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Review paper

Screen-printed carbon electrodes: processing, interface reconstruction, performance and sustainability

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Abstract

Screen-printed carbon electrodes (SPCEs) have evolved from inexpensive disposable transducers into tuneable platforms for point-of-care, wearable, and sustainable electrochemical systems. This critical narrative review synthesizes a focused 70-record evidence base, comprising a fixed core of 65 Scopus-indexed records spanning 1996-2026 and five targeted supplementary records on CNC-assisted screen printing, textured and multilayer carbon electrodes, and microfluidic electrochemical integration. Evidence is classified as direct mechanistic, direct analytical, adjacent-transfer, or secondary-context evidence. Across the corpus, SPCE behaviour is interpreted through three coupled control planes: bulk ink microstructure, post-print interfacial reconstruction, and functional device architecture. Carbon morphology, loading, binder chemistry, solvent balance, and print separation define a non-monotonic printability-conductivity window. Plasma, electrochemical, ozone, hydrogen peroxide, and laser treatments can improve charge transfer by exposing carbon sites and modifying surface chemistry, whereas excessive treatment may increase erosion, redeposition, capacitance, or instability. Functional nanostructures can enhance recognition and catalysis but may obstruct intrinsically porous electrodes. Translation therefore requires application-specific benchmarking, authentic-matrix validation, lot-to-lot and storage testing, reference-electrode control, manufacturing qualification, and lifecycle-based metrics. The review integrates operational processing and activation windows with a staged translation ladder, a five-level reproducibility hierarchy, and a gated research roadmap. These design rules provide a practical basis for moving SPCE development from material-centric optimization toward reproducible, scalable, and lifecycle-aware electrochemical technologies.

Keywords

Electrochemical sensing; nanocomposite sensor; rheological behaviour; surface activation; charge-transfer kinetics

Introduction

Screen printing converts a formulated ink into a patterned electrochemical interface through a manufacturing route that is inexpensive, scalable, compatible with small sample volumes, and adaptable to rigid, paper, textile, and polymer substrates. These attributes have established screen-printed electrodes as important platforms for clinical analysis, environmental and food monitoring, and portable measurement [1-3]. The same attributes are also relevant to industrial/process-monitoring and forensic-screening contexts, where rapid, low-cost, and portable electroanalysis is valuable [2,3]. Carbon is especially attractive because its composition, morphology, surface chemistry, and cost can be tuned over a much wider range than noble-metal inks. Recent reviews have accordingly shifted from describing screen-printed electrodes as disposable substitutes for conventional electrodes toward treating them as engineered interfaces whose analytical function emerges from materials selection and fabrication history [4,5].

That conceptual shift is necessary because the term SPCE conceals substantial heterogeneity. Graphite, carbon black, glassy-carbon particles, graphene, mesoporous carbon, and carbon nano-onions form different conductive networks and expose different electroactive sites [6-9]. Commercial inks also contain binders, solvents, plasticizers, and proprietary additives that influence ion exchange, wetting, fouling, and electron transfer. Consequently, even nominally similar bare electrodes can show relative standard deviations above 10 %, while the best electrode material depends on the redox process and target analyte [10,11]. Surface modification alone cannot compensate reliably for an uncontrolled bulk ink or an unstable printed film.

Recent evidence strengthens this process-centered interpretation. Studies from 2025-2026 quantify activation-induced surface reconstruction [12,13], connect particle size and solvent engineering with flexible ink performance [14], compare screen-printed graphene with binder-free laser-induced graphene [15], and extend SPCEs to clinically relevant creatinine measurements [16]. Complementary studies on low-cost mechanized screen printing, textured carbon electrodes, multilayer SPE fabrication, and microfluidic electrochemical integration further show that deposition kinematics, stencil history, layer construction, and device-level fluid handling can materially affect the realized electrode system [17-21]. Recent reviews cover clinical nanomodification [1], general sensor platforms [2], wearable carbon nanocomposites [22], interfacial nanomaterials [5], renewable electrodes [23], and broader printed-electrode technologies [3]. Rather than duplicating these material- or application-centered reviews, this review integrates formulation and print mechanics with activation-induced reconstruction, functional architecture, reproducibility, translation, and lifecycle performance within a single processing-to-deployment framework. The connection between ink rheology and print separation and reconstructed interfaces, functionalization, translation, and lifecycle performance remains fragmented. This review addresses that gap through three coupled control planes: bulk ink microstructure, post-print interfacial reconstruction, and functional device architecture.

This review therefore asks three linked questions. First, how do formulation and printing create the electrochemical surface that later treatments act upon? Second, when does activation improve charge transfer, and when does it damage or destabilize the printed composite? Third, which performance and sustainability claims are sufficiently validated for translation beyond proof-of-concept sensing? The central argument is that SPCE development should be managed as an integrated processing-structure-performance-sustainability problem, not as a sequence of independent material additions. The integrated control-plane logic used throughout this review is summarized in Figure 1.

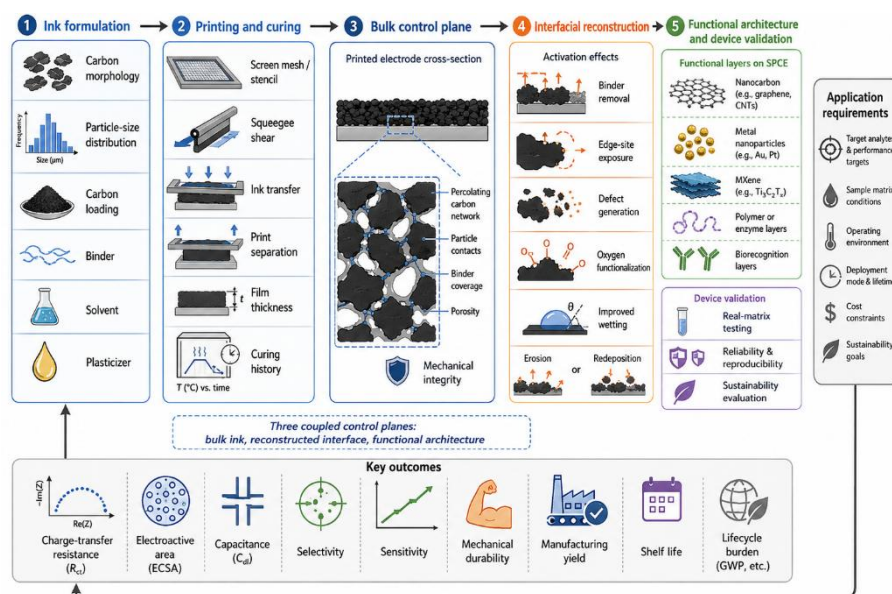


Figure 1. Integrated processing-structure-performance-sustainability framework for SPCE engineering. Ink formulation and printing establish the bulk control plane; activation reconstructs the accessible interface; functional layers create catalytic, recognition, transport, or mechanical functions; and device validation feeds performance, reliability, manufacturability, and sustainability requirements back to process design

Figure 1 establishes the organizing logic used throughout this review. The first control plane is the bulk ink and printed film, in which carbon morphology, particle distribution, binder-solvent balance, and print kinematics determine the percolating network and the amount of carbon accessible to the electrolyte. The second is the reconstructed interface created by activation, where binder removal, defect formation, oxygen functionalities, and wetting can improve charge transfer but can also introduce erosion or redeposition when treatment is excessive. The third is the functional architecture, in which nanocarbon, metals, MXenes, polymers, enzymes, or biorecognition layers address application-specific limitations. The framework closes the loop through reliability, yield, shelf life, and lifecycle burden, so analytical performance is treated as one output of a manufacturing system rather than an isolated material property.

