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Systematic Review

Role of Graphene Oxide and Reduced Graphene Oxide in Electric Double-Layer Capacitors: A Systematic Review

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Abstract: The evolution of electric double-layer capacitors (EDLCs) has significantly benefited from advancements in graphene-based materials, particularly graphene oxide (GO) and reduced graphene oxide (rGO). This systematic review consolidates and analyzes existing research on the roles of GO and rGO in enhancing the performance of EDLCs, focusing on synthesis methods, electrode fabrication, electrolytes, and performance metrics such as capacitance, energy density, and cycling stability. Following the PICOS and PRISMA frameworks, a comprehensive literature search was conducted across Scopus, Web of Science, PubMed, and IEEE Xplore, covering the period from 2010 to 2023. A total of 128 articles were initially identified, with 27 studies meeting the inclusion criteria after rigorous screening and full-text analysis. Key findings reveal that the incorporation of GO and rGO in EDLCs leads to significant improvements in specific capacitance, energy density, and cycling stability. Notable advancements include novel synthesis techniques and composite materials such as nitrogen-doped graphene, graphene/polyaniline hybrids, and various metal oxide–graphene composites, which exhibit superior electrochemical performance. However, challenges such as material scalability, environmental sustainability, and consistency in synthesis methods remain. This review stresses the great potential of GO and rGO in the development of high-performance EDLCs and highlights the need for continued research to address existing challenges and further optimize material properties and fabrication techniques.

Keywords: graphene oxide; reduced graphene oxide; electric double-layer capacitor; systematic review; capacitance; energy density; cycling stability

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1. Introduction

The evolution of electric double-layer capacitors (EDLCs) has seen significant contributions from advancements in graphene-based materials. Particularly, graphene oxide (GO) and reduced graphene oxide (rGO) have emerged as promising materials, enhancing the performance metrics of EDLCs through various innovative synthesis methods [1–3] and applications [4–6]. The objective of this systematic review is to consolidate and analyze existing research on the roles of GO and rGO in enhancing the performance of EDLCs, focusing on synthesis methods, electrode fabrication, electrolytes involved, and performance metrics to identify challenges and propose future research directions.

1.1. Advances in EDLCs

Recent advancements in the development of graphene-based materials have improved the performance of EDLCs. A novel one-pot hydrothermal carbonization approach for synthesizing flowerlike graphene-based particles significantly improved the specific surface area and electrochemical performance of EDLCs [7]. This method demonstrated efficiency in enhancing electrode capacitance and charge retention, indicating its potential for advanced energy storage systems. Building on this, a binder-free hybrid material combining copper(II) oxide (CuO) microspheres and rGO nanosheets exhibited enhanced electrochemical properties, including a high specific capacitance of 244 F/g and excellent stability over 1000 cycles [8]. The synergistic effect between the components improved charge transfer efficiency, making it a viable alternative for high-performance energy storage applications.

In exploring composite materials, a rGO/polyaniline (PANI) composite for supercapacitors was synthesized and characterized. The successful coating of PANI on rGO via in situ polymerization enhanced conductivity, resulting in a specific capacitance of up to 600 F/g [9]. This composite demonstrated excellent electrochemical performance, rate capability, and cycling stability, indicating its promise for energy storage applications. Additionally, glucose/graphene-based aerogels (G/GAs) developed using hydrothermal reduction exhibited exceptional gas adsorption capabilities and electrochemical performance, crucial for applications in EDLCs and gas separation technologies. The high specific surface area and hierarchical porosity of G/GAs highlight their potential in environmental and energy applications [10].

