

# Carbon-coated current collectors in lithium-ion batteries and supercapacitors: Materials, manufacture and applications

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## Abstract

The current collector is a crucial component in lithium-ion batteries and supercapacitor setups, responsible for gathering electrons from electrode materials and directing them into the external circuit. However, as battery systems evolve and the demand for higher energy density increases, the limitations of traditional current collectors, such as high contact resistance and low corrosion resistance, have become increasingly evident. This review investigates the functions and challenges associated with current collectors in modern battery and supercapacitor systems, with a particular focus on using carbon coating methods to enhance their performance. Surface coating, known for its simplicity and wide applicability, emerges as a promising solution to address these challenges. The review provides a comprehensive overview of carbon-coated current collectors across various types of metal and nonmetal substrates in lithium-ion batteries and supercapacitors, including a comparative analysis of coating materials and techniques. It also discusses methods for manufacturing carbon-coated current collectors and their practical implications for the industry. Furthermore, the review explores prospects and opportunities, highlighting the development of next-generation high-performance coatings and emphasizing the importance of advanced current collectors in optimizing energy device performance.

## KEYWORDS

batteries, carbon coating, current collector, energy storage devices, material solutions, supercapacitors

Hongqing Hao and Rui Tan contributed equally to this study.

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## 1 | INTRODUCTION

Low-carbon energy storage devices have found applications across a broad spectrum, from portable devices like wireless earphones<sup>1</sup> and personal laptops to larger systems such as energy grids and photovoltaic power stations. Batteries and supercapacitors stand out among existing energy storage devices due to their noteworthy features, including high energy and power densities and modest safety,<sup>2,3</sup> as well as their great potential for curtailing CO<sub>2</sub> emissions.<sup>4</sup> Despite these advancements, energy from fossil fuels still dominates market shares in sectors like the automotive industry.<sup>5</sup> Even with fervent governmental promotions, electric vehicles (EVs) constituted only 1% of the global automotive market in 2020, falling significantly short of the sustainability goals set for 2030.<sup>6</sup> Bridging such a substantial gap necessitates innovations in cost-effective, high-performance and safe energy storage systems, particularly focusing on batteries and supercapacitors.<sup>7,8</sup>

The structural configurations of Lithium (Li)-ion batteries (LIBs) and supercapacitors are illustrated in Figure 1, in which an electrolyte membrane is sandwiched between the cathode and the anode. The operational mechanisms differ between LIBs and supercapacitors. In the case of LIBs, Li ions migrate from the cathode to the anode during charging and vice versa during discharging, facilitating a current flow to deliver electrical energy. In contrast, supercapacitors adopt a distinct charge storage approach. Taking electrochemical double-layer supercapacitors as an example, they accumulate charges through a physical process where electrolyte ions adhere to electrode surfaces via electrostatic forces. This process maintains a consistent electrode structure while the

electrode potential shifts linearly.<sup>9</sup> Both energy storage devices can provide energy and power as tailored to the specific requirements of given application scenarios.

Regarding component materials, batteries typically incorporate cathode materials such as LiFePO<sub>4</sub>, LiNiMnCoO<sub>2</sub> and LiNiMnO<sub>2</sub>, while anodes are composed of Li metal, graphite and other materials such as silicon (Si)-based compounds.<sup>10,11</sup> Supercapacitors, on the other hand, utilize electrode materials primarily composed of carbon-based compounds, metal oxides, and conductive polymers.<sup>12,13</sup> Electrolyte separators in both batteries and supercapacitors facilitate charge transfer between the cathode and the anode.<sup>14–16</sup> The electrolyte types consist of liquid, gel, polymer, ionic liquid electrolyte and most recently solid electrolyte, whereas the separator is usually microporous polyolefin membranes, including polyethylene (PE) and polypropylene.<sup>17</sup> Notably, current collectors are indispensable for connecting the internal components of energy storage devices to external power sources, directly determining the energy and power output.<sup>18</sup> However, research on current collectors is often overlooked.

In practical terms, current collectors wield significant influence over the mass and overall weight of energy storage devices. For example, in LIBs, the mass percentage of the current collectors constitutes 15% of the total mass.<sup>19</sup> The development of lightweight current collectors provides a direct strategy to enhance the gravimetric energy density, and high safety of LIBs.<sup>20</sup> However, while energy can be increased with thinner and lighter current collectors, this comes at the cost of compromised heat transfer ability and electric conductivity, resulting in a reduction of device power and posing challenges to operational safety. A durable strategy is required to

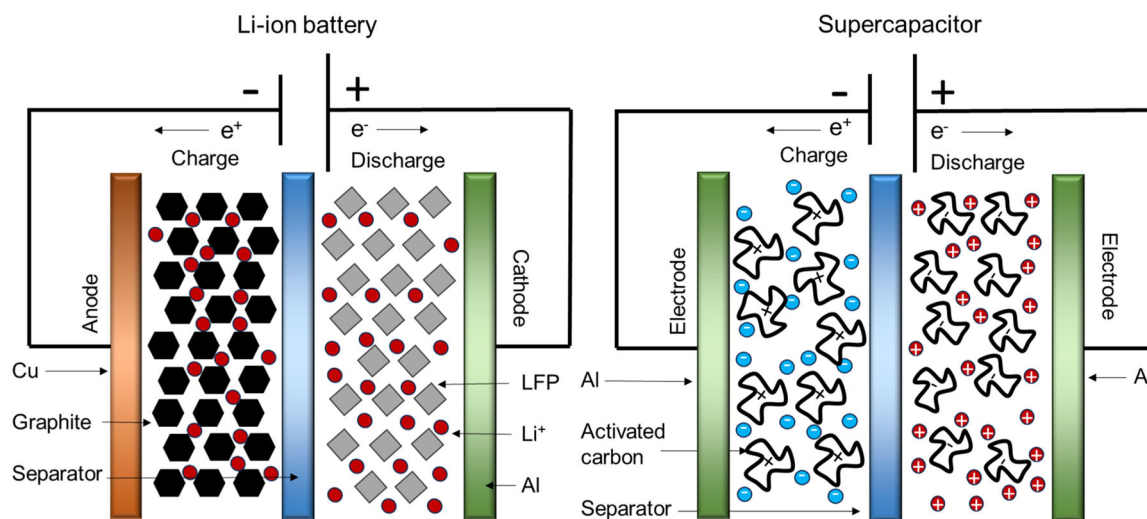


FIGURE 1 Prototypical configurations of the LIB and supercapacitor.

balance the aforementioned trade-off.<sup>21</sup> To realize this goal, various material solutions and surface engineering approaches need to be considered. Among different types of coating materials, carbon-based materials prove to be highly suitable as a coating medium to enhance the performance of energy storage devices due to their good chemical stability, supreme electric conductivity, and excellent corrosion resistivity.<sup>22,23</sup> Despite considerable research efforts successfully using the carbon-coating strategy in LIBs, a gap still exists between the know-hows of practical methods and know-whys behind the working principles. Although there have already been review articles discussing various current collectors within LIBs and supercapacitors, none of these has paid specific attention to the material, manufacture and application of the carbon coatings for the current collector of LIBs and supercapacitors.<sup>24–26</sup> This review aims to bridge this gap by systematically summarizing various material solutions, shedding light on the role of carbon coatings in enhancing performance, and providing guidance for future research.

## 2 | REQUIREMENTS FOR SELECTING CURRENT COLLECTORS

The optimal current collector should possess a combination of essential characteristics, including excellent electrical and thermal conductivities, strong adherence to electrode materials, a high surface area, mechanical durability, light-weight design, stability under electrochemical conditions,

cost-effectiveness and eco-friendliness. In LIBs and supercapacitors, metal foils (e.g., aluminium (Al) and copper (Cu)) and carbon-based materials are widely used. However, the selection criteria consistently emphasize the need to ensure efficiency and reliability across diverse applications. Critical parameters and properties are summarized in Table 1, with their detailed elaborations provided below.

### 2.1 | High electrical conductivity

The primary function of the current collector is to facilitate the flow of electrons generated by the electrodes. This involves both the internal conductivity of the current collector and the interfacial conductivity between the electrodes and current collectors. The selection process is influenced by the type of electrode and electrolyte.<sup>27</sup> As a physical attribute, internal electrical Conductivity = 1/Resistivity = 1/RA correlates with the ohmic resistance,  $R$ , and dimensions of conductive materials as expressed by the equation,  $\sigma = \frac{L}{RA}$ , where  $L$  is the length and  $A$  is the area. As summarized in Table 1, typical metal foils and carbon-based current collectors generally show high ionic conductivity up to  $1.0 \times 10^6 \text{ S cm}^{-1}$ , meeting the standard requirement for an effective conductive current collector.

### 2.2 | Heat dissipation

Safety concerns, particularly regarding thermal issues, have garnered significant attention in advanced LIBs and

**TABLE 1** Summary of the key properties of existing current collectors.

	Al	Cu	Ni	Ti	Stainless steel	Carbon material (graphene)
Internal electrical conductivity ( $\text{S m}^{-1}$ )	$3.77 \times 10^7$ ( $\pm 1.39 \times 10^6$ ) ( $\pm 3.6\%$ )	$5.95 \times 10^7$ ( $\pm 5.00 \times 10^5$ ) ( $\pm 0.8\%$ )	$1.44 \times 10^7$ ( $\pm 1.47 \times 10^5$ ) ( $\pm 1.0\%$ )	$2.56 \times 10^6$ ( $\pm 1.27 \times 10^5$ ) ( $\pm 4.9\%$ )	$1.39 \times 10^6$ ( $\pm 4.24 \times 10^4$ ) ( $\pm 3.1\%$ )	$1.09 \times 10^7$ ( $\pm 2.53 \times 10^5$ ) ( $\pm 20\%$ )
Thermal conductivity ( $\text{W m}^{-1} \text{K}^{-1}$ )	237 ( $\pm 1.52$ ) ( $\pm 0.6\%$ )	398 ( $\pm 2.08$ ) ( $\pm 0.5\%$ )	97.5 ( $\pm 4.44$ ) ( $\pm 4.5\%$ )	17 ( $\pm 2.53$ ) ( $\pm 14.8\%$ )	50 ( $\pm 0.61$ ) ( $\pm 0.01\%$ )	4000 ( $\pm 3.40$ ) ( $\pm 22.7\%$ )
Window voltage (V vs. $\text{Li}^+/\text{Li}$ ) <sup>a</sup>	1–5	0–2	3–5.5	3–5	3–5	0–5
Density ( $\text{g cm}^{-3}$ )	2.7	8.96	8.9	4.51	7.9	2.26
Mechanical strength (GPa) <sup>b</sup>	69	130	210	120	190	$2.4 \times 10^6$
Price per unit (\$ per $\text{m}^2$ in 2023) <sup>c</sup>	141.1	305.5	333.4	365.82	104.2	100,000

<sup>a</sup>Electrochemical voltages were confirmed by considering the redox reactions of current collectors at the potential versus  $\text{Li}^+/\text{Li}$ .

<sup>b</sup>Mechanical strength refers to Young's modulus, which is confirmed by the hardness test.

<sup>c</sup>Data were collected from the Goodfellow Cambridge Limited.

supercapacitors. The current collectors play a direct role in the heat transfer from internal batteries to the external environment. Current collectors with efficient heat dissipation ensure the elimination of locally accumulated heat and effectively reduce the elevated temperature to mitigate thermal runaways of batteries. Thermal conductivity can be calculated by the following equation:  $K = \frac{Qd}{A\Delta T}$ , where  $Q$  is the amount of heat transfer,  $d$  is the distance between the two isothermal planes,  $A$  is the area of the surface, and  $\Delta T$  is the difference in temperature. Compared with current collectors based on Al and Cu, the thermal conductivity of nickel (Ni), titanium (Ti), stainless steel and carbon-based current collectors is notably lower (Table 1), raising thermal concerns when using these materials in energy systems. New materials with high thermal conductivity, for example, graphene-based materials ( $5000 \text{ W m}^{-1} \text{ K}^{-1}$ ), are required for the development of safer and advanced batteries and supercapacitors.<sup>28</sup>

### 2.3 | Electrochemical stability

Ideal current collectors are expected to conduct electrons without undergoing any redox reactions. Redox reactions can lead to irreversible alteration in the structure of current collectors, resulting in undesirable capacity fade and reduced device longevity. It is crucial for current collectors to remain stable throughout these processes, particularly when paired with electrodes with high or low potentials, as seen in examples like Ni manganese cobalt oxide (NMC) (4.6 V vs.  $\text{Li}^+/\text{Li}$ )<sup>29</sup> and graphite (0.01–0.25 V vs.  $\text{Li}^+/\text{Li}$ ),<sup>30</sup> in which the stability requirement is especially pronounced during the cycling process. Existing current collectors typically use Al, Ni, Ti and stainless steel with high electrochemical oxidant windows on the cathode side, while Cu and carbon-based materials are widely adopted on the anode side. Materials with a broader electrochemical window, such as 0–5 V versus  $\text{Li}^+/\text{Li}$ , are highly required, ensuring the stable and reversible reactions of active species. For certain battery configurations, such as bipolar batteries, the current collector also needs to have a broad voltage window to cater to both cathode and anode requirements.

### 2.4 | Appropriate density

The mass of the current collector significantly contributes to the total mass of the device, typically around 15%. In the case of LIBs, optimal density can

significantly enhance the energy density of the device. The equation for density calculation is provided below. A lower-density current collector contributes to an overall reduction in the weight of the battery, thereby improving its mass and volumetric energy density, and allowing for a more compact design without sacrificing capacity. The desirable density for lightweight current collector materials is lower than  $0.4 \text{ g cm}^{-3}$ . When comparing materials with the same volume, the mass of the carbon fibre paper decreases by nearly six times compared to Al. In addition, using a conventional conductive carbon coating of  $1 \mu\text{m}$  with a  $10 \mu\text{m}$  Al current collector can result in a nearly 10% increase in mass density.<sup>31</sup> Density can be calculated by the following equation:  $\rho = \frac{m}{V}$ , where  $\rho$  is the density,  $m$  is the mass and  $V$  is the volume of the object.

### 2.5 | Mechanical strength

To accommodate volume expansion during cycling processes like Si anode,<sup>32</sup> robust mechanical strength in current collectors is vital for ensuring the adherence of electrode materials. The equation for calculating mechanical strength is shown below. Common properties used to assess mechanical strength include Young's Modulus (listed in Table 1), tensile strength, and elongation at break. The typical tensile strength of Al material is 90 and Cu is 210 MPa and the elongations at break for Al and Cu are 35% and 60%, respectively.<sup>33,34</sup> In general, for a current collector to be considered to have good mechanical strength, Young's modulus should exceed 750 MPa, tensile strength should surpass 250 MPa, and elongation at break should be more than 80%. The standard measurement for a current collector to bend without cracking is 200 times; however, a novel current collector should exceed this, aiming for over 500 times. Moreover, the aging life should extend beyond 30 years. Mechanical strength (Young's Modulus) is as expressed by the equation  $E = \frac{\sigma}{\epsilon}$ , where  $E$  is Young's modulus,  $\sigma$  is the uniaxial stress or uniaxial force per unit surface, and  $\epsilon$  is the strain or proportional deformation (change in length divided by original length).