Review scope and evidence framework

Corpus provenance and review design

This article is a critical narrative synthesis centered on a fixed core corpus, not a *de novo* systematic review or meta-analysis. Accordingly, the corpus is used as a focused, question-driven evidence base and is not intended to be statistically representative of the full publication universe retrieved by broad keyword searches. The core evidence base comprised 65 Scopus-indexed records exported on 6 August 2026 and spanning 1996–2026: 55 journal articles, 7 reviews, 2 conference papers, and 1 book chapter. Five targeted supplementary records [17–21] were added to address specific gaps in mechanized carbon-electrode screen printing, multilayer or textured electrode construction, and electrochemical microfluidic integration. These records were not part of the original 65-record export and are therefore kept analytically separate; they are used to support fabrication or device-integration arguments rather than to infer field prevalence or comparative superiority. A supplied Scopus AI thematic overview was used only for topic scoping. The original Boolean search string was not preserved in the supplied export and is therefore not reconstructed post hoc. Bibliographic identities of the core corpus were reconciled by title, authors, year, and DOI,

while the five supplementary records were retained as analytically distinct from the 65-record core corpus.

Evidence eligibility and source roles

All 65 core records were retained for bibliographic accounting, but their evidentiary roles were separated. Direct evidence comprised primary studies on SPCEs or direct comparisons that included an SPCE. Adjacent evidence comprised neighbouring manufacturing platforms, including laser-induced graphene, three-dimensional printed carbon electrodes, silver inks, fuel-cell anodes, natural printing inks, and device-level microfluidic integration; these records were used only to transfer process or translation principles, not to claim demonstrated SPCE performance when the platform differed. Reviews and the book chapter were used for field framing rather than quantitative ranking. The five supplementary records [17-21] were evaluated under the same rules and used only where their methods directly support fabrication, surface-texturing, multilayer-printing, or integrated-device arguments. Evidence strength was interpreted in four levels: A, direct mechanistic primary evidence with structural or interfacial characterization; B, direct analytical primary evidence; C, adjacent transfer evidence; and D, secondary contextual evidence. Mechanistic language was calibrated to this hierarchy.

Data extraction and synthesis logic

Each core and supplementary record used in the synthesis was coded across four layers: (i) ink and substrate formulation; (ii) printing, curing, activation, or modification; (iii) structural and interfacial evidence; and (iv) electrical, electrochemical, analytical, mechanical, or environmental outcome. Numerical values were retained only when explicitly stated in the supplied abstracts or in the verified supplementary source record. Because electrode areas, redox probes, electrolytes, signal processing, replication, and definitions of sensitivity or stability differ substantially, statistical pooling would imply false comparability. Quantitative values are therefore illustrative rather than a league table, and the synthesis emphasizes mechanistic convergence, boundary conditions, reproducibility, and translation readiness.

Ink formulation and printing: the bulk control plane

Carbon morphology, loading and percolation

An SPCE ink is both an electronic material and a transient fluid. Before printing, particles must remain dispersed and the paste must shear-thin under the squeegee. After transfer, it must recover sufficient structure to preserve line definition, release cleanly from the mesh, adhere to the substrate, and form a percolating conductive network after curing. Carbon black, graphite, and graphite nanoplatelets satisfy these requirements differently. Smaller carbon-black particles create more interparticle contacts and can improve conductivity, but they also raise viscosity and elasticity. Increasing carbon loading initially thickens the film and strengthens the conductive network, then reaches a regime in which agglomeration, mesh marking, filamentation, and roughness prevent further electrical improvement [24]. High-speed visualization confirms that elastic ink filaments correspond to local thickness peaks after separation from the screen [25]. The relevant design variable is therefore a process window, not maximum filler content.

Particle-size distribution also requires more rigorous measurement. Flow field-flow fractionation differentiated carbon-black dispersions that dynamic light scattering could not resolve and linked narrower distributions to better dispersion behaviour in ink [26]. For SPCE formulation, this suggests

that a mean particle diameter alone is inadequate. Distribution width, aggregate state, aspect ratio, and the ratio of particle size to mesh opening should be reported. The same principle appears in a 2026 graphite-graphene ink, where 200-mesh graphite, 2 wt.% graphene, and a binary solvent system produced 99.11 % thixotropic recovery, strong adhesion, and only a 1.6 % resistivity change after 300 bending cycles [14].

Mechanized fabrication studies provide a complementary view of the same processing window. A targeted supplementary fabrication study adapted a low-cost CNC 3018 desktop system for carbon-electrode screen printing and reported a repetition level of about 2.6 %, demonstrating that controlled squeegee motion can improve print uniformity without industrial-scale equipment [17]. Related beehive texturing work used two screen-deposition phases to realize a repeatable three-dimensional carbon texture of about 25 μm thickness, with line resolution near 1 mm and approximately 10 % dimensional error [21]. A later multilayer SPE study used a personalized CNC system, carbon conductive ink, PET, a T180 screen, a 2.5 mm snap-off distance, and up to five printed layers, while also documenting dimensional drift of the stencil after repeated use [20]. Together, these studies show that machine kinematics, snap-off distance, layer count, and stencil wear belong inside the bulk control plane because they determine the geometry and continuity of the printed carbon film before any activation or nanomodification is applied.

Binder and solvent as functional components

Binders are frequently described as inactive structural agents, yet they determine which carbon planes are exposed, how ions partition at the interface, and whether the film survives handling. Altering a polymeric ink formulation can shift screen-printed behaviour between edge-plane-like and basal-plane-like graphite reactivity [27]. Commercial inks likewise show target-dependent advantages because an ion-exchange polymer layer may impair alkaline oxidation but improve selective reductive sensing [11]. Evidence from screen-printed fuel-cell anodes, used here as adjacent manufacturing evidence, also indicates that binder content can affect film microstructure and conductivity more strongly than solvent identity within a printable range [28].

Rheology can itself generate electrochemical accessibility. In self-assembling graphene pastes, shear-induced agglomeration and subsequent de-agglomeration during printing exposed a less blocked surface without a separate treatment, with significant correlations among viscosity, thinning threshold, charge-transfer resistance, and peak separation [29]. Conversely, adding 10 % graphene to carbon paste improved responses to three redox systems, but higher addition was not reported as beneficial [7]. Carbon nano-onion/graphite inks similarly required coordinated selection of carbon fractions, binder, solvent, and plasticizer to obtain a uniform conducting network [9]. These findings reject the assumption that a high-conductivity filler automatically yields a high-performance electrode.