Three-dimensional graphene networks (3DGNs) fabricated using an electric field-assisted assembly method demonstrated a high specific capacitance of 238 F/g and maintained good rate capability [11]. Further advancements include the development of solid-state fractional-order electric double-layer capacitors (FO-EDLCs) that allow tuning of the low-frequency impedance phase angle. Using a gel electrolyte mixed with carbonaceous additives like rGO, these capacitors demonstrated subdiffusive charge transport and frequency dispersive capacitive-resistive behavior, impacting their performance and potential applications [12]. Laser-scribed graphene (LSG) fabricated from oil palm lignin resulted in high specific capacitance and excellent cycling stability, showcasing the potential of renewable resources in developing high-performance energy storage devices [13]. In the context of electric vehicles, advancements in electrode materials achieved a maximum efficiency of 95% using a composition of rGO, ketjen black, and PVDF, highlighting the importance of supercapacitors in enhancing cyclic stability and energy density for sustainable electric vehicle development [14].

Techniques such as laser reduction of graphene oxides and laser-induced graphene have significantly enhanced the electrochemical performance and integration of graphene-based supercapacitors, indicating major developments in energy storage devices [15]. The integration of metal oxides into carbon frameworks and various supercapacitor configurations based on charge storage mechanisms have been also explored, highlighting significant advancements in the field [16]. In the same way, environmentally friendly methods for synthesizing rGO have been introduced, emphasizing their applications in energy-related materials without the use of toxic reagents [17]. These sustainable methods are preferable for scalable industrial applications in energy harvesting. The distinct advantages of supercapacitors over traditional batteries and capacitors were discussed, covering various electrode materials and electrolytes and emphasizing the importance of material choice in performance, potential applications in grid stabilization, and automotive uses [18].

Alternative strategies include the use of ruthenium in supercapacitors, confirming the rapid reversible redox process and environmental adaptability of Ru-based materials. Various morphologies and structures of Ru composites were explored, demonstrating their potential to enhance supercapacitor performance [19]. Additionally, advancements in hybrid energy storage systems, focusing on hybrid supercapacitors that combine high specific energy and power with long cycle life and fast self-charging capabilities, show significant potential for portable electronics and wearable devices [20].

1.2. Innovations in Graphene for EDLCs

Various studies have explored different synthesis methods and modifications to enhance the electrochemical performance and structural properties of graphene materials, providing new insights into their potential applications. A novel synthesis route for creating graphene-cross-linked phenol-formaldehyde (PF) hybrid carbon xerogels involves the polycondensation of phenol and formaldehyde cross-linked by GO, followed by ambient pressure drying. This method resulted in significant improvements in structural integrity, reduced shrinkage during drying, and enhanced electrochemical properties as electrode materials in EDLCs [21]. The incorporation of GO not only strengthened the gel network but also contributed to a higher specific surface area and mesoporous volume, enhancing the electrochemical performance of the xerogels. Tailoring the oxygen content of rGO for specific applications was explored by controlling the oxidation levels. It was found that rGO with a 23.1% weight oxygen content exhibited an optimal balance between high surface area and adequate electron conductivity [22], providing guidelines for customizing the properties of graphitic materials for various applications, including energy storage.

Enhancements in carbon aerogels with graphene, GO, and carbon nanotubes (CNTs) were achieved for use as electrodes in symmetric supercapacitors. These modifications led to significantly improved morphological and capacitive properties, with CNT-modified aerogels achieving a capacitance of up to 326 F/g [23]. The potential of these modified aerogels in EDLCs was demonstrated by utilizing supercritical acetone drying to reduce material shrinkage and enhance performance. A comparison of ionic liquid electrolytes to aqueous electrolytes for carbon nanofiber supercapacitor electrodes derived from oxygen-functionalized graphene revealed that devices using ionic liquid electrolytes exhibited superior energy density and stability [24]. The carbon nanofibers synthesized using a reflux technique demonstrated excellent electrochemical properties, particularly in ionic liquid, due to its higher operating voltage and electrochemical stability.