### 2.6 | Cost-effectiveness and sustainability

Reducing current collector costs holds the potential to lower overall device price, enhancing market competitiveness without compromising performance. Material



selection should consider environmental impact, recyclability and raw material sourcing to ensure the sustainability of the current collector. Market prices for current collectors are listed in Table 1, indicating that novel current collectors should have a unit price below \$100 per square meter. Beyond raw material costs, recycling scrap metal is crucial. For example, the price of Al ranges between \$0.26 and \$0.28 per kilo and the price of Cu ranges between \$4.46 and \$4.93 per kilo. The replacement of the current collector can be achieved by the carbon-coating method, and the anticipated cost of recycling novel current collector material should be below \$0.18 in the future.<sup>35</sup>

The electroconductive substrates delineated in Table 1 offer distinct benefits. Al and Cu stand out as ideal substrates for cathodic and anodic interfaces, owing to their favourable electrochemical stability and superior internal electrical conductivity. Their cost efficiency further enhances their appeal. However, these materials have limitations; Al is susceptible to side reactions at lower electric potentials and Cu is prone to oxidative degradation at high voltages. Additionally, their volumetric density and mechanical robustness are sub-optimal compared to carbonaceous materials. An idealized current collector for LIBs and supercapacitors would combine these attributes, aiming to surpass the exemplary electroconductivity threshold of  $6 \times 10^7 \text{ S m}^{-1}$ , a thermal conductivity over  $5000 \text{ W m}^{-1} \text{ K}^{-1}$ , an operative potential window from 0 to 5 V versus  $\text{Li}^+/\text{Li}$ , with a density below  $2.2 \text{ g cm}^{-3}$  and a mechanical strength surpassing  $2.4 \times 10^6 \text{ Pa}$ , all while maintaining economic viability. This requires a synergistic integration of various collector types, leveraging the high internal electrical conductivity and modest cost from metals such as Al and Cu, along with the remarkable thermal conductivity, appropriate voltage window, reduced density and enhanced mechanical fortitude from carbon derivatives like graphene. Such integration often involves the carbonaceous coating of traditional metallic substrates, optimizing Al and Cu through the application of carbon layers. Subsequent sections will introduce various carbon-coated current collectors designed for LIBs and supercapacitors, aiming to advance energy storage devices towards enhanced performance.

### 3 | FUNCTIONS OF COATINGS FOR CURRENT COLLECTORS

Coating approaches are a durable and practical strategy to engineer the surface of current collectors, providing synergistic properties such as enhanced interfacial electrical conductivity while maintaining corrosion

resistance and electrochemical stability. As an alternative choice for current collector modification, the three-dimensional (3D) structuring method of coating, is promising due to its simple and versatile process. It can be directly applied to the top of the substrate material without causing morphology deformation of the current collectors. Current coating approaches, depending on the materials used, include metal coating, polymer coating and carbon coating.

Metal-based material coating is recognized for its significant potential in current collector applications due to several key factors: (a) Lithophilic metals facilitate uniform Li deposition during the charging process, promoting the formation of a uniform and stable solid-electrolyte interphase (SEI) layer that effectively inhibits dendritic growth.<sup>36,37</sup> (b) Metal coatings can substantially improve the mechanical properties of the substrate, alleviating the challenges associated with volume expansion during the charge/discharge cycles, particularly with active materials such as Si.<sup>38</sup> (c) Inert metal coatings serve as a corrosion protection layer, enhancing the corrosion resistance of the current collectors.<sup>39</sup>

Polymer-based coating has several advantages: (a) Polymer coatings show a variety of functional groups that impart ionophilicity to the substrate, enhancing ionic conductivity.<sup>40</sup> (b) Adhesive characteristics of polymer coatings improve the cohesion between the active material and the current collectors, ensuring mechanical stability. (c) Polymer coatings can serve as a physical reinforcement for the current collector, attributed to the internal chain structure of the polymer material. (d) Due to their relatively low density, polymer coatings minimally impact the overall energy density of the battery.<sup>41</sup>

As an alternative coating material to both metal and polymer, carbon coating emerges as a facile and effective approach capable of achieving the aforementioned targets. Specifically, the benefits of using carbon coatings on current collectors are manifold:

- (1) *Enhanced adhesion* between the active material and the current collector due to the rough surface structure and the expansive surface area of carbon.<sup>42</sup>
- (2) *Reduction in interfacial resistance* between the active and substrate materials due to the high conductivity of carbon.<sup>22</sup>
- (3) *Effective protection* against corrosion agents or electrolyte decomposition, serving as a shield for the underlying current collector.<sup>43</sup>
- (4) *Improved device cyclability* attributable to its robust structure and chemical stability.<sup>23</sup>
- (5) *Augmented robustness* of the current collector and prevention of internal short circuits caused by

dendritic growth, thanks to the carbon coating's mechanical strength.<sup>44</sup>

- (6) *Stabilization of metallic substrate* achieved by preventing direct contact between the solvent and the metallic current collector, facilitated by the SEI layer atop the carbon coating, which selectively transfers Li ions to the current collector.<sup>45</sup>

The carbon-coating materials have been categorized into four types, including graphitic carbon, amorphous carbon, doped carbon and carbon-metal composite, as illustrated in Figure 2.

The integration of carbon, metal and polymer coatings has been explored extensively in recent research, aiming to synergize their unique properties for superior current collector performance.<sup>46,47</sup> The combination of carbon and metal composites has been shown to facilitate uniform Li deposition and direct Li<sup>+</sup> flux, with carbon enhancing interfacial electrical conductivity and corrosion resistance, and metals providing lithophilic sites.<sup>48</sup> The incorporation of conductive materials into nonconductive polymer coatings addresses the limitation of electron diffusion, presenting a viable strategy for enhancing the interfacial electrical conductivity of the coating.<sup>49</sup> The multi-component coating will also be introduced in detail in the following sections.

The subsequent section will delve into the application of carbon coatings on both metal- and nonmetal-based current collectors, highlighting their

benefits and advancements for batteries and supercapacitors.

## 4 | METALLIC CURRENT COLLECTORS

### 4.1 | Al-based current collectors

Al is widely chosen as the current collector for cathode materials in LIBs and supercapacitors due to its impressive electrical conductivity, ranking fourth among metals at  $2.65 \times 10^{-8} \Omega \text{ m}$  at a standard room temperature of 20°C. It also possesses notable physical properties, including a tensile strength of 25 MPa, material density of  $2.7 \text{ g cm}^{-3}$  and thermal conductivity of  $237 \text{ W m}^{-1} \text{ K}^{-1}$ .<sup>50</sup> The electrochemical behavior of Al has been thoroughly analysed in previous studies.<sup>51</sup> Figure 3A illustrates anodic and cathodic peaks occurring around potentials between 0 and 1 V versus Li<sup>+</sup>/Li, indicative of the alloying and dealloying processes of Al and Li, respectively. This electrochemical profile highlights the challenge of using Al as a current collector for various anode active materials, with the exception of lithium titanate (LTO), which operates at a potential of around 1.5 V versus Li<sup>+</sup>/Li.<sup>58</sup> A magnified view of the graph (on the right) reveals two distinct peaks on the anodic side, signifying the formation of a passivation film on the Al surface when potentials reach 3.7 and 4.7 V versus Li<sup>+</sup>/Li. This

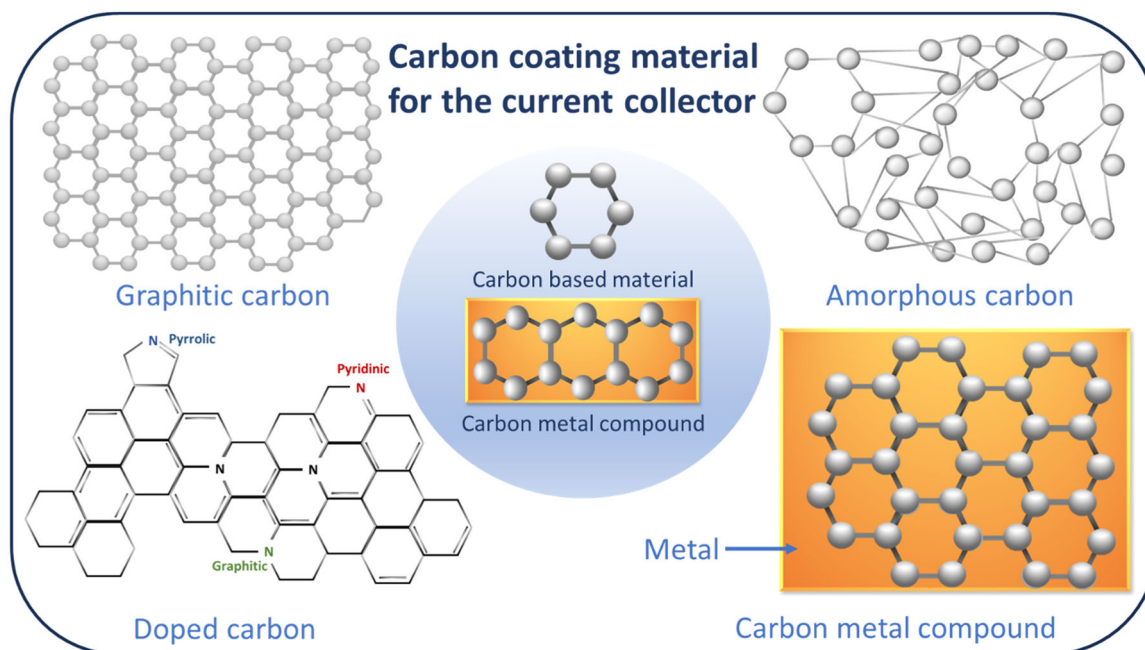
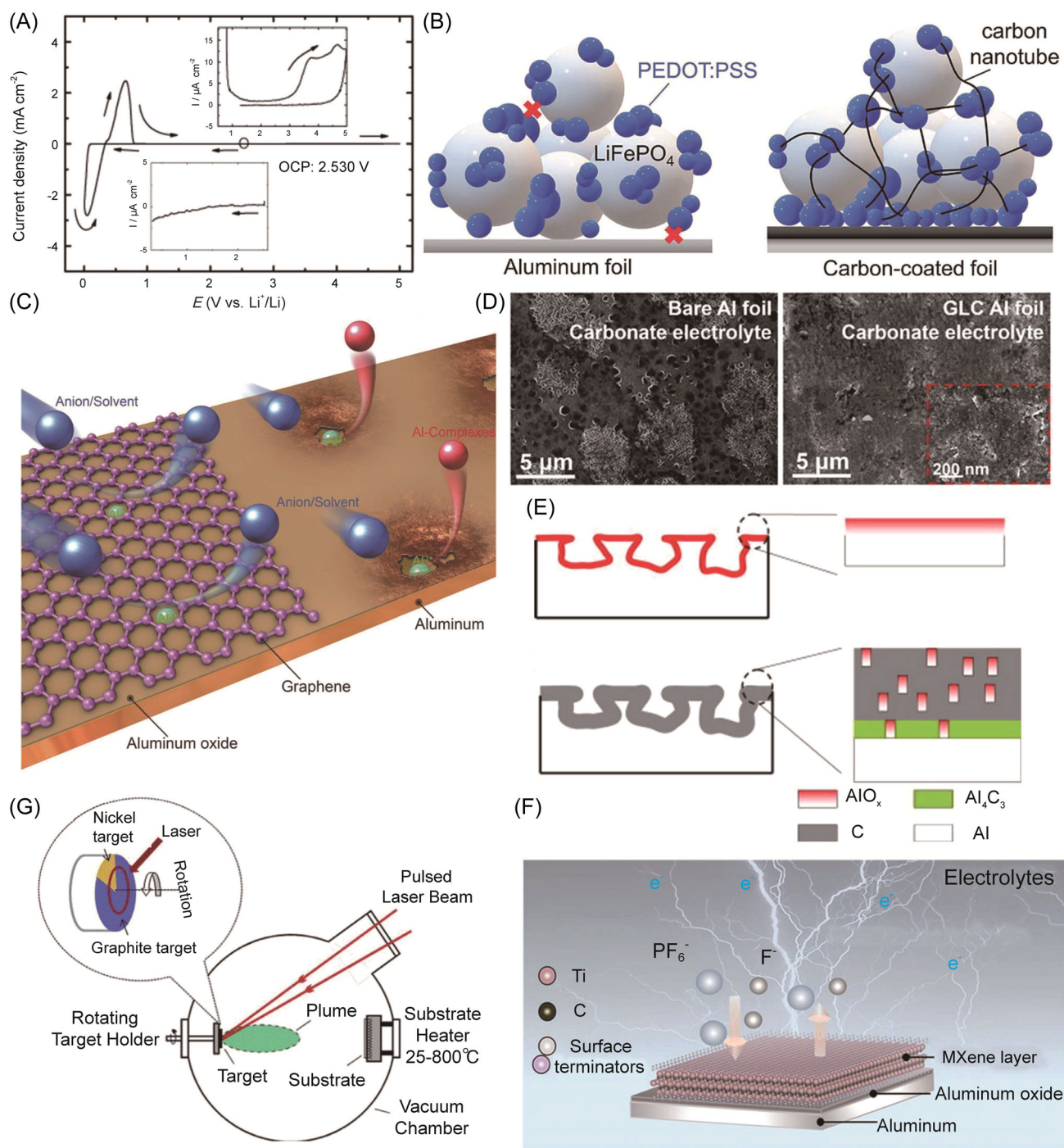


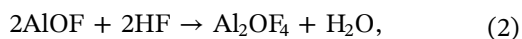
FIGURE 2 Classification of different types of carbon as coating materials for current collectors.



**FIGURE 3** Carbon coatings for Al-based current collectors. (A) Electrochemical behaviour of Al in 1 M  $\text{LiPF}_6$  in EC:DMC (1:1 vol%) electrolyte. Reproduced with permission: Copyright 2009, Elsevier.<sup>51</sup> (B) Nanotubes facilitating connections amongst discrete PEDOT:PSS particles. Reproduced with permission: Copyright 2009, AIP publishing.<sup>52</sup> (C) Graphene coating on Al-based current collectors. Reproduced with permission: Copyright 2017, Wiley.<sup>53</sup> (D) SEM images of an Al foil with and without graphene coating. Reproduced with permission: Copyright 2019, ACS Publications.<sup>54</sup> (E) Schematic diagram of oxide surface layer with and without carbon coating. Reproduced with permission: Copyright 2009, Elsevier.<sup>55</sup> (F) Illustration of carbon–metal compound coating on an Al foil. Reproduced with permission: Copyright 2022, Wiley.<sup>56</sup> (G) Example of the deposition method for the carbon–Ni composite using a KrF excimer laser. Reproduced with permission: Copyright 2021, IOP Science.<sup>57</sup>



protective film comprises layers of  $\text{Al}_2\text{O}_3$  at its base and  $\text{AlF}_3$  on its surface. The chemical reactions behind the formation of  $\text{AlF}_3$  from  $\text{Al}_2\text{O}_3$  and HF, especially during the decomposition of  $\text{LiPF}_6$  under optimal temperature, moisture conditions and high potentials, are presented in Equations (1), (2) and (3) as follows<sup>59</sup>:



Demonstrating commendable electrochemical stability, this passivation film formed on the Al surface acts as a safeguard, maintaining a near-zero current beyond 4.7 V, particularly when  $\text{LiPF}_6$  is used as the electrolyte. While certain electrolytes, such as  $\text{LiB}(\text{C}_2\text{O}_4)_2$ , also facilitate the formation of passivation films like  $\text{AlBO}_3$ , others, like  $\text{LiClO}_4$  and  $\text{LiCF}_3\text{SO}_3$ , prove incompatible. In these cases, the passivation layer fails to provide complete coverage on the Al surface.<sup>60</sup> To address these challenges, applying a carbon coating on Al emerges as a promising solution. This coating not only augments the conductivity of the Al current collector but also strengthens its resistance to corrosion. A detailed summary of carbon-coated Al current collectors can be found in Table 2.

Generally, carbon coatings are categorized into four different types: graphitic carbon, amorphous carbon, doped carbon, and carbon-metal compound.

#### 4.1.1 | Graphitic carbon

Graphitic carbon stands out as a preferred choice for current collector carbon coating material in various applications, owing to its features of high thermal stability, excellent electrical conductivity, industrial reliability, low cost and, most importantly, nontoxicity.<sup>86</sup> The category of graphitic carbons includes a wide range of materials, such as carbon nanotubes (CNTs), graphene, graphene oxide (GO), reduced GO (rGO), graphdiyne (GDY) and so forth.

