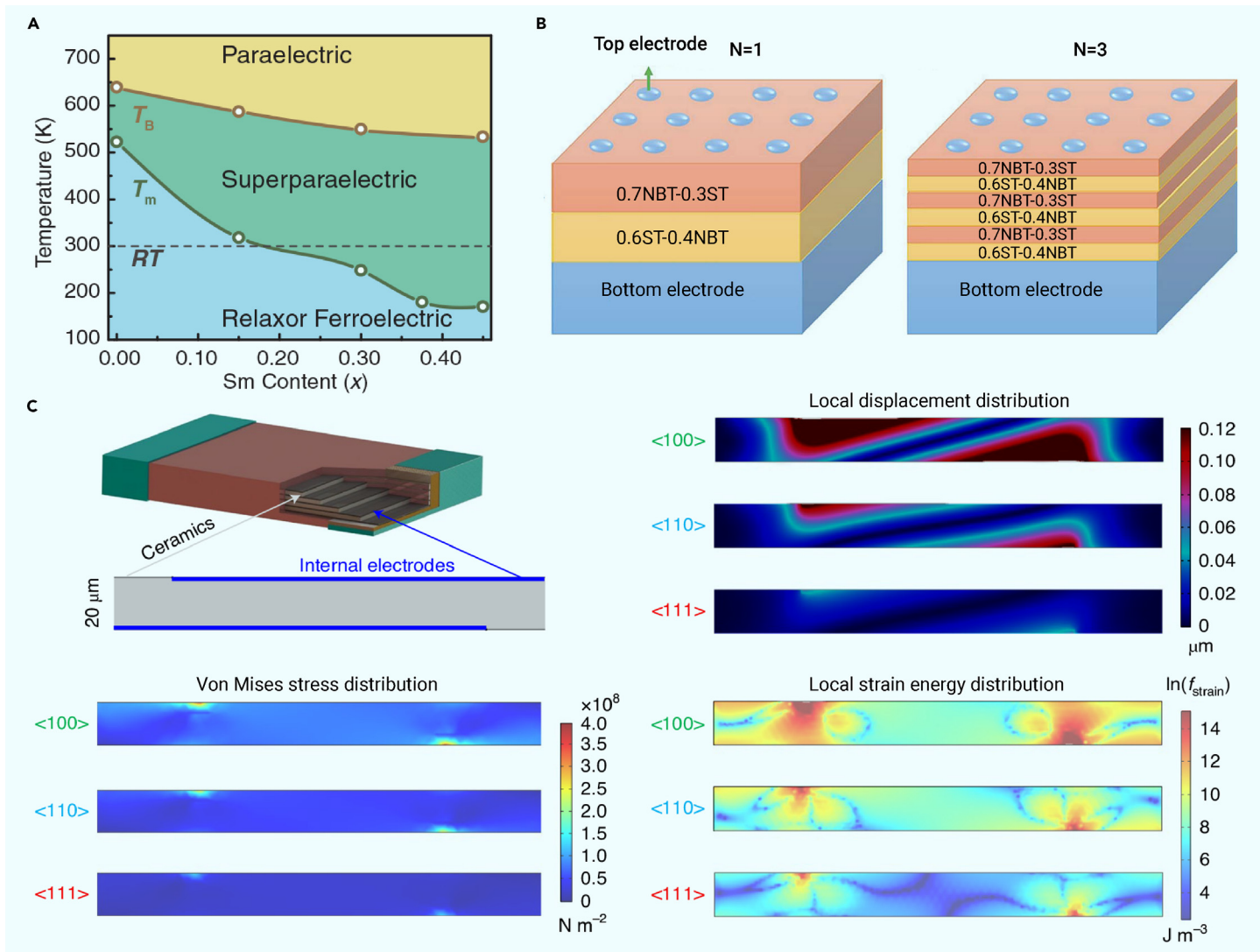
Figure 9. Optimization methods and developments of multilayer ceramics (A) Phase diagram of the Sm-BFBT films with respect to temperature and Sm content *x*. Reprinted with permission from Pan et al.[279] Copyright 2021, AAAS. (B) Sketch of the 0.7NBT-0.3ST/0.6ST-0.4NBT multilayer films with N = 1 and N = 3 and the same total thickness. Reprinted with permission from Zhang et al.[285] Copyright 2018, Royal Society Chemistry. (C) Finite-element simulations for the distribution of strains and elastic energies in a single layer of the MLCC. Reprinted with permission from Li et al.[292] Copyright 2020, Springer Nature.