Porous poly(vinylidenefluoride-co-hexafluoropropylene) (PVdF-HFP) composite membranes enhanced with GO were developed for use in EDLCs [25]. The inclusion of GO improved the physicochemical properties of the membranes, enhancing their electrochemical performance and ionic conductivity. This enhancement was attributed to the porous nature induced by GO, which facilitated ion transport and electrolyte uptake, presenting these composites as promising materials for future energy storage applications. An innovative method for synthesizing rGO from agricultural wastes such as coconut shells and coconut coir demonstrated a green and cost-effective process [26]. This method showed potential for large-scale production of rGO for use in supercapacitor electrodes, showing excellent cyclic stability and electrochemical performance. The simplicity of the method and the use of waste materials contribute to reducing environmental impact, aligning with sustainable manufacturing practices.

The synthesis of mesoporous nanohybrids composed of two-dimensional Cu–Cr phosphate (CCP) and rGO for high-performance asymmetric hybrid supercapacitors (AHS) exhibited superior electrochemical properties, including high energy and power densities and excellent cyclic stability [27]. These properties were attributed to the mesoporous structure and synergistic effects between the metal phosphate and graphene materials. Investigations into the effects of different thermally reducing temperatures on GO thin films for micro-supercapacitors found that varying the reduction temperature altered the electrochemical properties and structure of the films [28]. Lower reduction temperatures resulted in diffusion-controlled behavior, while higher temperatures led to surface-controlled behavior, optimizing the electrochemical performance of micro-supercapacitors. A promising approach to improve the performance of aqueous batteries was introduced by using a GO-modified glass fiber separator designed to selectively transport ions [29]. This separator enabled selective ion transport, enhancing the cycling stability of the battery and incorporating an intra-series architecture that provided extra capacity through the EDLC effect. The battery showed improved cyclic life and high-capacity retention, offering a new perspective for enhancing aqueous battery performance.

1.3. Advanced Synthesis and Processing Techniques for Graphene-Based EDLCs

The development of advanced synthesis and processing techniques for graphene materials has significantly enhanced the performance of EDLCs. For instance, the potential of polymer/graphite oxide composites as electrode materials for EDLCs was demonstrated by integrating graphite oxide with a poly(ethylene oxide)-based polymer. The composite exhibited a double-layer capacitance of up to 130 F/g, attributed to improved ion transport within the electrode material due to the influence of the polymer on increasing the interlayer spacing of graphite oxide. This approach offers a promising alternative to traditional electrode materials, aiming to enhance the energy storage capability and stability of EDLCs [30]. Similarly, superparamagnetic magnetite nanocrystal (Fe_3O_4)–graphene oxide nanocomposites were synthesized through a simple, one-step solution-processed method at ambient conditions [31]. The composite materials demonstrated enhanced electrochemical performance as electrodes for EDLCs, showing superior specific capacitance and high-rate charge–discharge capabilities compared to individual Fe_3O_4 nanocrystals and GO. This highlights the potential of combining magnetic nanoparticles with GO for improved performance in energy storage devices.

The influence of the electrochemical reduction process on the performance of graphene-based capacitors was examined, revealing that optimal reduction conditions—applying a potential of -1.0 to 1.0 V for 4000 s—resulted in a specific capacitance of 246 F/g [32]. The significant impact of residual oxygen functional groups and the structure of sp^2 domains on the capacitance performance of electrochemically reduced graphene oxide (ERGO) was demonstrated, showing that proper control of electrochemical reduction can optimize these factors for enhanced capacitor performance.

The assembly of GO for EDLC electrode application was explored by processing GO into microparticles through spray drying, creating rGO microparticles with corrugated surface morphology. This method maintained high specific capacitance even at high current densities, attributed to the unique surface morphology of the rGO microparticles. The simplicity and scalability of this process offer a practical approach to manufacturing EDLC electrodes [33,34]. A novel single-step method for the exfoliation and reduction of graphite oxide using benzidine was introduced [35], significantly impacting the electrochemical properties of the resultant graphene-based materials. The process enabled the accommodation of benzidine within the interlayer space of GO, achieving specific capacitance values of up to 178 F/g in aqueous solutions.