Figure 2 clarifies why rheology and electrochemical performance cannot be optimized independently. During screen transfer, a suitable paste must lose viscosity under squeegee shear yet recover rapidly enough to avoid lateral spreading. If elasticity or particle loading becomes excessive, filamentation and mesh marking generate local thickness peaks and roughness [24,25]. After curing, under-filled formulations remain below or close to percolation, whereas highly loaded systems can form agglomerates, binder-rich pockets, or inaccessible pore volume. The desirable region therefore lies between these extremes, where conductive continuity, print fidelity, accessible surface area, and mechanical integrity are simultaneously acceptable. Recent particle-solvent optimization and low-cost mechanized printing studies reinforce that this window depends on

formulation and process settings, not filler content alone. These formulation variables and their implications for printed-electrode performance are summarized in Table 1.

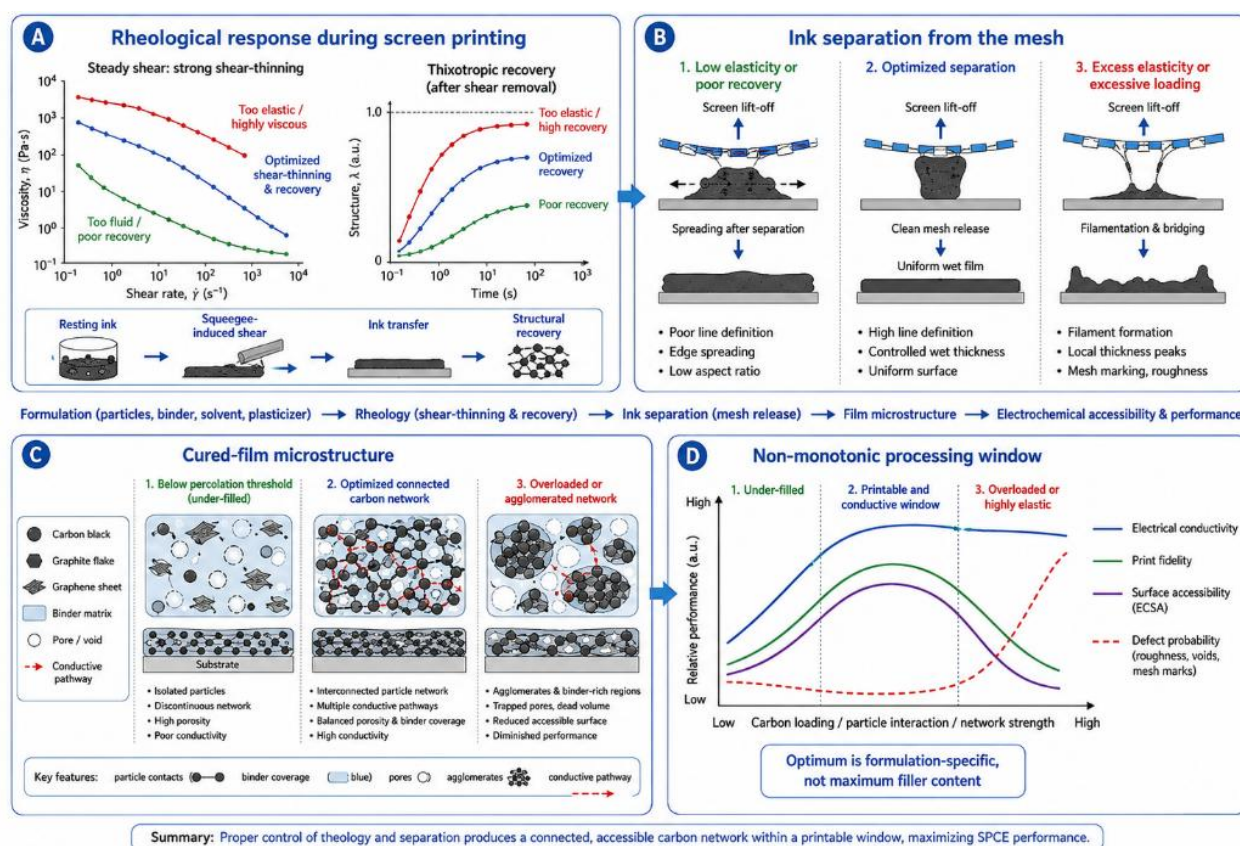


Figure 2. Coupled rheology, ink separation and percolation mechanisms in SPCE fabrication. The schematic links shear-thinning and structural recovery to mesh release, cured-film connectivity, and a non-monotonic printable-conductive window. Trend curves are conceptual and are not pooled quantitative data

Table 1. Formulation variables that connect ink processing to printed-electrode performance

Material system	Formulation lever	Printing or structural response	Functional outcome	Design implication	Ref.
Graphite, glassy carbon, and carbon black	Particle type, purity, size, morphology, and mass loading	Distinct microstructures and accessible carbon sites	Different solvent windows, capacitance, kinetics, sensing and fouling	Select carbon for the target reaction, not only bulk conductivity	[6]
Carbon-black dispersions	Aggregate-size distribution and dispersant hydrophobicity	Narrower, better-resolved particle distributions	Improved dispersion and ink clarity in adjacent ink evidence	Report distribution width and aggregation, not only mean size	[26]
Graphite, carbon black, and GNP pastes	Morphology and loading concentration	Agglomeration and filamentation occur beyond an optimum	Conductivity plateaus while roughness and defects increase	Define a formulation-specific process window	[24,25]
Graphite-graphene hybrid	Graphite mesh size, 2 wt.% graphene, binary solvent balance	Strong shear-thinning, 99.1 % recovery, 5B adhesion	Resistivity of 1.19 mΩ·m; 1.6 % change after 300 bends	Optimize rheology, electrical transport, and mechanics jointly	[14]
Self-assembling graphene paste	Static viscosity and shear-thinning threshold	Reversible clusters open the surface during print transfer	Lower resistance and peak separation without post-treatment	Use print stress as a controlled assembly step	[29]
Graphene-carbon paste	Graphene fraction	Multilayer graphene integrated into carbon-paste network	About 1.7-2.0-fold signal increases at 10 % graphene	Nanocarbon loading has an optimum, not a monotonic benefit	[7]

Material system	Formulation lever	Printing or structural response	Functional outcome	Design implication	Ref.
Three commercial carbon inks	Proprietary binder and additive chemistry	Different surface and ion-exchange behaviour	Performance changes with analyte and pH	There is no universally best commercial ink	[11]
Carbon nano-onion/graphite	Filler ratio, binder, solvent, and plasticizer	Uniform interconnected carbon network	Fourfold sensitivity gain over commercial graphite SPE	Dispersion quality governs the value of nanofillers	[9]
Polymeric carbon ink	Binder formulation	Tailored exposure of edge-like or basal-like graphite behaviour	Tuneable heterogeneous electron transfer	Treat binder chemistry as an electrochemical variable	[27]
Activated carbon with natural binders	Carbon and binder fraction, substrate compatibility	Adhesion fails at excessive carbon or insufficient beeswax	Stable fabric SPCE and 4.38 μM uric-acid LOD	Sustainability must preserve adhesion and wet stability	[30]
Binder-rich functional paste	Binder content within a printable rheological range	Stronger particle network and altered film microstructure	Conductivity and polarization respond to binder level	Adjacent evidence supports binder-first optimization	[28]

Evidence note: rows described as adjacent provide transferable manufacturing principles and are not treated as direct SPCE performance validation

The practical consequence is that sheet resistance cannot serve as the sole ink-qualification metric. Here, the printable-conductive window is operationally defined as the formulation range in which rheological recovery, clean mesh release, line fidelity, adhesion, thickness and roughness, electrical resistance, and electroactive area simultaneously remain within application-specific acceptance limits. A dense low-resistance film may expose few wettable carbon sites, whereas a rough high-area film can increase capacitive current, fouling, or modifier loss. Qualification should therefore combine rheology and print fidelity with electrical and electrochemical metrics, including at least two mechanistically different redox probes and the intended analyte.