bismuth titanate ([Na,Bi]TiO$_3$ [NBT]), and sodium niobate (NaNbO$_3$ [NN]),[275–277] which achieve induction and stabilization of AFE phases through specific chemical composition design and regulation. For instance, a high W_{rec} of 6.3 J cm^{-3} with η of 90% can be simultaneously achieved by constructing a room-temperature M2-M3 phase boundary in (1-x)AgNbO$_3$-xAgTaO$_3$ (Figure 8E).[278] However, the AFE-FE phase transition is usually accompanied by huge electric-field-induced strain, resulting in the fatigue phenomenon during switching cycles. Mechanical confinement, including external pressure and internal chemical pressure, is an effective method to reduce electric-field-induced strain and energy loss of AFE capacitors.[271] Compared with external physical pressure, the formation of internal chemical pressure can be easily achieved by introducing ions without using any external device.[273]

Emerged polymer-ceramic FE composites are expected to combine the advantages of polymers (high E_b) and ceramics (high ε_r) to achieve the high ε_r via introducing ceramic fillers into PVDF near the percolation threshold but at the expense of decreasing E_b and η. Further modifying the nano-/micro-structure of polymer-ceramic composites can improve E_b and η. Specifically, it can be achieved by inhibiting carrier mobility, tuning the crystal structure, improving the heterogeneity of the interface, etc. In the future, the key points for performance breakthroughs are tuning the organic-inorganic interface and reconciling the mutual constraints of filling content and flexibility.[245]

High W_{rec} is beneficial to minimize the size and weight of dielectric capacitors, meeting the miniaturization and lightweight requirement of equipment. Recently,

an ultrahigh W_{rec} of 152 J cm^{-3} with a high E_b of ~5.2 MV cm^{-1} was achieved in super-paraelectric samarium-doped bismuth ferrite-barium titanate (Sm-BFBT) films, demonstrating the potential of ceramic thin films in the field of energy storage (Figure 9A).[279] Numerous studies have proved the effectivity of multilayer ceramics (thin films/thick films) and emphasized influence of multilayer structural parameters (the number of layers, the thickness of the monolayer, interface density, layer order, and layer pairing) on energy storage performance with favorable controllability and fatigue life.[280–284] There are some special effects in the ultrathin nanolayers after combining two or more ceramic dielectric phases in a layered structure (Figure 9B),[285] such as the space-charges effect,[286] electric field redistribution,[285] heterojunction effect,[287] and interfacial dead layer.[288] Thus, it is an inevitable development trend for multilayer ceramic thin films to rationally design multilayer structures and fully use these effects. The interfacial polarization effect and blocking effect can effectively increase the P_{max} and E_b of multilayer thick films according to Maxwell-Sillar-Wagner's model and electric tree growth model.[289,290] Multilayer ceramic capacitors (MLCCs), which are ceramic thick films with a multilayer internal electrode structure, can obtain a larger capacitance capacity.[291] The development trend of MLCCs is as follows: (1) optimizing dielectric ceramics to increase ε_r, (2) reducing the thickness of dielectric monolayer to improve E_b, (3) increasing the overlap area of the inner electrode to improve capacitance, and (4) optimizing the co-sintering process to improve the reliability of capacitors. Linear and nonlinear inorganic materials have great potential to improve the energy storage performance of MLCCs.

Tokyo Denki Kagaku (TDK) of Japan pioneered the launch of CeraLink series capacitors on the basis of $(Pb,La)(Zr,Ti)O_3$ (PLZT). At present, many efforts have been made to reduce the strain, such as the preparation of high-quality textured $Na_{0.5}Bi_{0.5}TiO_3$-$Sr_{0.7}Bi_{0.2}TiO_3$ with lower electric-field-induced strain (Figure 9C),[292] providing a valuable direction for domestic enterprises to develop pulse-energy-storage MLCC products. Reducing strain and defects under the multiscale (atomic scale, nanoscale domain, micro-scale grain, and macro-scale multilayer) structure may be the main direction of optimizing the fatigue resistance of MLCCs in the future.

