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# *Nanocarbon Coatings for Graphite Anodes in Lithium-Ion Batteries: Recent Advances in Electrochemical Performance*

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***Abstract.*** Graphite is widely used as an anode material in lithium-ion batteries. Nanostructured carbon coatings can modify its surface properties without changing the bulk structure. They help control reactions between graphite and the electrolyte. This review examines the preparation and electrochemical performance of carbon coatings derived from pitch, resins, sugars, and other polymers. Pitch-derived coatings influence particle packing. Lower charge-transfer resistance has been measured for resin-derived nanoscale shells in single-particle tests. Saccharide and polymer coatings have produced both gains and losses in rate capability. Studies also report different lithium-storage performance at similar coating loadings. These differences are associated with coating uniformity, pore structure, surface chemistry, and electrode compaction. Suitable coatings may reduce interfacial charge-transfer resistance and improve rate capability or cycling stability. The added carbon surface can also increase irreversible lithium consumption. This may lower the initial Coulombic efficiency. High-rate delithiation alone does not establish fast-charging capability. Tests at practical electrode densities and areal loadings are needed to assess whether these improvements persist after compaction.

***Keywords:*** Lithium-ion batteries, graphite anodes, nanocarbon coatings

## 1. Introduction

Graphite is widely used in commercial lithium-ion battery anodes because of its low lithium intercalation potential, good cycling stability and relatively low cost. Lithium is stored reversibly between graphite layers, with fully lithiated $LiC_6$ corresponding to a theoretical specific capacity of 372 $mAhg^{-1}$ . Electrochemical measurements and X-ray diffraction studies have shown that lithium-graphite intercalation compounds form through successive stages and that the graphite structure is restored during delithiation [1].

Surface reactions and charging kinetics can limit this performance. During initial lithiation, electrolyte decomposition forms a passivating solid electrolyte interphase (SEI), consuming lithium irreversibly. For graphite and petroleum coke electrodes tested in PC/EC-based electrolytes, Fong et al. found that the irreversible capacity associated with initial film formation increased with the specific surface area of the carbon material [2]. Charging at low temperature introduces the possibility of lithium plating. Using voltage relaxation measurements and in situ neutron diffraction,

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von Lüders et al. found that the estimated extent of lithium plating increased with charging rate in commercial graphite-based batteries at −2 °C [3]. An effective surface coating needs to limit irreversible lithium consumption while allowing lithium ions to enter the graphite during charging.

Carbon coating modifies the graphite-electrolyte interface. Yoshio et al. reported an initial Coulombic efficiency of 94.5% for natural graphite with a 17.6 $wt\%$ carbon coating, compared with 87.2% for uncoated graphite, in 1 M $LiPF_6$ dissolved in EC/DMC (1:2 by volume). First-cycle irreversible capacity decreased from 50.3 to 19.6 $mAhg^{-1}$ [4]. The reported carbon content, however, does not specify the coating thickness or establish that the layer was nanoscale.

Nanoscale carbon coatings modify the surface while retaining micrometer-scale graphite particles. Mukhan et al. obtained a relatively uniform phenolic-resin-derived carbon layer approximately 4-5 nm thick. Single-particle measurements showed that the area-normalized charge-transfer resistance decreased from 81–101 to 49-67 Ω $cm^2$ after coating, supporting improved interfacial kinetics. However, initial Coulombic efficiency in composite electrodes decreased from 92% to 85%. A nanometer-scale layer can therefore improve charge transfer while introducing additional irreversible losses; its porosity and surface chemistry remain relevant to coating design [5].