CNTs, as quintessential one-dimensional carbon materials, are distinguished by their unique morphology and superior electrochemical attributes, including exceptional electrical conductivity, expansive surface area and rapid charge transport. These qualities persist even in the absence of consistent chirality and orientation.<sup>87,88</sup> Leveraging these inherent merits of graphitic carbon materials, single-walled CNTs (SWCNTs) have been used as coating agents for Al current collectors, using poly(3,4-

ethylenedioxythiophene) doped with polystyrene sulpho-nate (PEDOT:PSS) as a binder. As depicted in Figure 3B, the nanotubes facilitate connections among discrete PEDOT:PSS particles, significantly enhancing the adhesion of the active material. Consequently, the internal electrical conductivity across the PEDOT:PSS to the active material experiences a remarkable surge, increasing from  $0.21 \text{ S cm}^{-1}$  without SWCNT coating to  $8.0 \text{ S cm}^{-1}$  post-CNT coating. Furthermore, the resistance within these composite electrodes sharply decreases from  $181 \Omega$  to a mere  $5.6 \Omega$ .<sup>52</sup> Beyond SWCNTs, multiwalled CNTs (MWCNTs) are also gaining traction as preferred carbon coating materials, influencing the efficacy of LIBs. For instance, Rytel et al.<sup>64</sup> adopted the Langmuir-Schaefer technique to envelop the Al current collector in a thin MWCNT film. As a direct outcome, the charge-transfer resistance of the carbon-coated cell decreased from 39 to  $25 \Omega$ , and the capacity at an elevated rate (3 C) increased from 79 to  $107 \text{ mAh g}^{-1}$ .

Graphene, a prominent two-dimensional (2D) graphitic carbon material, is composed of hexagonal lattices of carbon atoms and offers advantages such as electrochemical stability, remarkable electrical conductivity, mechanical durability, flexibility and corrosion resistivity.<sup>89</sup> As depicted in Figure 3C, integration of a graphene coating significantly enhances the corrosion resistance of Al-based current collectors. In this application, graphene films are directly plated onto the Al current collector using chemical vapour deposition (CVD). Acting as a conductive shield, the graphene film protects Al from the detrimental effects of anions in corrosive media, thus drastically reducing corrosion susceptibility.<sup>53</sup> Moreover, the graphene layer notably improves the rate capability of the LIB, especially at high C rates. For instance, a graphene-armoured cell delivers a capacity of  $58 \text{ mAh g}^{-1}$ , in contrast to  $49 \text{ mAh g}^{-1}$  for bare Al. Graphene also finds application in lithium-sulphur batteries. Li et al.<sup>54</sup> used multilayer graphene as the carbon coating to suppress the corrosion of an Al foil, as shown in Figure 3D. The Al foil without graphene coating shows a severe porous corrosive structure, while the coated Al foil shows a homogeneous surface without any sign of corrosion. The carbon-coated cell shows an excellent capacity behaviour of  $150 \text{ mAh g}^{-1}$  after 100 cycles, which is 1.5 higher than the cell with bare Al. Apart from the mentioned types of graphene, Peng and colleagues<sup>84</sup> used plasma-enhanced CVD (PECVD)-coated hierarchical graphene as a powerful interlayer between lithium iron phosphate (LFP) and an Al current collector. The vertical graphene increases the area affected by van der Waals force, enhancing adhesion and facilitating free electron transfer between the active material and the current collector. Beyond applications



TABLE 2 Summary of Al-based carbon-coated current collectors.

Type of device	Carbon coating material	Electrode	Result with carbon coating (at C-rate) (mAh g <sup>-1</sup> )	Original result (mAh g <sup>-1</sup> )	Cycle number	References
LIB	Conductive carbon (C45)	NMC (cathode)	126 (1 C)	108	50	[61]
DIB	PAN film	Graphite (anode)	93 (2 C)	0	1000	[62]
LIB	CH <sub>4</sub>	LFP (cathode)	105 (5 C)	18	200	[63]
Lithium-sulphur battery	Graphene-like carbon	Sulphur (cathode)	750 (0.5 C)	500	100	[54]
LIB	MWCNT	LMO (cathode)	107 (3 C)	79	50	[64]
Lithium-sulphur battery	PEO	Sulphur (cathode)	650 (0.2 C)	550	100	[65]
LIB	Carbon-based ink	LFP (cathode)	79% capacity retention (2 C)	45% capacity retention (2 C)	200	[66]
Supercapacitor	CH <sub>4</sub>	AC (cathode)	18 Wh kg <sup>-1</sup> at 25 kW kg <sup>-1</sup>	0 Wh kg <sup>-1</sup>	/	[55]
Lithium-sulphur battery	Super P and layered graphite	Sulphur (cathode)	655 (0.2 C)	560	50	[44]
LIB	Carbon	LNMO (cathode)	120 (1 C)	100	200	[67]
LIB	Disordered carbon	LFP (cathode)	155 (0.1 C)	120	10	[68]
LIB	Graphene	LCO (cathode)	82.9% capacity retention (0.2 C)	75.8% capacity retention	100	[69]
Supercapacitor	HTMA and acac	AC (cathode)	100 F g <sup>-1</sup>	/	/	[70]
LIB	GO	LMO (cathode)	90 (3 C)	80	100	[71]
LIB	SWCNT	LFP (cathode)	150 (1 C)	40	100	[52]
Lithium-sulphur battery	Graphene	Sulphur (cathode)	659 (0.2 C)	390	150	[72]
SIB	Carbon	Carbon (anode)	600 (1 C)	0	200	[73]
LIB	Carbon-under coating layer	LCO (cathode)	250	230	/	[74]
LIB	CH <sub>4</sub>	LTO (anode)	170 (1 C)	130	25	[75]
LIB	CH <sub>4</sub>	LFP (cathode)	160 (1 C)	125	10	[76]
Supercapacitor	Carbon-nickel composite	AC (cathode)	28 F g <sup>-1</sup> (12.5 A g <sup>-1</sup> )	21 F g <sup>-1</sup>	10000	[57]
LIB	GNs, CNTs, ACs	LFP (cathode)	122 (GNs) 114.8 (CNTs) 106.8 (ACs) (1 C)	49.2	5	[77]
Supercapacitor	Carbon	Laminar composite (cathode)	134 F g <sup>-1</sup> (110°C) at 1 A g <sup>-1</sup>	81 F g <sup>-1</sup>	/	[78]
LIB	MXene-Ti <sub>3</sub> C <sub>2</sub>	NMC (cathode)	100 (0.5 C)	15	300	[56]

(Continues)

TABLE 2 (Continued)

Type of device	Carbon coating material	Electrode	Result with carbon coating (at C-rate) (mAh g <sup>-1</sup> )	Original result (mAh g <sup>-1</sup> )	Cycle number	References
LIB	CQDs	LMO (cathode)	115.1 (1 C)	92.5	100	[79]
LIB	Graphene-like carbon	LCO (cathode)	91.7 (1 C)	12.8	150	[80]
LMB	CNT	Li	92.4% Coulombic efficiency (1 mA cm <sup>-2</sup> )	40% Coulombic efficiency	50	[81]
SIB	GNs	NVP	82 (1 C)	64	10	[82]
LIB	Graphene	LMO (cathode)	58 (5 C)	49	10	[53]
Lithium-sulphur battery	Graphene-CNT hybrids	Sulphur	812 (0.5 C)	587	300	[83]
LIB	Hierarchical graphene	LFP	43 (20 C)	0	5	[84]
LIB	Prussian blue	LMO	87.8% capacity retention (1 C)	50.5% capacity retention	300	[85]

Abbreviations: AC, activated carbon; CNT, carbon nanotube; CQD, carbon quantum dot; DIB, dual-ion battery; GN, graphene nanosheet; GO, graphene oxide; LCO, LiCoO<sub>2</sub>; LIB, lithium-ion battery; LFP, lithium iron phosphate; LMB, lithium metal battery; LMO, lithium-rich manganese-based oxide; LTO, lithium titanate; MW, multiwalled; NMC, nickel manganese cobalt oxide; NVP, sodium vanadium phosphate; PEO, polyethylene oxide; SIB, sulphur-ion battery; SW, single walled.

in LIBs, graphene has also been utilized as a conductive film in sodium-ion batteries to enhance cell performance. Wu and colleagues<sup>82</sup> coated vertical graphene nanosheets using the PECVD method, significantly reducing interfacial resistance and increasing the contact between the cathode material (sodium vanadium phosphate [Na<sub>3</sub>V<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub>], NVP) and the current collector. The capacity at 1 C increased from 64 to 82 mAh g<sup>-1</sup> after 10 cycles compared to a pristine Al foil. In contrast, the cathode material with bare Al cannot even adhere suitably.

GO, a distinct variant of graphitic carbon, plays a crucial role as a conductive layer, particularly effective in batteries using nonaqueous electrolytes. Unlike graphene, which may exacerbate Al corrosion due to non-ion-selective unbiased ionic diffusion, GO, enriched with reactive oxygen functional groups, acts as a barrier against anions, effectively mitigating corrosion.<sup>90</sup> As an application, Richard Prabakar et al.<sup>71</sup> harnessed GO as a protective coating, demonstrating a significant reduction in Al corrosion. After 100 cycles, the bare Al foil shows a deeply rough surface, entirely corroded. In contrast, the GO-coated Al surface appears smooth, indicating successful corrosion inhibition with small trenches and pits. As a result, the lithium-rich manganese-based oxide (LMO)-based LIB with GO-coated Al shows a discharge capacity of 90 mAh g<sup>-1</sup> at 3 C, which is 11% higher than the performance achieved by the cell with bare Al.

Furthermore, GDY emerges as another promising coating material, lauded for its exceptional stability among various diacetylene-derived carbon allotropes.<sup>91</sup> Principally used as a coating for anode current collectors based on Cu, detailed applications of GDY will be elaborated upon in the following section.

In general, incorporating a graphitic carbon coating offers multiple performance enhancements in battery technologies. It can improve the electronic conductivity of the current collector, reduce the interface resistance between the electrode material and the current collector and establish a stable interface, ultimately enhancing the overall performance and lifecycle of the battery. Additionally, the carbon layer can act as a protective barrier, preventing Al from corroding or reacting with the electrolyte, particularly at higher potentials. While the addition of a graphitic carbon coating introduces an extra step in the manufacturing process, potentially increasing costs, the improved battery life and performance can often outweigh this drawback. Cost-effectiveness also depends on the scale of production; for large-scale manufacturing, economies of scale can make the coating process more affordable. However, for smaller-scale or custom applications, the additional cost might be more noticeable. Introducing a graphitic carbon coating to the Al current collector can complicate the manufacturing process. The coating needs to be uniform, adhere well to the Al surface and maintain its integrity

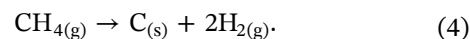
during battery cycling. Various methods like CVD or sputtering can be used to achieve this, but they require precise control and monitoring. The complexity also depends on the specific requirements of the battery application, as well as the quality and uniformity of the coating.

#### 4.1.2 | Amorphous carbon

Amorphous carbon, distinguished from its graphitic counterpart, is a favoured choice for carbon coating due to its exceptional interfacial electrical conductivity, robust mechanical strength and impressive elasticity. These attributes have positioned it as a popular choice for enhancing the performance of energy storage devices.<sup>66,92</sup> Conductive carbon was coated on an Al current collector to enhance the performance of NMC-based LIBs.<sup>61</sup> Notably, a morphological difference in Al becomes apparent when the cathode electrode is dried at 80°C in this application. Removing the precoated carbon exposes significant corrosion on the bare Al, attributed to the high reactivity of Al oxide at certain temperatures. In contrast, carbon-coated Al shows marked resistance to corrosive mediums, maintaining a smooth surface even at high temperatures. Performance-wise, the conductively coated cell outperforms its counterpart at a high rate of 5 C, delivering a capacity of 70 mAh g<sup>-1</sup> compared to the 21 mAh g<sup>-1</sup> by the bare Al. Additionally, the charge-transfer resistance of the carbon-coated cell shows a significant reduction. In another application, amorphous carbon enhances the performance of a dual-ion battery (DIB) on an etched 3D Al substrate with a porous structure.<sup>62</sup> DIBs, characterized by two ionic species as charge carriers, showcase a vast voltage window, commendable cycle performance and cost-effectiveness.<sup>93</sup> The process involves initial etching of the Al foil, followed by carbonization using polyacrylonitrile immersion and subsequent solidification; this carbon-coated foil serves as both the current collector and the anode material, with graphite used for the cathode. The results are promising, with the carbon-coated sample demonstrating an impressive high-rate cycling of 93 mAh g<sup>-1</sup> at 2 C for 1000 cycles. In contrast, the uncoated version shows zero capacity at this cycle number. The carbon-coated variant also strikes a balance between energy and power density, with the sample showing a good balance of energy/power density at an energy density of 204 Wh kg<sup>-1</sup> and a power density of 3084 W kg<sup>-1</sup>.

Amorphous carbon has proven to be beneficial for supercapacitors, especially in high-rate applications. Wu et al.<sup>55</sup> used CH<sub>4</sub> as a carbon source to coat an Al current

collector using CVD at 600°C. The process is represented as



The resulting carbon displaces Al oxide, forming a composite with an Al<sub>4</sub>C<sub>3</sub> interfacial layer topped with a carbon layer. The supercapacitor with this carbon coating shows significantly lower resistance than its pristine Al counterpart. Moreover, the carbon-coated cell achieves an energy density of 18 Wh kg<sup>-1</sup> at 25 kW kg<sup>-1</sup>, starkly contrasting with the uncoated version. The mechanism underlying this improvement in conductance due to carbon coating is elucidated in Figure 3E. The presence of the insulating oxide surface layer in the bare Al cell means that electrons are likely to traverse the thin side of the oxide layer, taking a lengthier path rather than flowing directly into the current collector. This elongated route diminishes the conductance of the current collector. However, introducing the conductive carbon layer ensures that electrons follow the shortest path to the current collector, replacing the oxide layer and substantially boosting electronic conductance in the contact area.

The application of amorphous carbon coating on Al current collectors plays a crucial role as a protective barrier, effectively guarding against corrosion and mitigating adverse reactions with electrolytes, thus extending the lifespan of the battery. Its unique ability to accommodate volume changes during cycling plays a pivotal role in preventing electrode delamination or degradation. Achieving a uniform and durable amorphous carbon layer relies on controlled deposition techniques, such as PECVD. However, maintaining a consistently amorphous structure without crystalline inclusions and ensuring strong adhesion to Al can be challenging. The complexity of the process varies depending on the required thickness and quality of the carbon layer. Despite the challenges, the deposition process contributes to the overall cost due to additional materials and steps involved. However, the benefits of an extended battery lifespan and improved performance can outweigh this increase. Cost implications are more pronounced in small-scale production but can be mitigated in larger operations through economies of scale.

#### 4.1.3 | Carbon-metal compounds

Carbon-metal compounds combine the advantageous properties of carbon, such as its high interfacial electrical conductivity and chemical stability, with the distinct characteristics of metals, like high internal electrical

conductivity or catalytic activity. This synergy results in enhanced electronic and ionic conductivities, improved mechanical durability and possibly additional electrochemical active sites. Yang et al.<sup>56</sup> used MXene ( $\text{Ti}_3\text{C}_2\text{T}_x$ ) nanosheets as an armoured layer for the Al current collector to counteract corrosion, particularly at high operating voltages exceeding 4.5 V versus  $\text{Li}^+/\text{Li}$ , as depicted in Figure 3F. These nanosheets, synthesized via a self-assembly process, involved etching Al layers from  $\text{MAX-Ti}_3\text{AlC}_2$  in a  $\text{LiF}/\text{HCl}$  solution. The resulting layer, observed through an atomic force microscope, boasted an ultrathin thickness of 1.5 nm. This MXene coating, uniformly spread across the surface of Al, amplified the electron conductivity of the current collector, spanning a wide voltage window from 2 to 5.5 V versus  $\text{Li}^+/\text{Li}$  in a  $\text{LiPF}_6$  electrolyte with a carbonate solution. Scanning electron microscopy (SEM) imagery revealed that, post-CV measurement, the uncoated Al showed significant surface damage, whereas the MXene-coated Al remained relatively unscathed. Consequently, the MXene-coated current collector paired with an NMC cathode delivered a commendable discharge capacity of  $100 \text{ mAh g}^{-1}$  at 0.5 C after 300 cycles. In contrast, the battery with bare Al managed a mere  $15 \text{ mAh g}^{-1}$  at this cycle number, highlighting the substantial contribution of the MXene coating to cell cyclability. Beyond these applications, the cycling stability at high rates of LMO is significantly constrained due to electrochemical polarization, a consequence of the sluggish kinetics associated with the  $\text{Li}_2\text{MnO}_3$  phase. To address this limitation, Hao et al.<sup>85</sup> have innovatively applied a Prussian blue interface onto the LMO surface. This modification aims to mitigate the adverse effects and prevent the release of oxygen from the surface of the active material. Because of this intervention, there has been a noteworthy enhancement in the capacity retention at a 1 C rate over 300 cycles, increasing from 50.5% for the uncoated electrode to 87.8% with the coated electrode.