Surface activation: interfacial reconstruction as a process window

Activation is often treated as a generic cleaning step. The evidence instead supports a dose-dependent reconstruction process that may combine binder removal, defect generation, oxygen functionalization, pore opening, and, at excessive dose, erosion or redeposition. These changes can affect adsorption-sensitive and outer-sphere redox systems differently. Oxygen plasma produced an approximately 80-fold response increase together with a 4.8 % coefficient of variation, while excessive power reduced reproducibility [31]. Subsequent low-power work showed improved electroactivity at 50-100 W and binder redeposition at 200 to 300 W [32]. Plasma etching also reduced inter-electrode variation from 21.6 to 4.6 %, with image colour level proposed as a manufacturing quality-control indicator [33].

Electrochemical routes provide comparable evidence of a bounded operating window. Saturated Na_2CO_3 activation lowered charge-transfer resistance from 5810 to 801 Ω and increased the heterogeneous rate constant by about one order of magnitude; XPS, Raman, EIS, and density-functional calculations linked the change to defects, oxygen groups, improved wetting, and lower interfacial barriers [13]. Earlier work found the same electrolyte mild and broadly applicable but showed that excessive potential or duration increased capacitance [34]. A two-stage NaOH treatment reduced charge-transfer resistance from about 2100 to 1.4 Ω and increased electroactive area 17-fold [35], while short pre-anodization provided a field-compatible activation route [36].

Oxidative chemical and photonic treatments add further options. Combined H_2SO_4 and H_2O_2 cycling generated a 518-fold sensitivity increase for hydrogen peroxide [37]. H_2O_2 activation across amorphous carbon, MWCNT, graphene, graphene oxide, and mesoporous-carbon inks improved reversibility and lowered impedance, but created material-specific changes in defects, crystallinity, and surface chemistry [12]. Ozone-assisted electrochemical activation required 12 min and

improved several analytes [38]. UV/ozone removed contaminants, increased oxygen groups, and renewed NADH response [39,40]. Laser ablation physically removed polymer binder and created microporosity, decreasing charge-transfer resistance from 41.83 to 5.26 k Ω before enzyme immobilization [41]. Mechanical polishing is supported only by adjacent platinum-SPE evidence and should not be generalized to carbon without direct validation [42]. The competing benefits and failure modes associated with increasing activation dose are summarized in Figure 3.

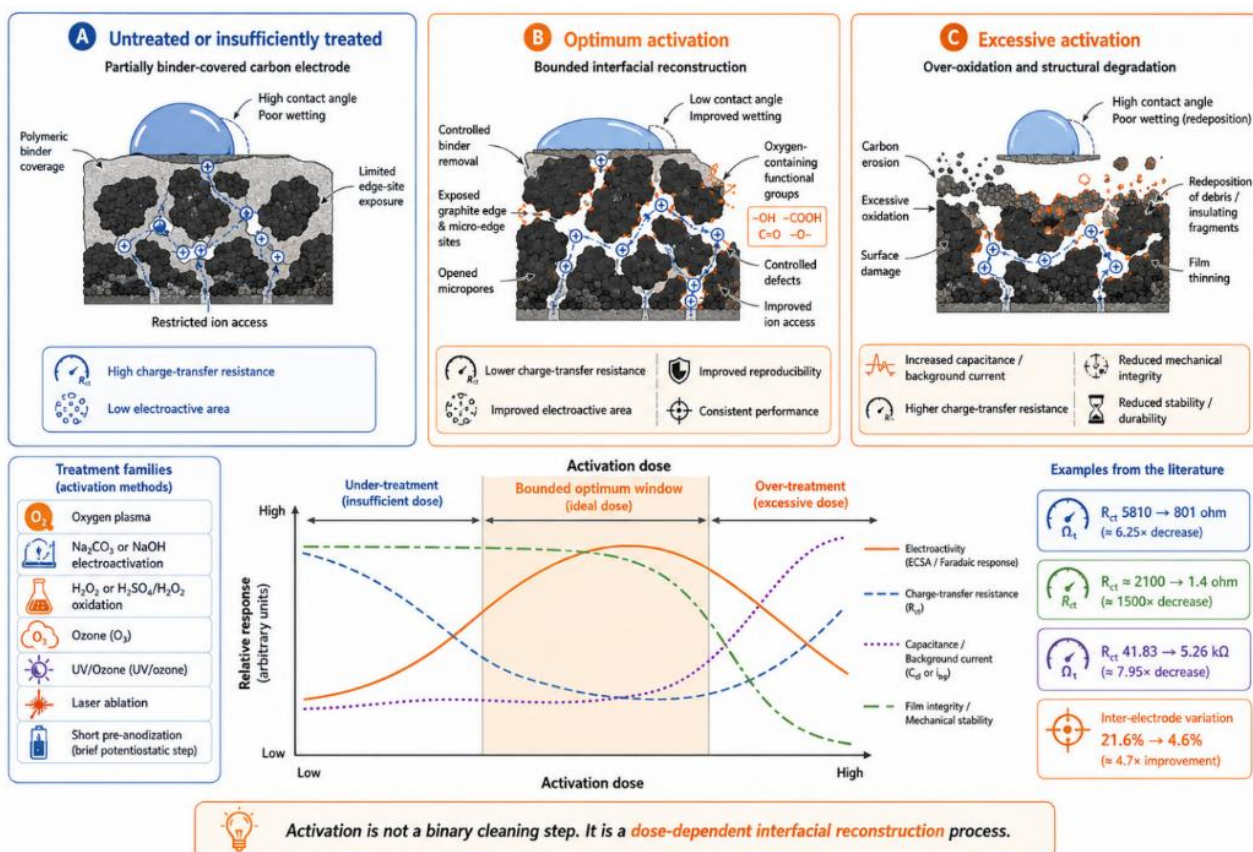


Figure 3. Surface activation as a bounded interfacial reconstruction window. Insufficient treatment leaves binder-covered carbon and restricted ion access; an intermediate dose can expose edge sites, introduce controlled defects and oxygen functionalities, improve wetting, and reduce charge-transfer resistance; excessive treatment can promote erosion, redeposition, capacitance, and loss of integrity. Dose-response curves are conceptual; numerical callouts are literature-specific examples from [13,33,35,41]

Figure 3 translates the surface-treatment evidence into a bounded activation window. Under-treatment leaves substantial binder coverage and restricted ion access. At an appropriate dose, removal of insulating material, exposure of edge sites, controlled defect formation, oxygen functionality, and improved wetting can lower charge-transfer resistance and increase electroactive area [13,31,32,35]. Beyond that window, aggressive oxidation, erosion, film thinning, or redeposition can increase background capacitance and compromise reproducibility or durability [32,34]. The quantitative callouts in the figure are illustrative examples from different studies and should not be interpreted as a pooled comparison because ink formulations, electrolytes, geometric areas, and measurement protocols differ [13,33,35,41]. Accordingly, activation should be reported as an ink-specific dose-response process with both electrochemical benefit and structural integrity used as release criteria. Representative surface-treatment mechanisms, quantitative effects, and boundary conditions are summarized in Table 2.