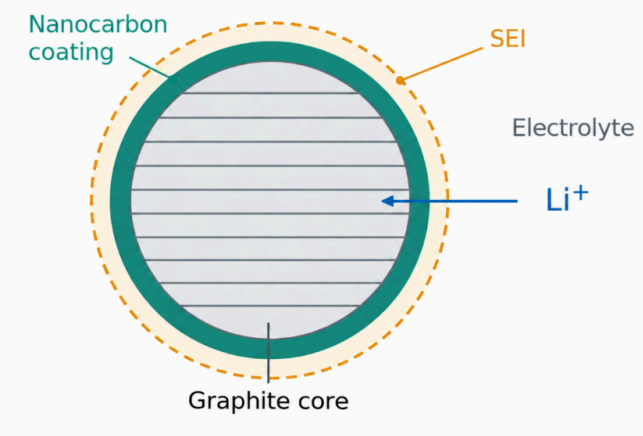



Figure 1. Precursor categories, coating structure. Original schematic based on [5-7], not to scale

## 2. Pitch-derived carbon coatings

### 2.1. Electrochemical performance and mechanisms

Pitch-derived carbon coatings can modify the electrode-electrolyte interface. Kim et al. reported an increase in initial Coulombic efficiency from 80.49% to 84.07% with the addition of 5 wt% petroleum pitch. The differential capacity peaks related to electrolyte decomposition became smaller during the first cycle. This suggests that fewer irreversible side reactions occurred. At a delithiation rate of 5C, coated graphite retained 80.58% of its capacity measured at 0.2C. Uncoated graphite retained only 9.07%. The authors linked this difference to lithium-ion transport channels in the disordered carbon layer. The coating also increased particle sphericity and reduced preferred orientation during electrode compaction (calendering). Lithiation, however, was performed at 0.1C throughout the tests. The reported improvement therefore concerns high-rate delithiation rather than fast-charging performance [6].

Li et al. examined a thinner carbon coating. Transmission electron microscopy (TEM) showed a dense amorphous carbon layer approximately 10 nm thick on graphitized needle coke. The coating

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was prepared using pitch derived from ethylene tar. After 100 cycles at 100 mA $g^{-1}$, the coated material delivered a specific discharge capacity of 347.5 mAh $g^{-1}$. The uncoated material reached 275.1 mAh $g^{-1}$ under the same conditions. The continuous carbon layer was reported to limit solvent co-intercalation and buffer volume changes. These results indicate improved cycling performance with a nanoscale coating. However, the relatively low-test current does not establish fast-charging capability [8].

Electrode compaction also affects performance. Kwon et al. used coal-tar and petroleum pitches to coat both the outer surfaces and internal graphite sheets of spherical natural graphite. The resulting electrode had a density of 1.60 g $cm^{-3}$. Its initial reversible capacity was 360.3 mAh $g^{-1}$, with an initial Coulombic efficiency of 93.0%. It outperformed artificial graphite at 1.55 g $cm^{-3}$ in separate rate tests for lithiation from 0.2 to 3C and delithiation from 0.2 to 10C. Impedance measurements showed lower charge-transfer resistance. The authors also suggested that the pore structure-maintained ion transport after calendaring. However, the artificial graphite used for comparison was itself carbon-coated. The performance difference therefore reflects changes in the graphite substrate and compacted electrode structure as well as the coating [9].

## 2.2. Coating modification and structural control

Air oxidation changes the composition and thermal behavior of pitch, which affect the carbon layer produced during carbonization. Guo et al. treated pitch at 310 °C with an air flow of 120 L $h^{-1}$. Increasing the oxidation time from 10 to 12 h raised the coking value from 70.19% to 71.85%, but also increased the quinoline-insoluble fraction from 0.97 to 2.55 wt%. The authors regarded this increase in insoluble material as undesirable for graphite coating, suggesting a trade-off between carbon yield and coating suitability [10].

Wang et al. examined pitches with softening points of 170, 220 and 250 °C, keeping the pitch-to-graphite mass ratio at 1:10. The coated graphite prepared using the 250 °C pitch had a BET specific surface area of 4.11 $m^2$ $g^{-1}$, compared with 10.77 $m^2$ $g^{-1}$ for the 170 °C sample. At 0.1C, their initial Coulombic efficiencies were 88.04% and 80.46%, respectively, versus 84.70% for uncoated graphite. Micropore formation during the release of light components on carbonization was proposed to explain the poorer initial efficiency of the 170 °C sample. Since coking value and composition also varied, these differences cannot be attributed to softening point alone [11].