Beyond batteries, supercapacitors have also harnessed the potential of carbon-metal composites as coating materials to enhance capacitance. One study roughened the Al surface using laser ablation in a high vacuum, followed by coating with a C-Ni ( $\text{C}_{0.97}\text{Ni}_{0.03}$ ) composite using the pulsed laser deposition method.<sup>57</sup> This composite, combining the pure  $\text{sp}^2$  bonding state of carbon with the metallic phase of Ni, demonstrated performance improvement of supercapacitors. Surface roughening ameliorated interface resistance by providing a larger contact surface and improved mechanical anchorage for the active material layer. As shown in Figure 3G, the deposition method for the carbon-Ni composite used a KrF excimer laser, projecting a pulsed beam with a 25 ns pulse duration and 248 nm

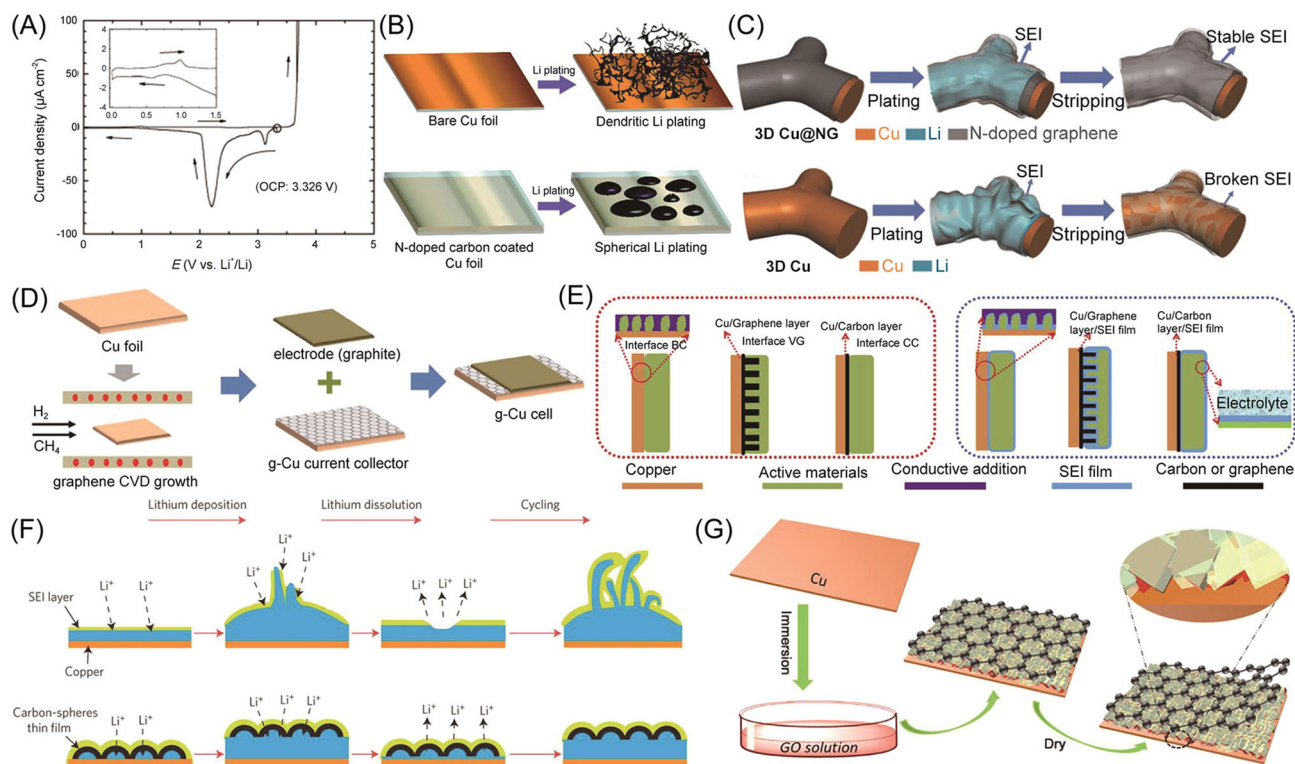
wavelength. This process took place within a state-of-the-art deposition chamber, focusing through a quartz window onto the Al foil surface. As for the cycle performance, as scan rates increased, the carbon-Ni composite consistently showed good performance, delivering  $28 \text{ F g}^{-1}$  at a current density of  $12.5 \text{ A g}^{-1}$  after 10,000 cycles. In comparison, the supercapacitor with bare Al yielded just  $21 \text{ F g}^{-1}$ , highlighting the enhanced capacitance achieved through the implementation of the carbon-metal compound.

Incorporating metals into carbon matrices for current collectors introduces additional costs, a factor significantly influenced by the type of metal used, with precious metals being more expensive than base metals. While the cost might be justified in high-end applications with substantial performance enhancements, the cost-benefit balance is crucial in consumer or large-scale uses. The production of a uniform carbon-metal compound coating on Al current collectors is a complex process. It involves steps like synthesizing the composite, ensuring an even distribution of the metal within the carbon and depositing the composite onto the Al. The challenge lies in establishing a robust bond between the composite and Al, while simultaneously maintaining the properties of the composite and preventing adverse reactions with other battery components. Despite the complexities, these composites can improve battery capacity, rate capability and cycle life, with the incorporated metal aiding in uniform current distribution.

## 4.2 | Cu-based current collectors

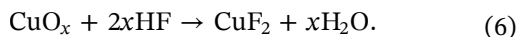
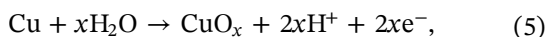
Cu is the predominant current collector for anode materials in commercial batteries, including the majority of commercial LIBs.<sup>94</sup> As the second most conductive metal, Cu boasts impressive properties: a resistivity of  $1.68 \times 10^{-8} \Omega \text{ m}$  at  $20^\circ\text{C}$ , a tensile strength of 325 MPa, a material density of  $8.96 \text{ g cm}^{-3}$  and a thermal conductivity of  $398 \text{ W m}^{-1} \text{ K}^{-1}$ .<sup>95</sup> Myung et al. reported the electrochemical behaviour of Cu within a  $\text{LiPF}_6$  electrolyte, as depicted in Figure 4A. Two distinct cathodic peaks emerge: the first, around 3.1 V versus  $\text{Li}^+/\text{Li}$ , is associated with the decomposition of  $\text{LiPF}_6$  to HF,<sup>98</sup> while the second, occurring between potentials of 1.5 and 3 V versus  $\text{Li}^+/\text{Li}$ , results from the surface oxidation of Cu, the formation of a solid electrolyte layer and the  $\text{Li}_2\text{O}$  formation process.<sup>51</sup> A closer inspection reveals subtle anodic and cathodic peaks at around 0.5 V versus  $\text{Li}^+/\text{Li}$ , indicating underpotential deposition of Li on the Cu surface and subsequent dissolution in the  $\text{LiPF}_6$ .<sup>96</sup> A notable anodic peak post 3.7 V versus  $\text{Li}^+/\text{Li}$  signifies the dissolution of Cu into the  $\text{LiPF}_6$  electrolyte, with no other discernible peaks evident below this threshold. Similar





**FIGURE 4** Carbon coatings for Cu-based current collectors. (A) Electrochemical behaviour within a 1 M LiPF<sub>6</sub> electrolyte, EC: Diethyl carbonate (DEC). Reproduced with permission: Copyright 2005, IOP Science.<sup>96</sup> (B) Li plating on the N-doped carbon-modified current collector. Reproduced with permission: Copyright 2019, Royal Society of Chemistry.<sup>97</sup> (C) Formation of SEI on a 3D porous Cu current collector with and without N-doped carbon. Reproduced with permission: Copyright 2018, Wiley.<sup>89</sup> (D) Growth of a graphene film on the Cu current collector using the CVD method. Reproduced with permission: Copyright 2018, Elsevier.<sup>43</sup> (E) Comparison of the performance of LIB using graphitic carbon coating (VG@CC) and amorphous carbon coating (AC@CC). Reproduced with permission: Copyright 2018, Elsevier.<sup>23</sup> (F) Example of a SEI layer presenting the notable differences when comparing cells with and without the carbon coating. Reproduced with permission: Copyright 2014, Nature.<sup>45</sup> (G) One-step redox strategy to produce a Cu current collector co-modified with rGO and Cu<sub>2</sub>O. Reproduced with permission: Copyright 2020, ACS Publications.<sup>47</sup>

dissolution processes with Cu are observed with other electrolytes like LiClO<sub>4</sub>.<sup>99</sup> Dai et al. elucidated the mechanisms accelerating the Cu dissolution, identifying a combination of heightened moisture levels and HF as accelerants.<sup>100</sup> The step-by-step progression of dissolution involves the oxidation of Cu reacting with water, followed by the resulting Cu oxide interacting with HF to yield CuF<sub>2</sub>. These results find validation in the immersion of a Cu foil in a LiPF<sub>6</sub> electrolyte with an ethylene carbonate (EC): dimethyl carbonate (DMC) (1: 1 vol%) solution as shown in Equations (5) and (6)



While the dissolution mechanisms of Cu suggest its unsuitability as a current collector for cathode materials, it remains a favoured substrate for anode materials. This preference stems from its stable electrochemical behaviour

under a potential of 3 V versus Li<sup>+</sup>/Li, signifying no alloying activity between Cu and Li. However, challenges emerge during overdischarging of the anode material, resulting in significant Cu dissolution. The re-formed Cu can then breach the separator, leading to critical issues such as short circuits and fires.<sup>101</sup> Additionally, using Cu (8.96 g cm<sup>-3</sup>) as the current collector, due to its higher mass density compared to Al, indicates that within the same energy storage device, Cu adds more weight, thereby reducing the overall energy density of the system. Nevertheless, this disadvantage comes with a caveat: thinner current collectors might compromise thermal stability, posing safety risks. To address these challenges, the introduction of carbon coatings is a viable solution. These coatings not only enhance the surface stability of Cu but also offer various benefits. They reduce both electrical and charge-transfer resistance and improve cycle stability.<sup>75,102</sup> A comprehensive summary of carbon-coated current collectors for both batteries and supercapacitors is presented in Table 3.

TABLE 3 Summary of Cu-based carbon-coated current collectors.

Type of device	Type of carbon coating	Electrode	Coated specific capacity (at C rate) (mAh g <sup>-1</sup> )	Original specific capacity (mAh g <sup>-1</sup> )	Cycle number	References
LIB	Graphite-like carbon	LTO (anode)	160.5 (1 C)	142.1	100	[103]
LIB	Conductive carbon	SiO <sub>x</sub> graphite	453.2 (1 C)	428.5	300	[104]
LIB	Nitrogen-doped carbon	Li	90% Coulombic efficiency (1 C)	40%	60	[97]
LIB	Graphene and graphite	LTO	133.86 (10 C)	106.15	2000	[105]
LIB	Rough carbon layer	Graphite	946.3 mAh (4 C)	887.9 mAh	400	[102]
LIB	Graphene	Graphite	453.3 (1 C)	340.5	50	[43]
LIB	Graphene	LTO	133 (2 C)	116.8	200	[42]
LIB	Graphene	Graphite	190 (3 C)	90	100	[23]
LIB	Graphene	Li	134.4 (0.5 C)	103.95	300	[106]
LIB	Nitrogen-doped graphene	Li	159.3 (0.5 C)	149.9	100	[89]
Li-Sulphur battery	Carbon nanofibres	Li	137.8 (1 C)	109.5	500	[107]
LIB	Graphene	Silicon	585.3 (1 C)	489.3	50	[108]
LIB	Carbon undercoating layer	Graphite	250 (1 C)	230	/	[74]
LIB	CH <sub>4</sub>	LTO	160 (3 C)	135 (3 C)	20	[75]
Supercapacitor	Nanocellular carbon	Ni(OH) <sub>2</sub>	86.5% (200 A g <sup>-1</sup> )	56.7%	10000	[109]
LIB	Carbon	Si	2000 (0.05 C)	750	100	[110]
LMB	MWCNT	Li	86.9 (0.5 C)	66.4	100	[111]
SIB	Copper-carbon composite	Na	270 (1 C)	230	100	[112]
LIB	3,4,9,10-PTCDA	Graphite	354.9 (0.2 C)	236.9	300	[113]
LMB	Hollow carbon sphere	Li	100% Coulombic efficiency (1 mA cm <sup>-2</sup> )	60% Coulombic efficiency (1 mA cm <sup>-2</sup> )	150	[45]
LMB	Nitrogen-doped graphene	Li	100% Coulombic efficiency (1 mA cm <sup>-2</sup> )	50% Coulombic efficiency	50	[114]
LIB	Amorphous carbon	LTO	157.11 (1 C)	105.03	400	[115]
Seawater battery	Graphene	Na metal	95% Coulombic efficiency (1 C)	50% Coulombic efficiency	100	[116]
LMB	Graphene	Li	100% Coulombic efficiency (1 mA cm <sup>-1</sup> )	90% Coulombic efficiency	120	[117]

TABLE 3 (Continued)

Type of device	Type of carbon coating	Electrode	Coated specific capacity (at C rate) (mAh g <sup>-1</sup> )	Original specific capacity (mAh g <sup>-1</sup> )	Cycle number	References
LMB	Crumpled graphene	Li	97.5% Coulombic efficiency (0.5 mA cm <sup>-1</sup> )	70% Coulombic efficiency	300	[118]
LIB	rGO	Graphite	870.3 (0.7 A g <sup>-1</sup> )	/	110	[119]
LIB	Graphdiyne	Silicon	1250 (1 A g <sup>-1</sup> )	700	200	[120]
LIB	CNT	LTO	90 (5 C)	50	100	[121]
LIB	CNF	Silicon	1100 (0.5 C)	250	100	[122]
LMB	N-doped graphdiyne	Li	99% Coulombic efficiency (0.5 mA cm <sup>-2</sup> )	75% Coulombic efficiency	50	[123]
Anode-free battery	Graphene, h-BN	Bare	93% Coulombic efficiency 0.5 mA cm <sup>-2</sup>	79% Coulombic efficiency	50	[124]
Anode-free battery	Ag, pD, graphene oxide	Bare	1.46 mAh cm <sup>-2</sup> (0.5 mA cm <sup>-2</sup> )	0.29 mAh cm <sup>-2</sup>	50	[125]
LMB	MWCNTs, Cu NWs	Si	2251 (0.2 A g <sup>-1</sup> )	1374	/	[48]
LMB	CNFs, Cu NWs	Li	124 (1 C)	80	400	[126]
LMB	Graphene, Au NPs	Li	220.8 (1 A g <sup>-1</sup> )	135.9	200	[127]
LMB	rGO, Cu <sub>2</sub> O	Li	140 (1 C)	95	200	[47]
LIB	N-doped rGO, NF	Sn-SnSb nanopowder	510 (0.1 A g <sup>-1</sup> )	10	100	[128]
LMB	N-doped carbon, ZnO	Li	140 (2 C)	50	175	[46]

Abbreviations: CNF, carbon nanofibre; LIB, lithium-ion battery; LMB, lithium metal battery; LTO, lithium titanate; MWCNT, multiwalled carbon nanotube; NW, nanowire; PTCDA, perylenetetracarboxylic dianhydride; rGO, reduced graphene oxide; SIB, sulphur-ion battery.