Table 2. Surface-treatment mechanisms, quantitative effects, and boundary conditions

Treatment	Dominant mechanism	Reported electrochemical change	Principal benefit	Boundary condition or risk	Ref.
Oxygen plasma	Binder removal and edge-plane exposure	About 80-fold response enhancement; coefficient of variation 4.8 %	Rapid dry activation	High power causes erosion or poor reproducibility	[31]
Oxygen plasma plus image analysis	Complete resin etching tracked by surface colour	Inter-electrode variation 21.6 $\rightarrow$ 4.6 %	Manufacturable quality-control concept	Requires ink-specific image calibration	[33]
Low-level oxygen plasma	Power-dependent removal versus binder redeposition	Improved repeatability and electroactivity at 50 to 100 W	Potential whole-device mass treatment	200 to 300 W can recoat active planes	[32]
Na ₂ CO ₃ electroactivation	Defects, oxygen groups, wetting, and electronic coupling	R_{ct} 5810 $\rightarrow$ 801 Ω ; k_0 $9.18 \times 10^{-9} \rightarrow 6.66 \times 10^{-8} \text{ cm s}^{-1}$	Mechanistically resolved reconstruction	Adsorption and diffusion contributions remain coupled	[13]
Mild Na ₂ CO ₃ pre-anodization	Organic-binder removal	Broad improvement across commercial inks	Simple aqueous protocol	More than 5 min or above 1.2 V raises capacitance	[34]
Two-step NaOH pretreatment	Binder extraction followed by functionalization	R_{ct} about 2100 $\rightarrow$ 1.4 Ω ; area increased 17-fold	Strong kinetics and dopamine discrimination	Long, concentrated alkaline exposure may affect integrity	[35]
H ₂ SO ₄ plus H ₂ O ₂ cycling	Oxygen functionalization and resistance reduction	518-fold H ₂ O ₂ sensitivity increase	Large analytical gain	Probe-specific response and multistep processing	[37]
H ₂ O ₂ electroactivation	Ink-specific defect and hole formation	Lower R_{ct} and improved reversibility across five carbons	Broad carbon-material comparison	A single recipe does not create the same surface on every ink	[12]
Ozone-assisted electroactivation	Area increase and carbon-oxygen groups	Improved H ₂ O ₂ , acetaminophen, hydroquinone and dopamine response	Twelve-minute routine pretreatment	Gas handling and dose standardization are required	[38]
UV/ozone cleaning	Contaminant removal and oxygenation	NADH sensitivity 13.31 $\rightarrow$ 19.11 $\mu\text{A mM}^{-1}$	Non-contact cleaning and renewal	Rough nanocarbon surfaces may retain fouling products	[39,40]
Short pre-anodization	Electrochemical exposure of reactive sites	Improved diverse irreversible and quasi-reversible processes	Compatible with on-site disposable use	Ink and curing temperature change the result	[36]
Laser ablation	Selective binder removal and micropore formation	R_{ct} 41.83 $\rightarrow$ 5.26 k Ω	Local, one-step patternable activation	Thermal dose and enzyme-compatible reproducibility need control	[41]

The synthesis therefore supports reporting activation as a dose-response experiment rather than a binary treated/untreated comparison. Operationally, the activation window should be bounded by treatment power or potential and duration, the desired reduction in charge-transfer resistance, acceptable background capacitance, wetting and surface-chemistry changes, and preservation of film integrity. Minimum reporting should include ink identity, curing history, treatment dose, electrolyte or atmosphere, exposed area, resistance, capacitance, morphology, wetting, and surface chemistry. These descriptors help distinguish true kinetic improvement from gains caused mainly by larger geometric area or higher background current.

Functional architectures and analytical translation

Nanocarbon, metal and MXene interfaces

Functional modifiers act on different bottlenecks. Mesoporous carbon increases accessible area and reduces electron-transfer resistance but can improve selectivity without necessarily producing the lowest detection limit [8]. Carbon nano-onions improve current density and dopamine sensitivity when a stable dispersion is achieved [9]. Three-dimensional MWCNT sponge and

graphene foam provide a conductive scaffold for dopamine and enzyme-free glucose sensing [43], while a porous-carbon comparator illustrates how hierarchical pores can separate dopamine and uric-acid signals [44]. These results show that porosity must be matched to analyte transport and signal discrimination, not maximized in isolation.

Metal and hybrid interfaces add catalytic or immobilization functions. Electrochemically deposited gold nanostructures yielded a 0.8 ng mL^{-1} lead detection limit with 2.4% RSD [45]. Nickel nanoparticles on activated SPCEs enabled enzyme-free glucose oxidation [46] and Pd-Pt nanodendrites on ordered mesoporous carbon combined high surface area with bimetallic catalysis for uric acid [47]. Gold and platinum nanostructures changed both antibody loading and electron transfer in CA125 immunosensors, demonstrating that biolayer formation cannot be interpreted as area blocking alone [48].

MXenes extend this architecture by combining metallic conductivity, surface functionality, and compatibility with particles or polymers. Pt/PANI/Ti₃C₂ supported hydrogen peroxide and lactate sensing [49]. Ti₃C₂T_x/MWCNT created a low-resistance path for simultaneous drug analysis [50]. MXene/carbon-dot SPCEs achieved creatinine measurements from 5 μL of serum [16], while enzyme@chitosan/Prussian-blue/MXene@AuNP interfaces coupled enzymatic conversion to efficient peroxide transduction [51]. Yet modifier addition is not universally beneficial. On intrinsically porous laser-induced graphene, AuNP or functionalized-MWCNT deposition partly blocked the conductive structure and reduced activity [15]. The relevant question is which interfacial limitation a modifier removes, and which transport pathways it may obstruct.

From analytical signal to decision-grade measurement

The corpus spans a useful translational ladder. Early demonstrations establish mediator stability and target response, such as poly(thionine) for NADH [52], thionine-modified homemade SPCEs for hydrogen peroxide [53] and pH- and potential-dependent IgG immobilization [54]. More developed systems test selectivity and real samples. Tyrosinase/chitosan/rGO suppressed uric acid and ascorbic acid interference during dopamine measurement [55]. Laser-ablated glucose electrodes were integrated with a smartphone-compatible analyzer and tested in artificial urine, sweat, and serum [41]. Creatinine studies compared results with Jaffe or clinical colorimetric methods [16,51] and an IL-6 aptasensor differed by only 1.38 % from ELISA in cell-culture media [56].