Solvent choice affects coating morphology at a fixed precursor loading. Jo and Lee used n-hexane, toluene, tetrahydrofuran (THF), and quinoline as solvents to process mixtures with a pitch-to-graphite mass ratio of 1:9. Transmission electron microscopy (TEM) observations revealed that the resulting coatings had thicknesses of approximately 200, 500, 150, and 300 nm, respectively; the sample processed with THF exhibited a smoother and more uniform surface. Given that coating thickness and uniformity tend to vary in tandem within these submicron-scale layers, it is not possible to simply designate 150 nm as the optimal thickness in a general sense [12].

Dry-mixing methods can reduce coating thickness to the nanometer scale. Youn and Lee mixed graphite with 10 wt% pitch (softening point: 150 °C) at a rotational speed of 9000 rpm. TEM measurements showed that the average coating thickness decreased from approximately 150 nm to 50 nm. The authors suggested that repeated collisions between particles rendered the coating more compact, thereby reducing its thickness [13].

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### 2.3. Preparation methods

Pitch-derived carbon coatings can be prepared by dry processing or solvent-assisted deposition, followed by carbonization. Dry routes distribute pitch through mechanical mixing and thermal softening, whereas solvent-assisted routes disperse the precursor in a liquid medium before solvent removal.

Kim et al. mixed natural flake graphite with petroleum pitch for 30 min and hot-pressed the mixture at 315 °C for 30 min, above the pitch softening point of 255 °C. Block formation was intended to limit pitch redistribution during heating. The blocks were carbonized at 900 °C for 1 h, then ground and classified into 10–25 μm particles [6]. Youn and Lee instead used a high-speed dry mixer, followed by heat treatment under argon at 500 and 1000 °C for 90 min at each temperature [13].

For solvent-assisted coating, Jo and Lee ultrasonicated petroleum pitch in a selected solvent for 10 min, then added artificial graphite at a pitch-to-graphite mass ratio of 1:9. Following a further 30 min of ultrasonication and 24 h of stirring, the solvent was evaporated at 80 °C. Carbonization proceeded under argon at 800 °C for 30 min and 1000 °C for 10 min [12].

Precursor fractionation can be incorporated before deposition. Huang et al. produced pitch from fluid catalytic cracking (FCC) slurry oil through air oxidation and subsequent thermal polycondensation under nitrogen. They removed the quinoline-insoluble material by Soxhlet extraction, then mixed the soluble fraction with natural graphite in tetrahydrofuran at a pitch-to-graphite mass ratio of 1:10. After solvent recovery at 60 °C, the powder was carbonized at 1000 °C for 2 h [14].

Dry mixing avoids the solvent recovery and drying steps needed for solution-based processing. In either route, the initial pitch loading differs from the final carbon content because some precursor mass is lost during carbonization.

## 3. Resin-derived carbon coatings

### 3.1. Electrochemical performance and mechanisms

Carbon derived from phenolic resin can form a disordered nanoscale shell on graphite, improving lithium-ion transfer across the interface. The coating may also introduce sites for irreversible reactions, so faster interfacial kinetics can come at the expense of initial Coulombic efficiency.

Mukhan et al. produced a relatively uniform carbon layer with a thickness of about 4–5 nm. Single-particle pulse-polarization measurements were performed at 293 K. The area-normalized charge-transfer resistance fell from 81–101 to 49–67 Ω $cm^2$ after coating. Meanwhile, the exchange current density rose from 0.25–0.32 to 0.38–0.52 mA $cm^{-2}$. Testing individual particles reduces the influence of binders and composite-electrode architecture. The measured changes therefore provide evidence of improved interfacial kinetics. The authors linked this improvement to the disordered carbon layer, which may assist lithium-ion transport toward graphite edge sites [5].

The improved interfacial kinetics did not translate into a higher initial Coulombic efficiency. In composite-electrode tests at 0.1C, the initial Coulombic efficiency dropped from 92% to 85% [5]. The additional irreversible loss was attributed to a larger specific surface area and oxygen-containing functional groups.