#### 4.2.1 | Doped carbon

Doped carbon has emerged as an effective strategy to reduce the nucleation overpotential of Li. This method enhances electrolyte wettability and introduces lithophilic sites on original carbon materials, offering a promising pathway for improved Li deposition. Doping has also been used as a technique for cathode material treatment including surface doping and bulk doping; the important strategy of doping (surface doping and bulk doping) has been considered an effective strategy to modulate the crystal lattice structure of cathode materials.<sup>129</sup> Typically, nitrogen and oxygen serve as the

primary doping materials, with nitrogen-doped carbon being more prevalent for Cu current collector coatings. N-doped carbon, characterized by its strong lithophilic properties, plays a crucial role in minimizing the energy barrier for Li deposition. Its composition includes graphitic, pyrrolic and pyridinic nitrogen. The presence of pyrrolic nitrogen introduces conjugated  $\pi$  bonds, while pyridine nitrogen groups possess a lone pair in the  $sp^2$ -hybridized orbital. These p-orbitals act as Lewis base sites, effectively adsorbing Lewis acidic Li ions in electrolytes via acid–base interactions, promoting a uniform distribution of metallic Li on the electrode surface.<sup>114</sup>

Polydopamine (pD), known for its diverse applications, is used to modify the Cu current collector. Zhang et al.<sup>97</sup> successfully produced N-doped carbon through a postcarbonization with the N-containing functional groups in pD. This polymerization and carbonization approach for pD provides both cost-effectiveness and scalability. Figure 4B illustrates the resulting Li plating on the N-doped carbon-modified current collector, effectively suppressing dendritic growth and showcasing spherical Li plating with a significantly reduced Li nucleation overpotential. Consequently, the Coulombic efficiency at 1 C notably increases, transitioning from 40% for the LIB with an uncoated Cu to 90% for the cell with N-doped carbon-coated Cu, sustained over 60 cycles.

Moreover, the CVD method, utilizing  $\text{NH}_3$  as the source, facilitates the application of a N-doped carbon coating.<sup>130</sup> Shi and colleagues<sup>89</sup> have augmented a 3D porous Cu current collector with N-doped carbon, as depicted in Figure 4C. This modification promotes even Li deposition and stable SEI formation, in contrast to the uncoated 3D Cu current collector, which shows a disrupted SEI layer after the Li-stripping process. As a result, the LIB with N-doped graphene demonstrates an enhanced performance of  $10 \text{ mA g}^{-1}$  at 0.5 C over 100 cycles.

Extending its applications beyond LIBs, doped carbon is integrated into lithium-sulphur batteries to improve their performance via the PECVD process.<sup>107</sup> The fabrication process involves depositing  $\text{NH}_3$ , nitrogen-heteroatoms and nitrogen- and oxygen-containing functional groups onto vertically aligned carbon nanofibres (CNFs). Among samples subjected to varying PECVD durations, the 1.5 h deposition sample outperforms the rest, showcasing a consistent cycling performance of  $137 \text{ mAh g}^{-1}$  for 500 cycles at 1 C.

Nitrogen- and oxygen-doped carbon improve their electronic and ionic conductivity, making them beneficial for supercapacitors and high-capacity battery anodes. This doping enhances capacitive properties and adhesion with electrode materials, resulting in a stable electrode-current collector interface. The introduction of heteroatoms increases active sites for charge storage or redox reactions, thereby boosting energy and power density. However, this process introduces additional steps and costs due to the requirement for specific raw materials, precursors or gases, and necessitates precise control during deposition. Despite these challenges, the performance gains, particularly in high-demand applications, can justify the investment. Achieving uniform dopant distribution and preserving the carbon structure's integrity are critical challenges, typically addressed through advanced synthesis techniques or postsynthesis treatments in controlled environments.

## 4.2.2 | Graphitic carbon

Graphitic carbon, as discussed in the previous section on Al foils, shows advantages in various applications as the substrate for anode material in batteries and supercapacitors when used as Cu current collectors. As shown in Figure 4D, Kim et al.<sup>43</sup> grew a graphene film on the Cu current collector using the CVD method, which increased the adhesion of the anode layer to afford a more hydrophobic Cu current collector surface. The uniform formation of Li metal nuclei on the current collector, facilitated by the graphene film contributed to reduced polarization and a significant decrease in the cell's charge-transfer resistance.

Liu et al.<sup>23</sup> applied a similar PECVD method to coat vertical graphene nanowalls and enhance the rate performance of the LIB using a graphite anode. The study compared LIB performance using graphitic carbon coating (VG@CC) and amorphous carbon coating (AC@CC) via various analysis techniques as shown in Figure 4E. After 100 cycles at 3 C with a graphitic anode cell, AC@CC showed an interfacial electron transfer resistance 11 times larger than that of VG@CC. Analysis of the SEI layer by x-ray photoelectron spectroscopy indicated that the oxygen-containing functional groups in AC@CC formed a thick SEI layer made of  $\text{LiCO}_3$  and  $\text{Li}_2\text{O}$  after several cycles, leading to increased interfacial resistance. In contrast, VG@CC showed no C=O peaks and a lower ratio of the LiF peak than AC@CC, which suggested the generation of a thinner SEI layer with high conductivity. This confirmed the higher conductivity and better electrochemical performance of graphitic carbon compared to amorphous carbon. Beyond these applications, graphene as a carbon coating material for Cu current collectors, composed of a Cu foil and foam based on graphite, LTO and a Li metal anode, stands out as a widely adopted choice for energy storage devices.<sup>43,106,117</sup>

As mentioned in the previous section, CNTs and CNFs are widely used as graphitic carbon coating materials to improve the performance of the Cu current collector. Current collectors with cross-stacked super-aligned CNTs, coated via electron beam deposition, have been applied to Cu substrates due to their good wettability, strong adhesion and low contact resistance.<sup>121</sup> The cell shows a superior rate capability with  $90 \text{ mAh g}^{-1}$  at 5 C for 100 cycles, whereas the cell with a conventional Cu current collector only showed  $50 \text{ mAh g}^{-1}$  at this stage. In applications involving CNFs, Shimizu et al.<sup>122</sup> used an electrodeposited vapour growth carbon fibre (VGCF) on a Cu current collector. The rough surface of the VGCF provides a high surface area and efficient Li-ion conduction, mitigating volume expansion of the VGCF@Cu/Si electrode during long-term cycling, thereby showing



minimal cracks compared to cells with a pristine Cu current collector.

In addition to conventional graphitic carbon, rGO and GDY also serve as coating materials for Cu current collectors. For instance, rGO has been used in LIBs to achieve binder-free integration via a one-step electro-reduction reaction.<sup>119</sup> The resulting rGO nanosheets, firmly attached to the Cu surface, display impressive Li-storage capabilities and conductivity, achieving a stable cyclability of 870.3 mAh g<sup>-1</sup> after 110 cycles at 0.7 A g<sup>-1</sup>. Similarly, GDY has been incorporated into Cu surfaces to address the significant volume expansion challenges of Si anodes during alloying processes.<sup>120</sup> The method for growing ultrathin GDY nanosheets creates 3D all-carbon conductive networks, enhancing Cu substrate and Si anode interfacial contact. This approach effectively addresses the disintegration challenges of Si anodes within the conductive network, boosting LIB capacity from 700 to 1250 mAh g<sup>-1</sup> over 200 cycles compared to uncoated Cu.

The coating on the Cu current collector offers advantages, primarily enhancing interfacial electrical conductivity for more efficient electron transport within the battery. Specifically for Cu, graphitic carbon can serve as a protective layer to guard against corrosion and oxidation, particularly in high-voltage conditions or in harsh environments. Similar to Al current collectors, the incorporation of the graphitic carbon coating involves additional steps that may increase the overall cost. Therefore, the cost-effectiveness needs to be evaluated in relation to potential performance benefits such as increased battery performance and efficiency. Addressing processing challenges, CVD is the most popular method for graphitic carbon coating. The adhesion of the carbon layer to the Cu substrate is crucial to ensure long-term stability and requires careful optimization of the coating process.

#### 4.2.3 | Amorphous carbon

Amorphous carbon plays an irreplaceable role in mitigating challenges related to Li deposition, particularly serving as a mould for Li plating. Cui et al. utilized hollow carbon nanospheres (HCNs) to enhance the performance of cells with Cu current collectors.<sup>45</sup> Carbon was coated on a polystyrene nanosphere layer to form HCN using flash evaporation. As described in Figure 4F, cells with the carbon coating showed a distinctive SEI layer on the HCN surface, effectively reducing the influx of Li ions into compromised SEI layers. In contrast, cells without the carbon coating showed a concentrated flow of Li ions around the crack region of

the SEI layer, increasing the potential risk of dendritic growth. The gap between HCN and the current collector expanded during Li plating; it reverted to its original structure during Li stripping, prompting further research into structural modification of coating materials for improved Li-ion flow management. Zhu et al.<sup>115</sup> also used amorphous carbon to enhance Cu current collectors by the magnetron sputtering technique. The carbon film notably improved the bond between the current collector and the active material, thereby reducing the internal resistance of the battery. Specifically, cells with the carbon film showed an interface resistance of 14.53  $\Omega$  between LTO and the current collector, whereas cells without the carbon film showed a resistance of 21.80  $\Omega$ . This enhancement is attributed to the rough carbon surface, which facilitates the movement of Li<sup>+</sup> ions and electrons.

In conclusion, the key advantage of amorphous carbon lies in its exceptional corrosion resistance, a critical attribute for Cu in electrolyte environments where degradation or oxidation may occur. This protective coating enhances the durability and longevity of the current collector. Moreover, the unique structure of amorphous carbon can accommodate the expansion and contraction of the electrode materials during charge and discharge cycles, potentially mitigating mechanical stresses and improving the overall stability of the battery. However, depositing a uniform and stable layer of amorphous carbon on Cu poses technical challenges. Techniques such as CVD or sputtering must be carefully controlled to achieve the desired properties of the carbon layer, including thickness, uniformity and adhesion to the Cu substrate.

#### 4.2.4 | Carbon-metal compounds

As previously emphasized, carbon coating materials offer key advantages such as enhanced interfacial electrical conductivity, corrosion resistance and improved adhesion between the active material and the current collector. Additionally, metals on the current collector can provide lithophilic sites.

Zhou et al.<sup>46</sup> showcased the modification of Cu foam using Nitrogen-doped carbon/ZnO, which was achieved by coating ZIF-8 on 3D Cu foam, followed by calcination to yield an Nitrogen-doped carbon/ZnO-modified Cu foam. Both ZnO- and Nitrogen-doped carbon served as lithophilic sites, leading to dendrite-free, flower-like Li metal deposition on the Cu foam, in contrast to the uneven plating and dendritic growth on uncoated Cu. ZnO has also been used in the fabrication of high-performance CC@ZnO@Li composite anodes,

developed through the meticulous atomic layer deposition of ZnO, followed by the infusion of molten Li. The chemical conversion of ZnO results in the in situ formation of a LiZn/Li<sub>2</sub>O configuration, which facilitates oriented Li deposition at lithiophilic Li<sub>2</sub>O sites. This configuration promotes more uniform Li deposition and ensures efficient charge transfer. Consequently, the fabricated CC@ZnO@Li anodes have shown superior cycling stability and rate performance, both in symmetric and in full-cell configurations.<sup>130</sup> Chen et al.<sup>47</sup> introduced a one-step redox strategy to co-modify a Cu current collector with rGO and Cu<sub>2</sub>O (designated rGO-Cu<sub>2</sub>O/Cu). This modification promotes uniform Li-ion nucleation and suppresses Li dendrite formation. The process involves immersing a Cu foil in a GO solution for several hours, inducing an in situ redox reaction between GO and Cu, leading to the formation of rGO and Cu<sub>2</sub>O, as illustrated in Figure 4G. The carbon composite-coated Cu foil demonstrated an enhanced capacity of 140 mAh g<sup>-1</sup> at 1 C, compared to the 90 mAh g<sup>-1</sup> capacity of pristine Cu. Zhao et al. studied a 3D porous MWCNT-Cu nanowire (Cu NWs)-coated Cu current collector using the spin-coating method and Si anode.<sup>48</sup> The structure, comprising intertwined Cu nanomaterials, allowed for straight Cu NWs to intertwine with MWCNTs and Si deposition via the PECVD method. This combination improved interfacial electrical conductivity and overall structural stability, harnessing the high conductivity of Cu NWs and the robustness of MWCNTs.

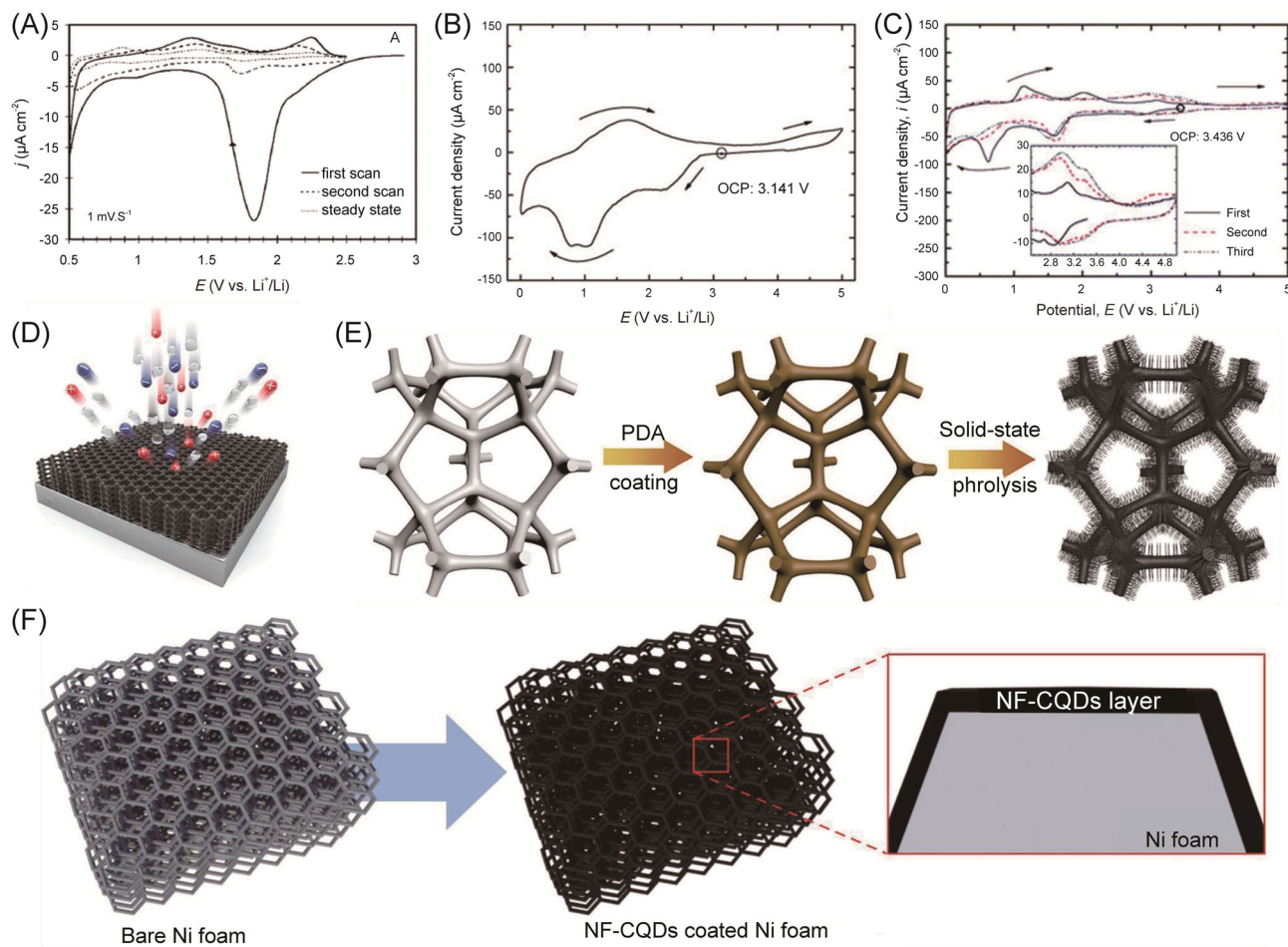
As mentioned in the previous section, carbon-metal compound coatings applied to Cu current collectors combine the conductive and protective properties of carbon with the unique attributes of the chosen metal. This combination significantly enhances the interfacial electrical conductivity and mechanical strength of the current collector. However, the complex process of synthesizing and applying a homogeneous carbon-metal composite layer contributes to higher processing costs. These increased expenses must be justified by corresponding improvements in battery performance or lifespan. In mass production, the potential economies of scale may help mitigate these additional costs. The challenge lies in achieving a consistent and high-quality carbon-metal compound coating on Cu, encompassing not only the deposition of the composite material but also ensuring a good distribution of metal particles within the carbon matrix and strong adhesion to the Cu substrate. Ensuring the long-term stability and integrity of the coating under operational conditions is another critical consideration.