Targeted supplementary device-level studies illustrate how a carbon interface can be embedded within a complete microsystem. A screen-printed carbon-based matrix was coupled to a microfluidic platform for voltage-controlled electro-adsorption of glutamate, demonstrating use of the carbon interface as an active transport and control element rather than only as a passive sensing transducer [18]. In a separate lab-on-chip platform, carbon-based screen-printed electrodes were integrated with a finger-primed pump, stop valves, and a portable electrochemical module for dopamine measurement up to 1 mM using microliter-scale fluid handling [19]. These studies are most appropriately treated as adjacent translation evidence: their value lies in showing that electrode performance must survive integration with sampling, pumping, fluidic confinement, and portable readout, not in providing a directly comparable SPCE detection-limit benchmark.

The reported metrics should not be read as a league table because detection limits depend on electrode area, noise treatment, calibration range, matrix preparation, and replication. Translation is better treated as a staged ladder: Stage 1, mechanistic electrochemical feasibility; Stage 2, target calibration and interference testing; Stage 3, authentic-matrix recovery; Stage 4, agreement with a reference method; Stage 5, lot, storage, and device-level qualification; and Stage 6, prospective

testing on independently collected samples. Only [16] reports a relatively large serum set in the supplied abstracts, while [41,47,48,51,56] and [50] provide progressively stronger real-matrix evidence. Clinical or field readiness also requires calibration transfer, reference-electrode stability, sample pretreatment, and predefined acceptance criteria, not sensitivity alone. For industrial/process-monitoring or forensic-screening use, analogous qualification should emphasize matrix-specific selectivity, calibration stability, contamination or fouling control, traceability, and portable-workflow robustness. Representative analytical platforms and translation-relevant performance are summarized in Table 3.

Table 3. Representative analytical platforms and translation-relevant performance

Target	SPCE architecture	Linear range	Key analytical metric(s)	Matrix and robustness evidence	Ref.
Creatinine	Ti ₃ C ₂ T _x MXene plus carbon dots	Not stated in supplied abstract	LOD 0.016 mg dL ⁻¹	5 μ L serum, $n = 89$, $R^2 = 0.99$ versus Jaffe; 97.8 % response after 15 days	[16]
Creatinine	Enzyme@CS/PB/MXene@AuNP	0.03 to 4.0 mM	LOD 0.01 mM	Serum, urine and saliva; 4 min; recovery 96.8 to 103.7 %	[50]
Dopamine	Tyrosinase/chitosan/rGO	0.4 to 8.0 and 40 to 500 μ M	LOD 22 nM	Human urine; designed against uric- and ascorbic-acid interference	[55]
Hydrogen peroxide	Electropolymerized thionine on home-made SPCE	Not stated	Not stated	Applied to real samples after fabrication and pretreatment optimization	[53]
Glucose	Laser-ablated SPCE with immobilized GOx	0.05 to 5 mM	LOD 2.0 μ M; 244 μ A mM ⁻¹ cm ⁻²	Artificial urine, sweat, and serum; recovery 95.4–109.4%; portable analyzer	[41]
NADH	Poly(thionine)-modified SPCE	5 to 100 μ M	Not stated	Stable, reproducible flow-injection response	[52]
Glucose	Activated SPCE with Ni nanoparticles	5 to 1500 μ M	LOD 0.28 μ M	Human serum and urine; enzyme-free operation	[46]
Uric acid	Pd-Pt nanodendrite on ordered mesoporous carbon	0.25 to 800 μ M	LOD 0.25 μ M; 154.45 μ A mM ⁻¹ cm ⁻²	Serum recovery 97 to 102 %; 380 injections; storage beyond 5 weeks	[47]
Dopamine and glucose	MWCNT sponge/graphene-foam SPCE	Not stated	Glucose LOD 4.96 μ M; 526 μ A mM ⁻¹ cm ⁻²	Selectivity demonstrated; real-matrix evidence not stated	[43]
pH	Flexible f-MWCNT working electrode	pH 4.0 to 8.9	33.66 mV pH ⁻¹	Artificial saliva; 19.71 s settling; 72 h stability; 78.57 % fabrication success	[57]
CA125	Au- or Pt-nanostructured antibody SPCE	Not stated	LOD 419 or 386 ng mL ⁻¹	Human plasma; binding-linked changes in electron-transfer rate	[48]
Lactate	Lactate oxidase on Pt/PANI/MXene	0.005 to 5.0 mM	LOD 5.0 μ M	Milk; reported high stability and reliability	[49]
IL-6	AuNW/AuNC/PAA plus thiolated aptamer	0.1–1000 pg mL ⁻¹	LOD 0.1 pg mL ⁻¹	Human serum recovery 93.9–104.8%; 1.38% difference versus ELISA	[56]
Paracetamol, caffeine, theophylline	Ti ₃ C ₂ T _x MXene/MWCNT	Analyte-specific ranges to about 90 μ M	LOD 0.23, 0.57 and 0.43 μ M	Human blood; $R_{ct} \approx 95 \Omega$; simultaneous non-enzymatic assay	[50]
Dopamine	Carbon-based screen-printed electrodes integrated with finger-primed microfluidics	Up to 1 mM	Portable electrochemical detection; analytical LOD not stated	50 to 100 microliter handling; finger pump and stop valves; integrated lab-on-chip demonstration	[19]

Flexible, additive, and sustainable manufacturing

Mechanical reliability and hybrid fabrication

Flexible devices expose the trade-off between electrochemical access and mechanical survival. TPU/MWCNT fabric electrodes changed resistance by only 0.8 % after 1000 bends and 5.0 % after 100 folding cycles, while retaining adhesion after washing [58]. Paper-based graphite sensors showed

better electrochemical resilience under contortion than polymer-supported counterparts [59]. A flexible pH platform reported a 78.57 % fabrication success rate, a valuable reminder that manufacturing yield should accompany sensitivity [57]. Broad wearable evidence identifies cost, biocompatibility, flexibility, mass production, and long-term stability as coupled constraints [22].

Laser-induced graphene and additive manufacturing are relevant comparators rather than direct substitutes in every application. Design-of-experiments optimization produced binder-free LIG with a 500 % higher nitrite peak current than a commercial graphene-sheet SPCE, but different laser conditions also changed ionic interference [60]. A 2026 comparison reported roughly sixfold higher current density and a 97.7 % lower process-related carbon footprint for LIG relative to in-house screen-printed graphene [15]. Three-dimensional printing enables patient-specific pyrolyzed carbon electrodes [61], MXene-coated carbon-black micro-supercapacitors with 95 % retention after 10,000 cycles [62] and heterolayered carbon architectures [63]. Broader additive-electrode studies show that printability, ionic tortuosity, inactive polymer, and throughput remain scale barriers [3,64]. Hybridization should therefore be justified by an architectural need, not by manufacturing novelty alone.