Gao et al. oxidized natural graphite with sulfuric acid and then applied a phenolic resin-derived carbon coating. At an initial resin content of 9 wt%, an amorphous carbon shell about 20 nm thick was obtained. The modified graphite delivered 361.6 mAh $g^{-1}$ after 40 cycles at 30 mA $g^{-1}$. By

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comparison, untreated graphite showed a specific capacity of 332.3 mAh $g^{-1}$. The carbon shell was proposed to act as a buffer against graphite exfoliation caused by solvent co-intercalation. However, the oxidation treatment was carried out before coating. Its contribution to the capacity improvement cannot be separated from that of the carbon shell based on these results. The modified graphite thus showed a higher capacity after cycling under low-current conditions [15].

### 3.2. Coating modification and structural control

He et al. studied natural microcrystalline graphite coated with resin-derived carbon at coating contents between 4% and 16%. Scanning electron microscopy (SEM) images showed that a 4% coating made the particle edges less sharp and filled some gaps between surface crystallites. However, the specific surface area increased as more carbon was added. At 16% coating content, it reached 117.7 $m^2$ $g^{-1}$, compared with 18.3 $m^2$ $g^{-1}$ for uncoated graphite. The authors attributed this increase to open micropores formed as volatile substances escaped during carbonization. At 0.1C, the initial Coulombic efficiency increased from 79.7% to 81.4% with a 4% coating. When the coating content reached 16%, it fell to 70.2%. Higher coating content therefore resulted in greater irreversible losses during the first cycle [16].

Mukhan et al. reported a similar increase in surface area despite using a much thinner carbon layer. The coating was approximately 4–5 nm thick. The BET specific surface area increased from 4.27 $m^2$ $g^{-1}$ for uncoated graphite to 47.67 $m^2$ $g^{-1}$ after coating. X-ray photoelectron spectroscopy (XPS) also showed a higher proportion of oxygen-containing species on the coated surface. A uniform nanoscale coating therefore does not necessarily make the graphite surface more "inert" (i.e., less reactive) [5].

### 3.3. Preparation methods

Resin-derived carbon coatings are commonly prepared by applying a resin solution to graphite. The coated material is then dried and carbonized.

He et al. dissolved phenolic resin in ethanol and mixed it with natural microcrystalline graphite to obtain a paste. The paste was cured at 160°C and then cooled. After passing through a 325-mesh sieve, the material was carbonized at 800°C for 2 hours under oxygen-free conditions [16].

Mukhan et al. prepared the coating using acetone as the solvent. They dissolved 2 g of phenolic resin in 30 mL of acetone and mixed it with 3 g of natural graphite. The mixture was stirred for 5 hours at room temperature. The solvent was then removed, followed by drying at 110°C for 2 hours. Carbonization took place under an argon atmosphere. The temperature was increased to 1000°C at a heating rate of 3°C/min and held for 1 hour. A carbon layer approximately 4–5 nm thick was obtained [5].

## 4. Saccharide- and other polymer-derived carbon coatings

### 4.1. Electrochemical performance and mechanisms

Coatings derived from saccharides and polymers can lower initial Coulombic efficiency and rate capability. Sharova et al. examined sucrose- and glucose-coated graphite in half-cells containing 1 M $LiPF_6$ in EC/DMC with 2 wt% fluoroethylene carbonate. The sucrose- and glucose-derived samples had initial Coulombic efficiencies of 80.8% and 83.0%, respectively, versus 91.2% for pristine graphite. Their capacities at 3C delithiation were also lower: 112 and 180 mAh $g^{-1}$,

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compared with 211 mAh $g^{-1}$ for uncoated graphite. Capacity was calculated using graphite mass alone, excluding the coating. Lithiation was carried out at 0.5C with a constant-voltage step [7].

The authors attributed the lower initial efficiencies to the larger surface area available for electrolyte decomposition during SEI formation. However, surface area did not fully explain the rate performance. The poly(vinyl alcohol) (PVA)-derived sample had a lower specific surface area than pristine graphite but delivered only 116 mAh $g^{-1}$ at 3C [7]. Reducing surface area was therefore not enough to improve high-rate delithiation in these tests.

Different results were reported by Wang et al. for a PVA-derived coating prepared at a PVA-to-natural-graphite mass ratio of 1:5. The coated graphite delivered an average capacity about 24% higher than the uncoated sample and retained 72.8% of its capacity after 100 cycles at 0.2C [17]. These low-rate cycling measurements are not directly comparable with the high-rate delithiation results of Sharova et al., which used different graphite substrates and processing conditions [7].