### 4.3 | Other metal-based current collectors

In addition to the commonly used Al cathode current collector and Cu anode current collector, other metals including Ni, Ti and stainless steel also serve as current collectors for carbon coating in batteries and supercapacitors.

Ni, with an electrical resistivity of  $6.93 \times 10^{-8} \Omega \text{ m}$  at 20°C, is particularly noteworthy. Figure 5A illustrates the previously investigated electrochemical performance of Ni in 1 M LiPF<sub>6</sub> in EC:DMC. Among various current collectors, Ni shows the highest tensile strength at 730 MPa and a density of 8.9 g cm<sup>-3</sup>.<sup>131</sup> On the anodic side, the peak is attributed to the dissolution of Li deposited on the Ni surface and the SEI decomposition. Conversely, on the cathode side, a significant peak emerges around a voltage of 1.85 V versus Li<sup>+</sup>/Li, attributed to SEI formation on the Ni surface and Ni oxide reduction reaction. The subsequent drop indicates the underpotential deposition of Li on the Ni surface.<sup>135</sup> The entire anodic potential region demonstrates stable electrochemical behaviour, with the current density consistently below 5  $\mu\text{A cm}^{-2}$ . This makes Ni a suitable choice for an anode material current collector. However, the potential of Ni as cathode material current collectors is debated. While Geoffroy et al. argue that the electrochemical behaviour of Ni remains stable in LiPF<sub>6</sub>- and LiClO<sub>4</sub>-based electrolytes (both in EC: Diglyme (DG) at 1:1 vol%) within a potential range of 3–5.5 V vs Li<sup>+</sup>/Li,<sup>136</sup> other researchers counter that Ni corrosion occurs at a potential of 3.6 V versus Li<sup>+</sup>/Li when the electrolyte is LiPF<sub>6</sub> in EC/DMC/DEC (1:1:1 vol%).<sup>137</sup> As a result, Ni has not been widely adopted as a cathode current collector. Nevertheless, by introducing a carbon coating to the surface of Ni, its application range as a current collector can be expanded, simultaneously reducing internal and charge-transfer resistance and corrosion resistivity.

Ti possesses an electrical resistivity of  $3.9 \times 10^{-7} \Omega \text{ m}$  at 20°C, a material density of 4.51 g cm<sup>-3</sup> and a tensile strength of 360 MPa. Myung et al. measured Ti electrochemical behaviour in a 1 M LiPF<sub>6</sub> electrolyte in EC:DMC (1:1 vol%).<sup>51</sup> As illustrated in Figure 5B, the initial anodic peak is observed between 1 and 2 V versus Li<sup>+</sup>/Li, resulting from the oxidation reaction involving Ti and Li. As discussed in the Al section, a secondary peak in the anodic region stems from the formation of a dual passivation layer, comprising bottom TiO<sub>2</sub> and top TiF<sub>4</sub>. Similar to the Al substrate behaviour, this passivation film acts as a protective layer, enhancing corrosion resistance during high cell potential.<sup>99</sup>



**FIGURE 5** Carbon coatings for Ni, Ti and stainless-steel-based current collectors. (A) Electrochemical performance of Ni in 1 M  $\text{LiPF}_6$ , EC:DMC. Reproduced with permission: Copyright 2005, ACS Publications.<sup>131</sup> (B) Electrochemical performance of Ti in 1 M  $\text{LiPF}_6$ , EC:DMC. Reproduced with permission: Copyright 2011, Royal Society of Chemistry.<sup>51</sup> (C) Electrochemical performance of stainless steel in 1 M  $\text{LiPF}_6$ , EC:DMC. Reproduced with permission: Copyright 1994, IOP Science.<sup>96</sup> (D) Graphene used as a protective layer on Ni current collectors using the CVD method. Reproduced with permission: Copyright 2022, ACS Publications.<sup>132</sup> (E) Nitrogen-doped CNT hierarchical scaffold on Ni foam through in situ growth. Reproduced with permission: Copyright 2020, Elsevier.<sup>133</sup> (F) N and F-Co-doped CQDs on a Ni foam substrate as a current collector for Li-air batteries. Reproduced with permission: Copyright 2023, Hindawi.<sup>134</sup>

Among the previously mentioned metal current collectors, stainless steel has the highest electrical conductivity of  $7.2 \times 10^{-7} \Omega \text{ m}$  at  $20^\circ\text{C}$ , a material density of  $7.9 \text{ g cm}^{-3}$  and a tensile strength of 454 MPa. Comprised primarily of Fe, Cr, Ni and Mn, stainless steel is notable for its exceptional corrosion resistance, attributed to the formation of a Cr oxide passivation film on its surface.<sup>96,138</sup> Figure 5C illustrates the electrochemical behaviour of stainless steel in a 1 M  $\text{LiPF}_6$  electrolyte mixed with EC/DMC in a 1:1 ratio. An initial peak emerges at around 1.8 V versus  $\text{Li}^+/\text{Li}$ , indicating the reduction of air-formed Fe and/or Cr oxide films, the formation of SEI and the creation of the electrolytic salt  $\text{Li}_2\text{O}$ . A subsequent cathodic peak appears at around 0.6 V versus  $\text{Li}^+/\text{Li}$ , attributed to the under-potential deposition of Li on stainless steel. Notably, as the potential approaches 0 V versus  $\text{Li}^+/\text{Li}$ , no alloying process

occurs between stainless steel and Li. In the anodic zone, an initial peak emerges at around 1 V versus  $\text{Li}^+/\text{Li}$ , representing the oxidation of deposited Li. Another peak emerges at 2 V versus  $\text{Li}^+/\text{Li}$ , related to the oxidation of Cr. A smaller anodic peak at around 3 V versus  $\text{Li}^+/\text{Li}$  likely results from the oxidation of Fe and  $\text{Cr}^{3+}$ . The subsequent voltage increases without a corresponding current rise indicate the formation of a passivation film. While this passivation layer can provide protection in certain scenarios, it may be insufficient when paired with  $\text{LiCoO}_2$  or  $\text{LiMn}_2\text{O}_4$  cathodes or specific electrolytes. Use of carbon coatings could be a viable strategy to enhance corrosion resistance, thereby improving cell performance.<sup>139,140</sup> Table 4 below summarizes batteries and supercapacitors using carbon-coated metal current collectors other than Al and Cu.

TABLE 4 Summary of Ni, Ti and stainless-steel-based carbon-coated current collectors.

Current collector	Type of device	Type of carbon coating	Electrode	Coated specific capacity (at C rate) (mAh g <sup>-1</sup> )	Original specific capacity (mAh g <sup>-1</sup> )	Cycle number	References
Ni	Lithium-sulphur battery (Ni)	Carbon fibre	Sulphur (cathode)	397 (0.5 C)	194	50	[141]
Ni	LMB (Ni)	Sp <sup>2</sup> -hybridized carbon	Li metal (anode)	87.4 (4 C)	60	500	[142]
Ni	Lithium-air battery (Ni)	NF-CQDs	Li <sub>2</sub> O <sub>2</sub> (cathode)	5121.91	2917.97	1	[134]
Ni	LJB (Ni)	Graphene	AC (cathode)	0.7 W m <sup>-2</sup>	0.4 W m <sup>-2</sup>	/	[132]
Ni	LIB	Graphene	Graphite (anode)	510 (0.4 A cm <sup>-2</sup> )	380	100	[143]
Ni	LMB	CNT	Li (anode)	127 (2 C)	76	400	[133]
Ni	Supercapacitor	Carbon cloth	MnO <sub>x</sub>	816 F g <sup>-1</sup> (100 mV s <sup>-1</sup> )	415.25	/	[144]
Stainless steel	Aqueous rechargeable batteries	Carbon	LMO	90.1 mAh g <sup>-1</sup> (2 C)	60 mAh g <sup>-1</sup>	800	[139]
Stainless steel	LIB	CNT ink	Silicon (free standing)	1711 mAh g <sup>-1</sup> (0.1 C)	/	50	[145]
Stainless steel	Supercapacitor	Nitrogen-doped carbon	PANI	642 mF cm <sup>-2</sup> (0.05 mA cm <sup>-2</sup> )	376 mF cm <sup>-2</sup>	/	[146]
Stainless steel	LIB	Maleic acid	V <sub>2</sub> O <sub>5</sub>	696 mAh g <sup>-1</sup> (100 mA g <sup>-1</sup> )	550 mA g <sup>-1</sup>	100	[147]
Stainless steel	Aqueous batteries	Carbon black, polyethylene	LMO	74.29 (0.5 C)	32.59	100	[148]
Ti	Supercapacitor (Ti)	FWCNT	AC (cathode)	132 F g <sup>-1</sup>	74 F g <sup>-1</sup>	/	[149]
ZnO	LMB (ZnO)	Graphene-nested porous carbon	Li (anode)	129.5 (0.5 C)	78.6	50	[37]

Abbreviations: AC, activated carbon; CNT, carbon nanotube; CQD, carbon quantum dot; FWCNT, few-wall carbon nanotubes; LIB, lithium-ion battery; LMB, lithium metal battery; LMO, lithium metal battery; LMO, lithium-rich manganese-based oxide; NF, nanofibre; PANI, polyaniline; PANI, Polyaniline.



### 4.3.1 | Graphitic carbon

As highlighted in the previous section, multilayered graphene stands out for its exceptional interfacial electrical conductivity and corrosion resistance. It has been utilized as a protective layer on Ni current collectors via the CVD method, as depicted in Figure 5D.<sup>132</sup> This graphene layer not only promotes corrosion resistance but also facilitates efficient charge percolation due to its high interfacial electrical conductivity. Chu et al.<sup>143</sup> applied a graphite layer on Ni-Fe foam by the CVD method as well, resulting in both enhanced lithophilicity and increased corrosion resistance. SEM images reveal notable differences: the uncoated Ni-Fe foam shows evident corrosion with a loose grain boundary, whereas the graphite@Ni-Fe shows a dense structure with large grains and numerous pinholes on particle surfaces.

Beyond graphene and graphite, CNTs have been harnessed for Ni, stainless-steel and Ti current collectors to enhance the performance of batteries and supercapacitors.<sup>133,145,149</sup> As illustrated in Figure 5E, a Nitrogen-doped CNT hierarchical scaffold was integrated onto Ni foam through in situ growth. The as-prepared current collector showed no cracks, dendrite-free morphology and resisted dead Li deposition. Its electrochemical performance was commendable, demonstrating low overpotential and maintaining an average Coulombic efficiency of 98.2% over 200 cycles at  $1 \text{ mA cm}^{-2}$  during charge and discharge processes. Cui et al. introduced a distinctive anode structure that eliminated the need for heavy-metal current collectors. By weaving a flexible, conductive CNT network into a Si anode, they achieved a free-standing structure reminiscent of steel bar-reinforced concrete. In this configuration, the CNT network served a dual function as both a mechanical support and an electrical conductor, with Si serving as a high-capacity anode material for LIB. This structure demonstrated low areal resistance, an impressive specific capacity ( $\sim 2000 \text{ mAh g}^{-1}$ ) and commendable cyclability. For supercapacitors, few-wall CNTs were coated on Ti current collectors to enhance capacitance and cost-efficiency. This is achieved through a straightforward three-step process involving mixing, dispersion and filtration, with activated carbons acting as the cathode material. The supercapacitor shows an increase in capacitance from 3.9 to  $107 \text{ F g}^{-1}$  at a practical voltage scan rate of  $100 \text{ mV s}^{-1}$ , associated with improved interfacial electrical conductivity.

### 4.3.2 | Amorphous carbon

Amorphous carbon has been applied to stainless steel, primarily through the slurry coating process. Wen et al.<sup>139</sup> utilized a conductive slurry, comprising carbon

black, polyacrylic acid sodium salt and carboxymethyl cellulose, to treat stainless steel for use in aqueous LIB cathodes. The initiation of oxygen evolution at high potentials was reduced with the introduction of the carbon coating. Moreover, batteries equipped with a carbon-coated stainless-steel mesh outperformed their counterparts using an uncoated stainless-steel mesh in terms of both cyclic performance and rate capability. A carbon cloth was integrated into a nanoporous Ni architecture in supercapacitors, achieving a significant increase in specific capacitance. While the pristine current collector showed a specific capacitance of  $353.2 \text{ mF cm}^{-2}$ , the carbon-coated variant achieved an impressive specific capacitance of  $906.6 \text{ mF cm}^{-2}$  at  $1 \text{ mA cm}^{-2}$ .<sup>144</sup>

### 4.3.3 | Doped carbon

Ma et al.<sup>134</sup> utilized nitrogen and F-Co-doped carbon quantum dots (CQDs) on a Ni foam substrate as a current collector for Li-air batteries, achieved through ultrasonic spray-coating as shown in Figure 5F. The CQDs were hydrothermally synthesized using citric acid as the carbon source and urea as the nitrogen source. During heating, citric acid reacted with urea to form citric acid amide, while sodium fluoride served as the fluorine source, integrating F atoms into the carbon's honeycomb structure. The NF-CQD/Ni-based cathode showed superior electrochemical attributes, including enhanced cycle stability and substantial discharge capacity. This was attributed to the NF-CQD coating, which prevented Ni foam oxidation and enhanced both the electrical conductivity and the electrocatalytic activity of the cathode. A similar technique was used for a stainless-steel current collector-based supercapacitor by Cao et al.<sup>146</sup> The nitrogen-doped carbon-coated stainless-steel mesh was fabricated through a straightforward hydrothermal carbonization process using polyaniline (PANI). The carbon layer, derived from glucose and dicyandiamide, significantly improved the bond between the stainless-steel mesh and PANI. As a result, the tailored supercapacitor showed a substantial specific capacitance of  $624 \text{ mF cm}^{-2}$  at  $0.05 \text{ mA cm}^{-2}$ , nearly doubling the performance of the unmodified current collector.

Carbon coating on Ni current collectors improves corrosion resistance, especially in aggressive electrolytes, and enhances conductivity. This synergistic combination leverages the inherent properties of Ni with the stability and conductivity of carbon contributing to the overall performance of high-energy batteries. For Ti, known for its corrosion resistance and mechanical strength but

insufficient internal electrical conductivity, the application of carbon coating boosts its efficiency in battery systems. For stainless steel, carbon coating increases interfacial electrical conductivity while providing protection against corrosion, which is crucial when operated with corrosive electrolytes or at high temperatures. The cost impact of carbon coating varies; it is moderate for Ni and stainless steel due to well-established coating processes, but more substantial for the less common and relatively expensive Ti. The selection of graphitic carbon coating depends on the balance between performance enhancement and cost. Various techniques such as CVD or physical vapour deposition are used for the coating process. While it is relatively easier to coat Ni and stainless steel with carbon, Ti requires careful interface management due to its reactivity. Optimizing the coating process is essential to achieve uniform conductive coatings that preserve the substrate's inherent properties. In summary, carbon coatings substantially improve the performance of Ni, Ti and stainless-steel current collectors in batteries. However, the decision to use carbon coating should carefully balance the improved functionality with the additional costs and processing challenges, tailored to meet the specific requirements of the application.