A bottom-up method for fabricating activated carbon fiber (ACF) using GO as both a dispersant and a binder was described. The resulting ACFs exhibited high electrical conductivity, significant mechanical flexibility, and large specific surface areas [36]. These fibers were used to construct all-solid-state flexible supercapacitors, demonstrating high energy and power densities, a robust cycle life, and excellent mechanical flexibility. This method offers a scalable solution to the challenges of incorporating conventional activated carbon in flexible and wearable electronic devices.

A novel composite material consisting of SnO_2 nanoparticles embedded in a rGO matrix was synthesized for use as a negative electrode in lithium-ion capacitors (LICs) [37]. The SnO_2 nanoparticles, with a size below 10 nm, were synthesized via an in situ synthetic approach involving freeze drying and thermal reduction. This nanostructuring enhanced the electrochemical performance of the electrode by providing short lithium-ion diffusion paths and a more homogeneous particle distribution, mitigating volume changes during operation. The charge transfer kinetics at carbon/hydroquinone interfaces in redox-active-electrolyte supercapacitors (RAESs) were explored using scanning electrochemical microscopy (SECM) [38]. This study highlighted the advantages of SECM over traditional cyclic voltammetry in obtaining more accurate kinetic constants by avoiding errors like ohmic drop and charging current. These findings suggest significant potential improvements in energy storage technology through better understanding and manipulation of interfacial reactions.

Lastly, chemical vapor deposition-grown graphene (CVD-G) on nickel foil used as a current collector in the fabrication of supercapacitors was optimized to reduce equivalent series resistance (ESR) [39]. Growing CVD-G at different cooling rates revealed that a cooling rate of 100 °C/min resulted in the lowest ESR of 0.38 Ω, significantly enhancing charge–discharge characteristics. This approach improved supercapacitor performance by reducing interface resistance, leading to faster charge and discharge rates and better high-frequency response in cyclic voltammetry tests.

1.4. Enhanced Performance of Graphene-Based Materials in Energy Storage Applications

Groundbreaking applications of graphene materials have led to significant advancements in the performance of EDLCs and other related technologies. High-performance cellulose-based nanocomposite soft actuators were developed, incorporating a porous high-conductivity electrode made from a cellulose–chitosan framework doped with graphene-coated carbon nanosheets [40]. This structure enhanced the electrochemical and electromechanical properties of the actuators, demonstrating significant improvements in peak-to-peak displacement and specific capacitance, indicating potential for advanced applications in fields requiring precise motion control and high performance. Enhancing the structural integrity and electrochemical properties of 3D graphene, carbon nanodots (CNDs) were incorporated into the laser reduction process of GO [41]. This method increased the electrical conductivity and active surface area of the resultant 3D graphene, significantly improving its performance as EDLC electrodes. Additionally, investigations into the interactions at the electrode/electrolyte interface in carbon-based EDLCs using solvent-free ionic liquids as electrolytes provided crucial insights into ion-electrode compatibility and factors limiting charge accumulation, which are essential for optimizing supercapacitor behavior and performance [42].

A novel approach for fabricating high-performance supercapacitors involved the use of free-standing and flexible graphene electrodes created by laser holography reduction of graphene oxide [43]. This technique enabled the creation of hierarchically structured photoreduced graphene oxide (PRGO), significantly enhancing areal capacitance through precise patterning and beneficial micro-nanostructures that facilitated rapid ion transport and increased electrochemically accessible surface area. Similarly, flexible micro-supercapacitors were fabricated using laser-scribed graphene with fractal designs [44], enhancing capacitance and energy density compared to conventional interdigitated designs. These fractal designs optimized the electrochemically active surface area, improving performance through efficient space utilization and increased edge effects, enhancing ion transport and charge storage capabilities.