### 4.2. Coating modification and structural control

The choice of precursor can affect the amount of residual carbon, even when the final coating contents are similar. Sharova et al. used graphite-to-precursor mass ratios of 4:1 for sucrose and glucose, 3:1 for poly(acrylic acid) (PAA), and 2.5:1 for poly(vinyl alcohol) (PVA). Thermogravimetric analysis (TGA) showed residual coating contents of 5.3 wt%, 4.7 wt%, 4.2 wt%, and 4.8 wt%, respectively. The BET specific surface area also differed between samples. It was 26.1 $m^2 g^{-1}$ for sucrose-coated graphite and 4.0 $m^2 g^{-1}$ for the PVA-coated sample. Pristine graphite had a surface area of 6.9 $m^2 g^{-1}$. Raman spectroscopy showed greater surface disorder in the sucrose-derived coating. The differences in surface area and structure may partly explain the variation in electrochemical performance at comparable coating contents [7].

Polymer precursors can also be used to introduce heteroatoms into carbon coatings. Wang et al. used poly(vinylidene fluoride) (PVDF) and melamine to prepare an amorphous carbon shell containing fluorine and nitrogen. The shell was approximately 15 nm thick. After cycling, the coated graphite showed a stronger LiF signal in the F 1s XPS spectrum than uncoated graphite. Its charge-transfer resistance was also lower. The authors associated these observations with a more stable interphase and improved lithium-storage kinetics [18].

At 0.1C, the initial Coulombic efficiency of the co-doped graphite was 89.02%, slightly below the 89.26% measured for uncoated graphite. After 350 cycles at 2C, the co-doped sample retained a capacity of 113.19 mAh $g^{-1}$. The uncoated graphite delivered 61.21 mAh $g^{-1}$ under the same conditions [18]. The combined carbon coating and fluorine-nitrogen co-doping treatment therefore improved capacity retention during cycling, despite the slightly lower initial Coulombic efficiency.

### 4.3. Preparation methods

Water-soluble sugars and polymers can be deposited onto graphite from aqueous solutions. Sharova et al. compared three preparation methods: solution mixing, dry mixing, and solution mixing followed by ball milling. Initial tests with sucrose showed less particle agglomeration when solution mixing was used. The authors therefore selected this method for the remaining experiments [7].

Graphite and the precursor were stirred in ultrapure water for 8 hours. The mixture was then dried at 80°C, and the resulting powder was ground. Heat treatment was carried out at 500°C for 4 hours under argon. The amount of precursor was adjusted according to its carbon yield. This produced a residual carbon coating of approximately 5 wt% for the different sugar- and polymer-derived samples.

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Unlike the pitch- and resin-based methods described earlier, the aqueous process does not require organic solvents. Drying and heat treatment under an inert atmosphere are still necessary. Heteroatom-containing coatings can also be prepared using polymer precursors. Wang et al. mixed polyvinylidene fluoride (PVDF), melamine, and graphite by ball milling. The mixture then underwent multi-stage high-temperature carbonization. This procedure combines precursor deposition with the incorporation of dopant elements [18].

## 5. Conclusion

Nanocarbon coatings can change the graphite-electrolyte interface. Coating coverage, surface chemistry, and particle packing influence electrochemical performance. Pitch-derived coatings can alter particle morphology and reduce preferential orientation during compaction. Lower charge-transfer resistance has been measured for resin-derived nanoscale shells in single-particle tests. Saccharide and polymer coatings have shown both gains and losses in rate capability.

Adding a carbon layer does not necessarily improve every performance measure. Electrochemical results can differ at similar coating loadings. A larger surface area can increase irreversible lithium consumption. Lower resistance or slower capacity fade may occur alongside reduced initial Coulombic efficiency.

Further testing should use practical electrode densities and areal mass loadings. High-rate delithiation alone provides insufficient evidence of fast-charging capability. Comparisons should account for differences in graphite substrates and preparation conditions. Coating selection involves processing requirements as well as electrochemical performance. Dry mixing avoids solvent recovery. Aqueous deposition avoids organic solvents but still requires drying and carbonization.

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