Sustainability beyond renewable ingredients

Natural and waste-derived materials are expanding the SPCE design space. Activated carbon, coconut oil, and beeswax produced a fabric uric-acid sensor, but adhesion failed when carbon was too high or beeswax insufficient [30]. Coconut-shell activated carbon with a cellulose-derived binder yielded a biodegradable ink, with 40 wt.% carbon balancing rheology, printability, and biodegradability [65]. Reviews of waste-derived disposable electrodes emphasize paper, agricultural by-products, and recovered materials [23]. Adjacent ink studies add bio-based PLA/Cyrene silver formulations that survived 5000 bending cycles [66], neem-gum formulations with tuneable viscoelasticity [67], and natural paper inks that still face water-resistance and scale challenges [68].

Renewable content, however, is not equivalent to lower lifecycle burden. A device can contain a natural binder yet require toxic solvents, energy-intensive curing, noble-metal modifiers, single-use plastics, or cold-chain storage. Conversely, a non-biodegradable electrode may reduce total burden if it is reusable, highly stable, or requires very little sample and reagent. Sustainability claims should therefore define a functional unit, such as validated analyses delivered within specification, and include material origin, solvent hazard, curing energy, manufacturing yield, packaging, shelf life, assay consumables, disposal, and recovery options.

The evidence supports a three-tier hierarchy for environmental claims. Tier 1, material-origin sustainability, documents renewable, recycled, or waste-derived feedstocks. Tier 2, manufacturing sustainability, demonstrates printability, conductivity, adhesion, wet stability, shelf life, solvent safety, energy demand, and manufacturing yield under intended use conditions. Tier 3, functional lifecycle sustainability, compares the complete device with a conventional SPCE using a defined functional unit and includes reagents, packaging, storage, use, disposal, and recovery. Most studies in the supplied corpus remain at Tiers 1 or 2 [23,30,65-68]. The LIG comparison adds process-level carbon-footprint evidence [15], but does not by itself establish whole-device superiority. Trade-offs should be reported explicitly because biodegradability can reduce shelf life or increase material consumption when yield is low. Representative manufacturing and sustainability strategies, evidence maturity, and remaining validation needs are summarized in Table 4.

Table 4. Manufacturing and sustainability strategies, evidence maturity, and next validation needs

Strategy	Demonstrated benefit	Scale or durability evidence	Unresolved issue	Required next validation	Ref.
Image-based plasma quality control	Surface colour correlated with complete binder etching	Inter-electrode variation reduced to 4.6 %	Transfer across inks, cameras, and lighting	Calibrated inline imaging linked to electrochemical release limits	[33]
DoE-guided laser processing	Captures interactions among power, speed, and focus	Stable porous LIG and large signal gain	LIG is an adjacent platform and interference changed with settings	Cross-lot optimization with predefined robustness criteria	[60]
Data-driven property prediction	Nonlinear models link multiple carbon features to capacitance	Demonstrated on literature-derived supercapacitor data	Domain shift and incomplete manufacturing descriptors	SPCE-specific datasets with external and prospective validation	[69]
TPU/MWCNT textile ink	Strong adhesion and resistance stability	1000 bends, 100 folds, and washing tests	Electrochemical drift under sweat and repeated wear	Combined mechanical, chemical and biocompatibility aging	[58]
Paper substrate	High flexibility and low material demand	Resilient electrochemistry under contortion	Humidity, barrier coatings, and storage	Standard bend radius, cycles, humidity, and shelf-life protocol	[59]
Biodegradable activated-carbon ink	Renewable carbon and binder-compatible printing	Printable on packaging and flexible substrates	Conductivity, wet stability and end-of-life conditions	Functional biodegradation plus full sensor performance	[65]
Bio-based silver ink	Lower-hazard binder and solvent; high conductivity	Electrical continuity after 5000 bends and over 30 % strain	Adjacent non-carbon evidence; silver recovery burden	Comparative lifecycle and recovery assessment	[66]
Waste and renewable electrodes	Reduces virgin feedstock and supports circular design	Multiple material routes summarized	Variable purity and inconsistent functional units	Traceability, contaminant control and application-specific LCA	[23]
Neem-gum conductive ink	Natural binder with tuneable rheology	Screen-printable viscoelastic formulations	Conductivity remains formulation-dependent and non-SPCE	Carbon-filled SPCE trials, wet aging, and adhesion testing	[67]
Multi-material 3D carbon architecture	Spatially separates complementary carbon allotropes	Improved sensing and energy-conversion response	Polymer dilution and throughput	Hybrid screen/3D pilot lines with cost and yield data	[63]
MXene-coated 3D carbon electrode	Custom geometry and long cycling life	95 % capacitance retention after 10,000 cycles	Energy-storage evidence does not establish sensing reliability	Application-specific transport, fouling, and scale studies	[62]
CNC-assisted and multilayer carbon screen printing	Low-cost mechanized deposition, textured carbon surfaces, and controlled layer construction	About 2.6 % repetition in CNC printing; about 25 μm textured films; up to five printed layers	Stencil wear, snap-off coupling, pressure control, and transfer to higher-throughput production	Cross-lot DoE with dimensional, electrical, and electrochemical release criteria plus inline stencil/print inspection	[17,20,21]

Evidence note: LIG, 3D-printed energy-storage electrodes, silver inks, and natural-ink studies are adjacent evidence used only where a transferable manufacturing principle is explicit.

Cross-cutting design rules and research agenda

Design rules for reproducible SPCE engineering

Six design rules emerge. First, ink disclosure should extend beyond a commercial name to carbon morphology and distribution, filler fraction, binder and solvent composition, rheological recovery, mesh, print speed, squeegee kinematics, snap-off distance, layer count, stencil history, film thickness, cure, and storage history [6,11,14,17,20,21,24-26,28,29]. Second, activation should be specified as a dose with before-and-after evidence because the optimum is bounded by incomplete binder removal at low dose and capacitance, erosion, redeposition, or instability at high dose [12,13,31,32,34,37]. Third, electrochemical benchmarking should combine an outer-sphere probe, a surface-sensitive probe, and the intended analyte with EIS, background capacitance, and electroactive-area estimates because ink chemistry and pretreatment affect different redox processes differently [6,10,11,36,40,70].

Fourth, nanomodification should be hypothesis-driven: each modifier should address a defined limitation in catalysis, recognition, antifouling, or transport and should be checked for pore blockage and added variability [5,15,43,48-50]. Fifth, reproducibility should be qualified hierarchically at five levels: within-device repeatability, electrode-to-electrode reproducibility, batch-to-batch reproducibility, operator or instrument reproducibility, and ultimately inter-laboratory reproducibility. Mean sensitivity should therefore be accompanied by distributions, manufacturing yield, reference-electrode drift, storage stability, mechanical cycling where relevant, and failure analysis [10,17,20,33,57-59]. Sixth, real-world readiness requires predefined matrix, comparator, fluidic-integration, and sustainability criteria; spiked-sample recovery alone is not equivalent to application-specific decision agreement, system-level robustness, or lifecycle superiority [15,16,18,19,41,47,51,56].

Research priorities and qualification gates

Five priorities follow. Cross-laboratory round-robin studies should test reference inks and shared activation protocols using common probes and acceptance limits. Multiscale models should connect particle distribution and rheology with screen separation, binder exposure, wetting, and charge transfer; DoE, inline imaging, and low-cost mechanized printing studies provide practical starting points [14,17,20,21,33,60], while machine learning requires standardized SPCE descriptors and external validation [69]. Scalable functionalization should compare in-ink, post-print, and in situ routes under identical lot and aging tests. Flexible devices require combined mechanical, chemical, and electrochemical aging. Lifecycle and techno-economic assessment should compare manufacturing routes using application-specific functional units.