Fatigue resistance and thermal stability in extreme environmental conditions are also important issues with dielectrics, which have made remarkable progress on the basis of various compositional and structural design approaches. Typically, thermally crosslinked divinyl-tetramethyldisiloxane-bis(benzocyclobutene) (BCB) in the presence of boron nitride nanosheets (BNNSs) can afford the crosslinked nano-composite c-BCB/BNNS, exhibiting a discharged energy density (W_{dis}) of 1.8 J cm^{-3} at 250°C.[293] Recently, self-assembling ladderphane copolymers into highly ordered arrays by π-π stacking interactions resolved the contradiction between electrical resistivity and thermal conductivity, and achieve a W_{dis} of 5.34 J cm^{-3} with a charge-discharge efficiency of 90% at 200°C.[294] Compared with bulk ceramics, ceramic thin films have the additional advantage of mechanical flexibility. In particular, a high-entropy stabilized $Bi_2Ti_2O_7$-based film achieves W_{rec} as high as 182 J cm^{-3}. It also exhibits favorable cycling reliability and temperature stability.[295]

CONCLUSION AND OUTLOOK

Over the past century, carbon emissions have drastically increased, resulting in global climate change and increasing natural disasters that call for sustainable development. Since the Paris Climate Change Agreement, many countries and parties have made pledges and commitments to a zero-carbon world. Renewable energy is key to reduce carbon dioxide emissions. However, until now, the use of renewable energy has been quite low. With increasing share of intermittent renewable energies, energy storage technologies are needed to enhance the stability and safety of continuous supply. Among various energy storage technologies, mobile energy storage technologies should play more important roles, although most still face challenges or technical bottlenecks. In this review, we have provided an overview of the opportunities and challenges of rechargeable batteries, fuel cells, ECs, and dielectric capacitors, which will be beneficial to the further development of mobile energy storage technologies and boosting carbon neutrality. Despite enormous advances being made, there are still many issues to be solved.

Keeping up with the development of novel materials, the combination of machine learning algorithms and materials genome engineering could be used to advance the materials informatics and promote the development of materials science. The use of data-driven machine learning algorithms to build material-performance prediction models and then to assess key properties of new materials has shown its merit in various applications[296,297] and is expected to accelerate the process of material innovation, shorten the discovery-development-production-application cycle, and reduce the human and material costs for energy storage applications.[298]

Currently, there are always gaps between the performance reported in scientific reports and the practical performance in commercial devices. Gaps may also exist among different laboratories. Therefore, there is a need for all-around standards for not only material characterization, device fabrication, and performance testing but also the processing and description of data. Such standards are especially important for the potential application in machine learning with uniform data.

To obtain deep understanding of the materials and structure-property relationships, more advanced characterization techniques should be developed to reveal high-throughput, in situ and real-time investigations with high accuracy at even nanometer scales or even angstrom levels, which could provide information on the structural evolution, charge transport and storage mechanism, and side reactions, etc. Quantitative characterizations are of high priority. Such experimental results could be aided by advanced theoretical simulation techniques.

Last but not least, the aforementioned energy storage technologies have their own strengths and shortcomings. To meet the energy demands for various complex real scenarios, an effective and feasible solution is to couple different types of energy storage technologies to take advantage of their strengths and compensate for their weaknesses. Taking electric vehicles as an example, ECs or dielectric capacitors with high power density could be used to start the equipment instantaneously or provide power for rapid acceleration, while the fuel cells or rechargeable batteries with high energy density could be combined to provide steady endurance. Moreover, power device could be used to recycle energies in seconds from sudden braking. Collaborative efforts in interdisciplinary fields such as chemistry, materials science and engineering, and applied physics are expected to break through the limits of energy storage devices, which will boost carbon neutrality.

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AUTHOR CONTRIBUTIONS

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DECLARATION OF INTERESTS

The authors declare no competing interests.

LEAD CONTACT WEBSITE

The lead contact's website is at http://flexbatt.oei.hust.edu.cn.