## 5 | NONMETAL-BASED CURRENT COLLECTORS

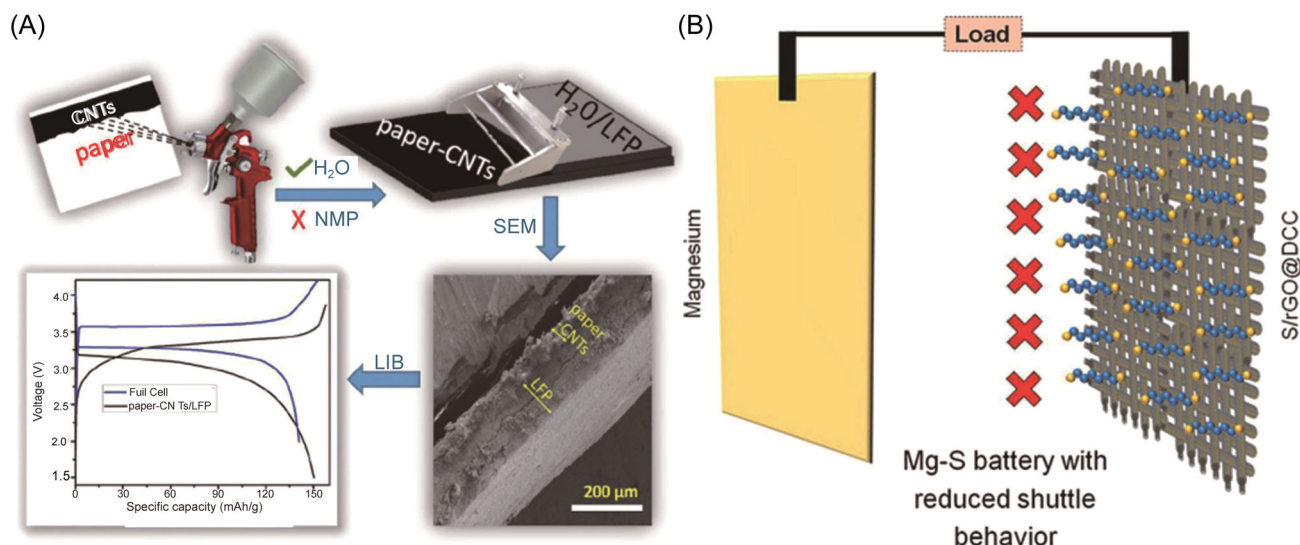
### 5.1 | Carbon-coated nonmetal-based current collectors

Beyond metal-based current collectors, carbonaceous materials can also function as substrates for carbon coatings, enhancing the efficiency of batteries and supercapacitors. Carbonaceous current collectors offer advantages such as being lightweight and enabling a higher active-to-nonactive material ratio at electrodes. These are due to their porous structure, which allows for increased mass loading of active material compared to metal foils.<sup>150,151</sup> For example, carbon fibre paper has an electrical resistivity of  $8 \times 10^{-4} \Omega \text{ m}$  and a tensile strength below 5 MPa. Despite having the highest resistivity and the lowest tensile strength among current collectors, its flexible nature makes it an excellent choice for pliable current collectors. It also boasts an ultra-low density of  $0.44 \text{ g cm}^{-3}$ , significantly enhancing the energy density of devices. Unlike metal-based collectors that undergo plastic deformation, carbonaceous materials can be folded multiple times, rendering them ideal for future flexible LIBs.<sup>152</sup> A list of carbon-coated nonmetal current collectors is provided below.

Unlike metal-based counterparts, there is limited online information available about nonmetal or carbonaceous current collectors. However, each example possesses distinctive features. In Figure 6A, Ventrapragada et al.<sup>153</sup> introduced a method to coat commercial cellulose-based paper with CNTs for LIBs using a surfactant-free spray-coating process. The advantages of paper as a current collector include cost-effectiveness, ready availability, biodegradability, seamless integration into manufacturing lines and enhanced safety compared to conventional counterparts. While paper is not intrinsically conductive, the use of CNTs ensures a consistently conductive coating. Compared to an Al-based current collector for LFP cathodes, the LFP with the paper-CNT cathode showed a 17% improvement in areal capacity, maintaining stability over 450 cycles at  $150 \text{ mA g}^{-1}$  (Table 5).

For supercapacitors, He et al.<sup>155</sup> used CNT foam chemically vapour-infiltrated with pyrolytic carbon. This current collector is lightweight, highly conductive and compressible, making it suitable for rGO electrodes. The CNT-based supercapacitor showed superior rGO utilization, reduced charge-transfer resistance and enhanced electrolyte diffusion compared to vinyl film collectors. With a knee frequency of 1.3 Hz, it outperformed vinyl-based ones (0.4 Hz), achieving a balance of  $14.1 \text{ Wh kg}^{-1}$  at  $3100 \text{ W kg}^{-1}$ . In Mg-S batteries, carbon-coated carbonaceous current collectors using nitrogen-doped and sulphuric acid-treated carbon cloth effectively addressed polysulphide issues (Figure 6B). The doped variant increased battery capacity from 10 to  $388 \text{ mAh g}^{-1}$  at  $20 \text{ mA g}^{-1}$ , enhancing longevity and stability.<sup>154</sup>

In conclusion, carbon coatings on nonmetal current collectors, like carbonaceous materials or papers, primarily enhance interfacial electrical conductivity, which is a critical factor often lacking in these substrates. This coating significantly improves charge-transfer efficiency, making these lightweight and potentially more flexible substrates viable for battery applications. Additionally, carbon coatings provide chemical and thermal stability to nonmetal substrates, increasing their durability under various operating conditions. The strategic pairing of substrate material with a carbon coating opens doors for innovative battery designs, especially in weight-sensitive and flexible applications such as wearable electronics or flexible devices. While nonmetal substrates may offer cost advantages, the processing required for carbon coating can contribute to the overall cost. Long-term cost reduction could occur with economies of scale and advancements in coating technologies. Applying carbon coatings to nonmetal substrates involves challenges distinct from those encountered with metal substrates, which includes ensuring good adhesion of the carbon



**FIGURE 6** (A) Coating of commercial cellulose-based paper with CNTs using a surfactant-free spray-coating process. Reproduced with permission: Copyright 2019, De Gruyter.<sup>153</sup> (B) Nitrogen-doped and sulphuric acid-treated carbon cloth serving as a reservoir to anchor polysulphides within the cathode scaffold. Reproduced with permission: Copyright 2017, Elsevier.<sup>154</sup>

layer, maintaining substrate integrity and properties during coating process and achieving coating uniformity and conductivity. The complexity varies based on the substrate material. Some polymers or composites might undergo deformation or degradation under typical coating conditions, necessitating the development of specialized, low-temperature coating processes.

## 5.2 | Research progress of conductive polymer-based current collectors

Among the materials utilized for the fabrication of LIBs current collectors, electrically conducting polymers like polypyrrole (PPy), PANI and polythiophene (PTH) are popular choices, potentially enabling the free-standing and flexible formation of batteries because of their superior mechanical robustness, excellent thermal and chemical resistance, significant mass density, ease of fabrication, affordability and versatility.<sup>156–159</sup> Apart from being used as substrate materials, these polymers also find applications as binders and active materials, contributing to environmental sustainability by minimizing waste. However, the widespread adoption of these conductive polymers is hampered by several drawbacks, including their propensity for self-discharge, limited conductivity, and suboptimal energy density and rate capability.<sup>160–162</sup> Additionally, the efficiency of these polymers is constrained by the restricted rate of ion transfer across thick polymer membranes and a limited degree of doping.<sup>163,164</sup> The complexity of postsynthesis processing, arising from issues such as infusibility, low solubility and brittleness,

further complicates their use.<sup>164</sup> Therefore, to satisfy the next-generation application on LIBs, improvement of the electrochemical and mechanical performances of customized polymer material is needed.

PPy is recognized as one of the most extensively researched and promising conductive polymers, characterized by its low monomeric mass, exceptional mechanical strength, notable electrical conductivity, adequate biocompatibility and a significant degree of doping.<sup>165,166</sup> The versatility of PPy is further underscored by its compatibility with both aqueous and nonaqueous solvents, facilitated through chemical or electrochemical polymerization processes. This adaptability allows PPy to be used across a broad spectrum of current collectors, making it an ideal candidate for conducting substrates or active materials in flexible electrodes for LIBs.<sup>167</sup> Recent advancements have highlighted the synergistic integration of PPy with carbon materials, exemplifying this through the development of a highly flexible fibre-shaped battery system. This system integrates a PPy/PF<sub>6</sub> cathode with an SWCNT/poly (vinylidene difluoride) hollow anode.<sup>168</sup> While the incorporation of PPy into composite electrodes mitigates structural collapse during cycling or deformation and enhances conductivity and capacity, challenges such as high irreversible capacity and suboptimal rate performance persist. Addressing these issues is crucial for advancing the performance of PPy-based composite electrodes.

PANI is distinguished as another conductive polymer notable for its appropriate conductivity, significant flexibility, affordability, straightforward synthesis, commendable environmental compatibility and distinctive

TABLE 5 Summary of nonmetal-based carbon-coated current collectors.

Type of device	Type of carbon coating	Electrode	Coated specific capacity (at C rate) (mAh g <sup>-1</sup> )	Original specific capacity (mAh g <sup>-1</sup> )	Cycle number	References
LIB (regular copy paper)	CNT	LFP	90 (1 C)	74.7	450	[153]
Supercapacitor (CNT foam)	Pyrolytic carbon	Graphene oxide	14.1 Wh kg <sup>-1</sup> 3100 W kg <sup>-1</sup>	5 Wh kg <sup>-1</sup> 1200 W kg <sup>-1</sup>	/	[155]
LIB (microfibre paper)	CNT	V <sub>2</sub> O <sub>5</sub> /graphene	283 (1 C)	196	100	[200]
Supercapacitor (CNT fibre)	Nitrogen-doped CNT	LTO	100% capacity retention (15 C)	79% capacity retention	1000	[201]
Mg-S battery (carbon cloth)	N-, <i>s</i> -doped nitric acid, sulphuric acid	LTO	388 (20 mA g <sup>-1</sup> )	10	40	[154]

Abbreviations: CNT, carbon nanotube; LFP, lithium iron phosphate; LIB, lithium-ion battery; LTO, lithium titanate.

redox characteristics. These attributes position PANI as a promising contender for use as a polymer substrate and material for free-standing electrodes. Typically, PANI serves as a flexible matrix, which, when combined with various organic or inorganic materials, shows enhanced electrochemical performance due to a synergistic effect.<sup>169</sup> An example of such innovation involves the creation of free-standing hybrid aerogels, comprising interwoven SWCNTs and PANI nanoribbons. These aerogels are utilized as flexible, free-standing substrates and anodes for LIBs.<sup>170</sup>

PTH, characterized by its high conductivity (approximately 500 S cm<sup>-1</sup>), exemplifies another conductive polymer with remarkable mechanical flexibility, commendable thermal stability and favourable chemical resilience. These properties have facilitated its adoption as both a substrate and active material for cathode and anode components in energy storage devices.<sup>171</sup> The strategic integration of CNTs, PEDOT:PSS, PTH and Si as building blocks enhances the composite's overall performance.<sup>172</sup>

Owing to good electrical conductivity, favourable theoretical capacity, excellent chemical stability and flexible frameworks capable of mitigating the volumetric expansion and subsequent pulverization of active materials<sup>173</sup> and also their environmentally friendly nature and recyclability, these polymers further underscore their potential as promising materials in LIB applications. However, several challenges hinder their widespread utilization as current collectors and free-standing active materials. For example, their relatively low ionic conductivity, which hampers ion transfer across the material, impeding optimal performance,<sup>174</sup> and poor postsynthesis handleability, such as limited solubility, infusibility and mechanical fragility, necessitate complex processing techniques. These issues, coupled with difficulties in achieving the desired degrees of doping, restrict the overall effectiveness of conductive polymers in LIBs.<sup>175</sup> Hence, dedicated research efforts are crucial to overcoming these obstacles, aiming to harness the full potential of conductive polymer-based high-performance current collectors for next-generation LIBs.

## 6 | MANUFACTURING TECHNIQUES OF CARBON-COATED CURRENT COLLECTORS

Various manufacturing techniques have been applied to coat carbon materials onto current collectors, such as CVD and slurry casting, offering distinct advantages. Specifically, CVD is a process for producing high-quality, solid and thin films, which includes atmospheric-pressure



CVD, low-pressure CVD and PECVD.<sup>22,23,42</sup> In the CVD coating process, substrates are exposed to volatile precursors, which decompose or react on the substrate surface, forming a layer of the desired composition.<sup>176</sup> The initial application of CVD for carbon coating was reported by Somani et al. in 2006, demonstrating the deposition of few-layer graphene on nickel sheets.<sup>177</sup> In addition to CVD, casting methods, including blade casting, drop casting and solution casting, are used for carbon coating. Zhang et al. reported a composite of VGCF (90 wt%), polyethylene oxide (5 wt%) and carboxymethyl cellulose (5 wt%), demonstrating remarkable flexibility, adhesion and mechanical strength, with a VGCF layer that possesses a uniformly nanoporous structure with nanoscale pores.<sup>65</sup> Similarly, solution-cast crumpled graphene balls on Cu current collectors showed uniformity and mechanical robustness, with a highly smooth surface at the single-particle level.<sup>118</sup>

Other notable methods include magnetron sputtering, which involves depositing carbon-based materials onto current collectors in a high-pressure environment within a magnetron sputtering device. This technique preserves the integrity of the Cu foil, achieving uniform coverage and substantial contact surface area.<sup>103</sup> Electroplating VGCF on Cu surfaces, as conducted by Shimizu et al., produced a VGCF layer characterized by high internal electrical conductivity and mechanical strength.<sup>122</sup> Jia et al.<sup>126</sup> prepared CNFs via electrospinning, creating an advanced 3D current collector with a conductive network acting as a porous equipotential body, promoting uniform Li deposition. A comprehensive summary of the coating method for carbon-coated current collectors of batteries and supercapacitors is presented in Table 6.

## 7 | INDUSTRIAL CARBON-COATED CURRENT COLLECTOR

In the contemporary automotive industry, carbon-coated current collectors are indispensable for advancing light-weight and high-energy-density LIBs in EVs. The carbon coating ensures uniform current distribution across the collector surface, preserving electrode materials during extensive charge–discharge cycles. This is essential in mitigating the detrimental effects of localized current densities, preventing uneven electrode degradation and maintaining optimal battery performance. Transitioning to supercapacitors, crucial for swift energy delivery in applications such as power grid stabilization and backup power systems, carbon-coated current collectors provide an expanded surface area. This facilitates expedited

electron transfer processes, advantageous in hybrid vehicles with stop-start technology, ensuring immediate energy surges for engine restarts. The carbon coating acts as a protective barrier enhancing the longevity of these devices in demanding industrial settings, particularly in supercapacitors vital for reliable, low-maintenance energy management systems.

In the dynamic landscape of the global carbon-coated Al foil market, valued at approximately \$277.3 million in 2021 and projected to reach \$2998.09 million by 2031, that is, a compound annual growth rate of 26.9%,<sup>178</sup> a discernible shift is underway. Carbon-coated Al foils are emerging as favoured cathode substrates in battery technology, boasting superior properties compared to conventional Al foils. This trend is set to propel a significant surge in demand for these advanced materials, contributing to overall market growth.