Evidence limitations and scope boundaries

Four limitations constrain interpretation. First, the core corpus was predefined and the original search string was unavailable, so exhaustive systematic coverage is not claimed; the five supplementary records [17-21] were identified during revision and were therefore kept analytically separate from the original export. Second, abstract-level extraction leaves some experimental conditions, replication details, and negative results unavailable. Third, heterogeneous probes, areas, matrices, and performance definitions preclude defensible statistical pooling. Fourth, adjacent evidence supports manufacturing or device-integration inference but not direct SPCE validation. Accordingly, the 70-record corpus should be interpreted as a focused evidence base for the stated review questions rather than as a census or bibliometric representation of the thousands of SPCE records retrievable by broad keyword searches. These constraints are addressed through explicit evidence-role separation, transparent provenance, and conservative mechanistic language.

Figure 4 converts the cross-cutting design rules into a gated development sequence. The first two stages establish traceable descriptors and common benchmarks before predictive models or advanced functionalization are attempted. The third and fourth stages connect process control with interface reconstruction, scalable modifier integration, aging, and manufacturing yield. Only after these gates are satisfied does the roadmap advance to authentic-matrix comparison, independent sample sets, and field or clinical acceptance criteria. Lifecycle and techno-economic assessment is placed at the final decision stage but is informed by sustainability across the entire pathway. The three horizontal themes emphasize that reproducibility, reliability, and sustainability are not end-stage checks. They are constraints that must be maintained across formulation, fabrication, activation, validation, and deployment [17,20,21,23,33,58,60,65,69].

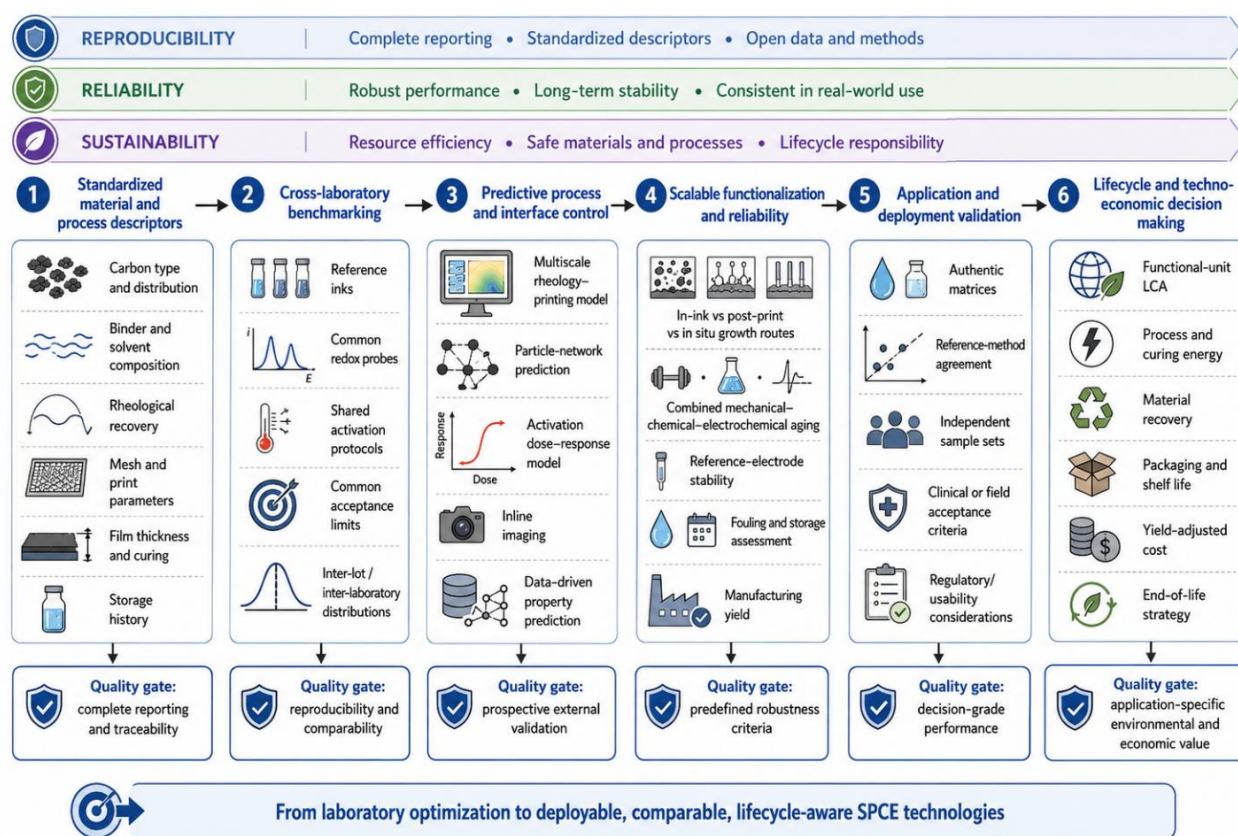


Figure 4. Gated research roadmap for reproducible and deployable SPCE systems. Six stages progress from standardized process descriptors and cross-laboratory benchmarking to predictive control, scalable functionalization, application validation, and lifecycle and techno-economic decision making, with reproducibility, reliability, and sustainability treated as cross-cutting constraints

Conclusions

SPCE performance emerges from a coupled manufacturing system. Ink particles and polymers establish the conductive network and determine how much carbon is accessible. Printing and curing create spatial variability. Activation can remove binder, create defects, improve wetting, and accelerate charge transfer, but the same treatment can also raise capacitance, erode the film, or redeposit insulating material. Functional nanostructures can supply catalytic, recognition, and transport advantages, yet they add interfaces and may block an already porous electrode.

The most credible recent studies move beyond detection-limit reporting by linking physicochemical characterization with electrochemical response, testing authentic matrices, comparing reference methods, reporting mechanical or storage stability, and considering manufacturing yield or environmental burden. Targeted supplementary evidence on low-cost CNC-assisted fabrication, multilayer screen printing, and microfluidic integration further emphasizes that reproducibility is created across the entire manufacturing and device chain rather than at the electrode surface alone [17-21]. The field should therefore move from material-centric experimentation toward controlled integration of three linked planes: bulk ink structure, reconstructed interface, and functional architecture. Standardized descriptors, orthogonal electrochemical probes, cross-lot statistics, staged translation criteria, inline quality control, and lifecycle functional units would make results more comparable and transferable. Within the limits of the focused 65-record core corpus and the explicitly separated targeted supplements, this processing-to-interface framework provides a defensible route for converting versatile laboratory SPCEs into reproducible technologies for point-of-care diagnostics, wearable monitoring, environmental analysis, industrial/process monitoring, forensic screening, and

distributed sensing. Taken together, the integrated control-plane framework in Figure 1 and the gated roadmap in Figure 4 provide a practical basis for moving from material-by-material optimization toward reproducible, application-specific, and lifecycle-aware SPCE development.

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