In terms of specifications, the majority of carbon-coated current collectors use Al foil, optimizing adhesion with the active material and enhancing corrosion resistance. Featuring conductive carbon, with some variants incorporating carbon, polymer and graphene, these collectors maintain a coating thickness of around 1  $\mu\text{m}$ , with thinner coatings less common. The reduced thickness minimizes impacts on the overall energy density of batteries and supercapacitors, ensuring that weight increase is negligible. Manufacturers cater to diverse applications, with some focusing on specific active materials like graphite or LFP, while others offer products compatible with a broad range of cathode materials, including lithium NMC, lithium nickel manganese oxide and anode materials such as Si and LTO. Despite the advancements, cost remains a key barrier to the widespread adoption of carbon-coated current collectors. Urgent development of cost-effective methods for fabricating thinner carbon coatings is crucial to fully realize their potential benefits. Table 7 summarizes the company list of commercialized carbon-coated current collectors.

## 8 | CONCLUSION

Current collectors play a crucial role in facilitating the transfer of electrons from the active material to the external circuit, with their weight significantly influencing the overall energy density of energy storage devices. While Al and Cu are common choices for current collectors in cathode and anode materials, the potential for enhancing battery and supercapacitor performance through carbon coatings remains. This review systematically examines various types of carbon-coated current collectors, focusing on their roles in improving energy

TABLE 6 Summary of coating methods for carbon-coated current collectors.

Coating method	Applied current collector	Coating thickness ( $\mu\text{m}$ )	Carbon coating material	Electrode material	Charge-transfer resistance reduced ( $\Omega$ )	Cycling performance increased	References
Magnetron sputtering	Al	2	GLC	LCO	25	39.5 mAh g <sup>-1</sup> (1 C)	[103]
CVD	Cu	22	Graphene	Graphite	/	50 mAh g <sup>-1</sup> (0.2 C)	[22]
PECVD	Cu	1	Vertical graphene nanowalls	Graphite	35.6	130 mAh g <sup>-1</sup> (3 C)	[23]
LPCVD	Cu	90	Graphene	LTO	99.13	16.2 mAh g <sup>-1</sup> (2 C)	[42]
Spreading	Al	5	Super P and layered graphite	Sulphur/carbon composite	70.4	95 mAh g <sup>-1</sup> (0.2 C)	[44]
Blade casting	Al	100	VGCF layer	Sulphur	/	450 mAh g <sup>-1</sup> (50 mA g <sup>-1</sup> )	[65]
Drop casting	Cu	40	Graphene oxide	Li	120	97% for 200 cycles (1 mA cm <sup>-2</sup> )	[117]
Solution casting	Cu	8	Reduced graphene oxide	Li	/	37.5% CE after 300 cycles	[118]
Electrobeam deposition	Cu	1	SACNT	LTO	/	16.5 mAh g <sup>-1</sup> (5 C)	[121]
Buffing process	Al	0.2	GLC	LNMO	20	100 mAh g <sup>-1</sup> (1 C)	[54]
Electroplating	Cu	1	MWCNT	Silicon	19	300 mAh g <sup>-1</sup> (1 C)	[122]
Spin-coating	Cu	1	MWCNTs, Cu NWs	Li	/	1074 mAh g <sup>-1</sup> (0.42 A g <sup>-1</sup> )	[48]
Slurry coating	Stainless steel	/	Carbon black	LMO	42	27 mAh g <sup>-1</sup> (10 C)	[139]
Electrospinning	Cu	70	CNFs, Cu NPs	Li	97	17 mAh g <sup>-1</sup> (1 C)	[126]
Immersing and drying	Cu	/	rGO, Cu <sub>2</sub> O	Li	45	35 mAh g <sup>-1</sup> (1 C)	[47]

Abbreviations: CNF, carbon nanofibre; CVD, chemical vapour deposition; GLC, graphene-like carbon; LCO, LiCoO<sub>2</sub>; LMO, lithium-rich manganese-based oxide; LNMO, lithium nickel manganese oxide; LPCVD, low-pressure CVD; LTO, lithium titanate; MWCNT, multiwalled carbon nanotubes; NP, nanoparticle; NW, nanowire; PECVD, plasma-enhanced chemical vapour deposition; rGO, reduced graphene oxide; SACNT, super-aligned carbon nanotube; VGCF, vapour growth carbon fibre.

storage device performance. The following summarizes how carbon coatings address/mitigate challenges during the operation of batteries and supercapacitors.

1. *Prevent cathodic corrosion*: For cathodic current collectors like Al foil, Al foam and etched Al, carbon coatings effectively address the issue of pitting corrosion caused by trace amounts of H<sub>2</sub>O or organic radical cations, thereby stabilizing battery cycling.
2. *Avoid cracks and regulate ion transport*: In anodic current collectors like Cu foil, Cu foam and etched Cu, carbon coatings mitigate crack formation, counter acidic conditions contributing to corrosion products and simultaneously reduce charge-transfer resistance. These coatings also regulate ion plating and enhance cyclability.
3. *Enhance charge transfer*: Carbon coatings enhance current transfer at high C rates by increasing the contact area with the active material due to their rough surface. In supercapacitors, they reduce charge-transfer resistance, resulting in higher power density.
4. *Enhance adhesion*: Carbon coatings improve adhesion between the active material and the current collector,

TABLE 7 Summary of industrialized carbon-coated current collectors.

Company name	Product name	Content	Thickness ( $\mu\text{m}$ )	Current collector	Type of device	Application with Active material	Country	References
ARMOR battery films	En'Safe	Carbon and polymer	0.5–1 $\mu\text{m}$	Al, Cu	LIB, EDLC	LFP, LMFP, rich nickel NMC, LNMNO, LTO, silicon, graphite	France	[179]
Jinxi new material	Nano-modified carbon-coated aluminium foil	Conductive carbon	0.3 $\mu\text{m}$	Al	LJB	LFP	China	[180]
MSE supplies	Single-side conductive carbon-coated copper foil	Conductive carbon	1 $\mu\text{m}$	Cu	LJB	Graphite	USA	[181]
TOB New Energy	Carbon-coated aluminium foil for lithium battery cathode	Conductive carbon	1 $\mu\text{m}$	Al	LJB	LFP, LTO	China	[182]
Toyal group	Toyo carbo	Carbon particles	20–30 nm	Al	LJC and supercapacitor	Carbon	Japan	[183]
Kokuen group	Primer-coated foil	Carbon particles	1 $\mu\text{m}$	Al	LJB	LFP, NMC	Japan	[184]
Shenzhen YuQiang	Carbon-coated Al and Cu for LIB	Conductive carbon	0.1–1 $\mu\text{m}$	Al	LJB	LFP, NMC, graphite, silicon	China	[185]
Jiangsu Dingsheng	Carbon-coated Al foil	Conductive carbon	0.1–1 $\mu\text{m}$	Al	LJB	LFP, NMC	China	[186]
Blueglownano	Conductive primer-coated Al and Cu foil	Conductive carbon	Less than 1 $\mu\text{m}$	Al, Cu	LJB, EDLC	LFP, NMC, NiC, Li, SiC, SiO, anode free, LTO, graphite	China	[187]
Dunmore	Conductive coated aluminium	Carbon black	/	Al	LJB	/	Germany	[188]
Shanghai metal	Carbon-coated aluminium foil	CNT	2 $\mu\text{m}$	Al	LJB	LFP, NMC	China	[189]
Landt	Cathode current collector for lithium-ion batteries	Nanosized conductive graphite and carbon particles	1 $\mu\text{m}$	Al	LJB	LFP, NMC, LTO	Germany	[190]
Cambridge Energy Solution	Carbon-coated aluminium foil	Conductive carbon	1 $\mu\text{m}$	Al	LJB	LFP, NMC, LNMNO	UK	[191]
Stanford Advanced Materials	AL4180 conductive carbon-coated aluminium foil	Conductive carbon	3 $\mu\text{m}$	Al	LJB	LFP, NMC, LNMNO	USA	[192]

(Continues)

TABLE 7 (Continued)

Company name	Product name	Content	Thickness ( $\mu\text{m}$ )	Current collector	Type of device	Application with Active material	Country	References
TMAX battery equipment	Conductive carbon-coated aluminium foil for lithium-ion battery	Conductive carbon	2–3 $\mu\text{m}$	Al, Cu	LIB	LFP, NMC, LNMNO	China	[193]
Nsfoil	Graphene-coated aluminium foil	Graphene	0.5 $\mu\text{m}$	Al	LIB, supercapacitors	LFP, NMC, AC	China	[194]
GYC group	Graphene-coated aluminium foil	Graphene	0.5 $\mu\text{m}$	Al	LIB, supercapacitors	LFP, NMC, LNMNO, AC	China	[195]
Primet	Carbon-coated aluminium foil	Conductive carbon	1 $\mu\text{m}$	Al	LIB	LFP, NMC, LNMNO, LCO	USA	[196]

Abbreviations: AC, activated carbon; EDLC, electric double layer capacitor; LFP, lithium iron phosphate; LIB, lithium-ion battery; LNMNO, lithium nickel manganese oxide; LTO, lithium titanate; NMC, Ni manganese cobalt oxide.

especially beneficial in anode materials with large theoretical capacities like Si and Ge.

However, the challenges associated with carbon coatings include (a) *Cost*: Techniques like CVD used for carbon coating can be expensive. There is a need for innovative cost-effective methods to improve the overall affordability of carbon coating. (b) *Volume sacrifice*: Certain 3D structured carbons, like MWCN or carbon nanowalls, may occupy significant space, potentially reducing energy density. Balancing carbon coating thickness with its protective and conductive roles is crucial to address this challenge effectively.

In the competitive energy storage market, from wearable devices to EVs, the key objective is to develop storage solutions with higher energy and power densities while maintaining cost efficiency. This necessitates the crucial need for conductive, lightweight and cost-effective current collectors. Therefore, carbon coatings must meet the criteria of being lightweight, uniformly distributed and economically viable. Despite their considerable potential, research on carbon coatings remains underrepresented, highlighting the necessity for more comprehensive studies.

## 9 | OUTLOOK AND FUTURE PROSPECTS

Looking to the future of battery technology, the focus turns to carbon-coated ultra-lightweight current collectors. Aligned with the ongoing trend towards sustainable, lightweight and flexible power sources, these current collectors could significantly impact the industry. Especially with some newly developed electrode materials such as  $\text{Li}_5\text{Cr}_7\text{Ti}_6\text{O}_{25}$  and  $\text{CoNiO}_2$  materials as the anode material for LIBs and  $\text{NiO@NiMoO}_4\text{@PPy}$  for pseudocapacitor active material with relatively low conductivity compared to traditional electrode material, the application for the surface coating could greatly impact the performance.<sup>197–199</sup> The synergy between lightweight, flexible polymer and conductive carbon coatings presents an appealing combination for next-generation electronics, including wearable devices, bendable smartphones and foldable screens. Figure 7 illustrates a shift from the traditional design and material selection to the future for coatings of the current collectors, advocating for a departure from conventional methodologies towards the exploration of novel materials. As mentioned in Section 5, the advancement of carbon-reinforced conductive polymers embodies the integration of high conductivity and low resistance of carbon coatings with the lightweight and flexible nature of polymers. This



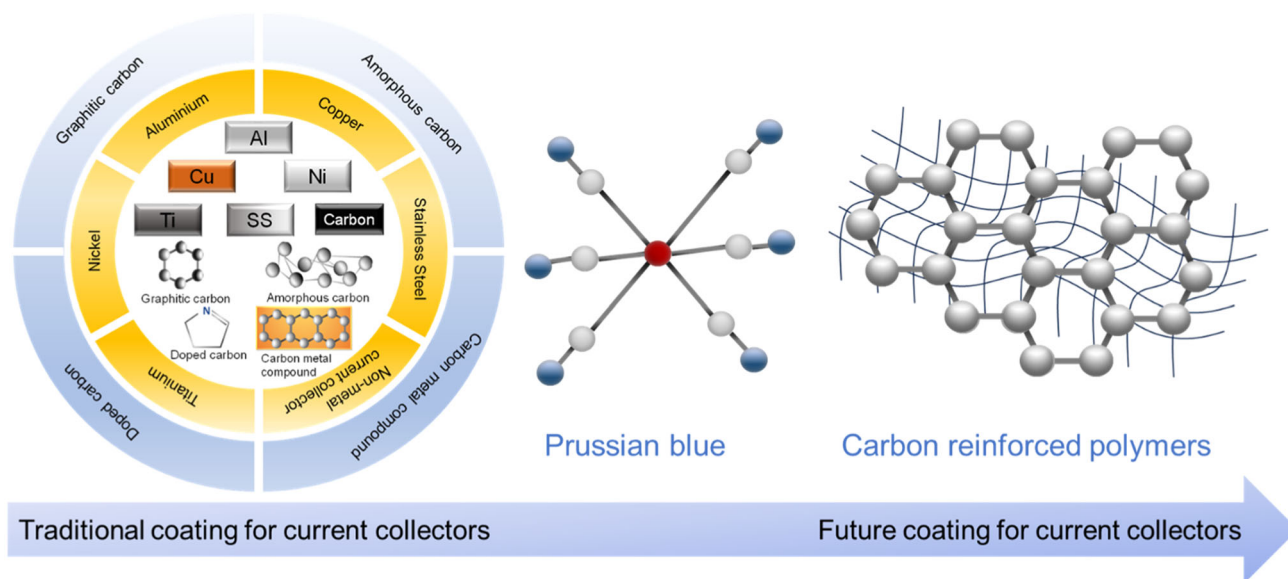


FIGURE 7 Current and future directions of material-coated current collectors.

amalgamation harnesses the strengths of both components, optimizing the overall performance of current collectors. As mentioned before, the application of Prussian blue has been recognized for its effectiveness in addressing surface issues of the current collector, such as oxidation. Although Prussian blue has a moderate level of conductivity, it is significantly lower than that of metallic conductors. This can limit the efficiency of electron transfer within the battery, reducing its overall performance. To address these limitations, ongoing research efforts are critically focused on modifying Prussian blue through doping, creating composites and optimizing the microstructure to enhance its stability and conductivity. Therefore, it is imperative that future research endeavours concentrate on the innovation of surface coatings. This entails the adoption of new carbon-based, lightweight materials that can further enhance the durability, efficiency and functionality of battery systems, ultimately contributing to the development of next-generation energy storage solutions.

The development trajectory of carbon-coated current collectors is expected to align with advancements in nanotechnology and material science. Among various substrate materials, polymers show promise, with the key objective being the enhancement of interfacial electrical conductivity. Carbon-coated polymer current collectors, incorporating innovations such as graphene or CNT coatings, have the potential to further enhance conductivity and mechanical strength while maintaining the inherent flexibility of polymers. In response to escalating environmental concerns driving the quest for greener and more sustainable battery components, carbon-coated

polymers derived from renewable resources stand out as a compelling alternative to traditional metal-based current collectors. Anticipated advancements in processing techniques, particularly the emergence of low-temperature, cost-effective coating methods, could make the production of these carbon-coated polymers more feasible on a commercial scale. Moreover, the integration of these current collectors with novel electrode materials and electrolytes holds the promise of yielding batteries with unprecedented energy densities, charging speeds and life cycles. However, challenges such as ensuring long-term stability, optimizing the interface between the carbon coating and polymer substrate and mass manufacturing scalability need to be addressed. Research and development efforts in these areas will be crucial for transitioning from experimental prototypes to mainstream applications. In conclusion, the potential of carbon-coated current collectors aligns with the broader trends in technology and sustainability, ushering in an era of lightweight, flexible and high-performance batteries poised to revolutionize how we power our devices and our daily lives.

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### CONFLICT OF INTEREST STATEMENT

The authors declare no conflicts of interests.

### DATA AVAILABILITY STATEMENT

Data will be made available on request.

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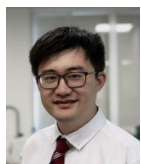
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