Further advancements include the preparation of composite materials integrating EDLC and pseudocapacitance characteristics using nickel hydroxide ($Ni(OH)_2$) and rGO, enhanced with cobalt (Co) doping. The Co-doped nanosheets exhibited a remarkable initial specific capacitance of 2006 F/g and retained 80.60% of this capacitance after 1000 cycles, significantly outperforming the undoped composite [45]. This substantial improvement highlights the potential of Co doping for enhancing nickel-based supercapacitor materials. In the context of capacitive deionization (CDI) applications aimed at water desalination, novel electrode materials were developed by coating single-layer graphene oxide (SGO) onto activated carbon fibers (ACF). The resulting SGO@ACF composites exhibited a high specific surface area of 1532.73 m^2/g and impressive electrochemical properties, achieving a specific capacitance of 188.36 F/g and demonstrating high salt adsorption capacity [46]. Additionally, a two-dimensional nanocomposite material combining molybdenum disulfide (MoS_2) and rGO was developed to improve the energy density of EDLCs. These nanocomposites achieved higher energy densities and slower discharge rates than typical rGO-based EDLCs, marking a significant advancement in supercapacitor technology [47]. Finally, highly condensed porous carbon materials derived from sulfonated pitch and GO were developed for use in EDLCs, focusing on achieving high voltage and high volumetric energy density [48]. The material underwent KOH activation and high-temperature treatment, resulting in a porous carbon with a significant specific surface area and high packing density. This material demonstrated excellent voltage characteristics, capable of handling up to 3.2 V in TEABF4/PC electrolyte and exhibited stable performance over 2000 cycles at high current density.

2. Methodology

2.1. Goal of the Systematic Review

As noted, there is an enormous adoption of GO or rGO in EDLCs. Hence, this systematic review aims to gather and evaluate current research on the roles of GO and rGO in improving the performance of EDLCs. Specifically, we examine synthesis methods, electrode fabrication techniques, the electrolytes used, and performance metrics to identify challenges and suggest future research directions. Unlike previous reviews, which have predominantly taken a narrative approach, this systematic review employs the PICOS and PRISMA frameworks to ensure a rigorous and comprehensive analysis of the existing literature. Particularly, narrative reviews, while valuable, often lack the structured methodology that is essential for minimizing bias and providing a clear, reproducible analysis of the literature. These reviews typically offer a broad overview of the topic, summarizing findings without a standardized protocol for data extraction and analysis. In contrast, a systematic review follows a predefined protocol, ensuring that all relevant studies are identified, appraised, and synthesized using transparent and replicable methods.

Then, the PICOS framework (Population, Intervention, Comparison, Outcomes, Study Design) [49] is instrumental in structuring the current systematic review. The research question guiding this systematic review is as follows:

- "What are the effects of using GO and rGO on the performance metrics of EDLCs?".

This question is addressed by considering the PICOS frameworks, as follows (see Table 1):

Table 1. Research question by PICOS framework.

P	Population	EDLCs
I	Intervention	The use of GO and rGO
C	Comparison	EDLCs using other materials or no GO/rGO
O	Outcomes	Performance metrics including capacitance, energy density, cycling stability, and other relevant electrochemical properties
S	Study Design	A systematic review of peer-reviewed articles, research papers, and conference papers

Otherwise, the PRISMA (Preferred Reporting Items for Systematic Reviews and Meta-Analyses) [50] guidelines further ensure the transparency and completeness of the review process. By adhering to these guidelines, this systematic review aims to provide a detailed and unbiased synthesis of the current evidence, identifying gaps in the literature and suggesting areas for future research. Additionally, by highlighting the challenges and limitations associated with the use of GO and rGO in EDLCs, this review aims to guide future research toward addressing these issues, ultimately advancing the field of energy storage.

This complete approach ensures that our systematic review, illustrated in Figure 1, encompasses the latest advancements and trends, providing a comprehensive and up-to-date synthesis of the evidence.

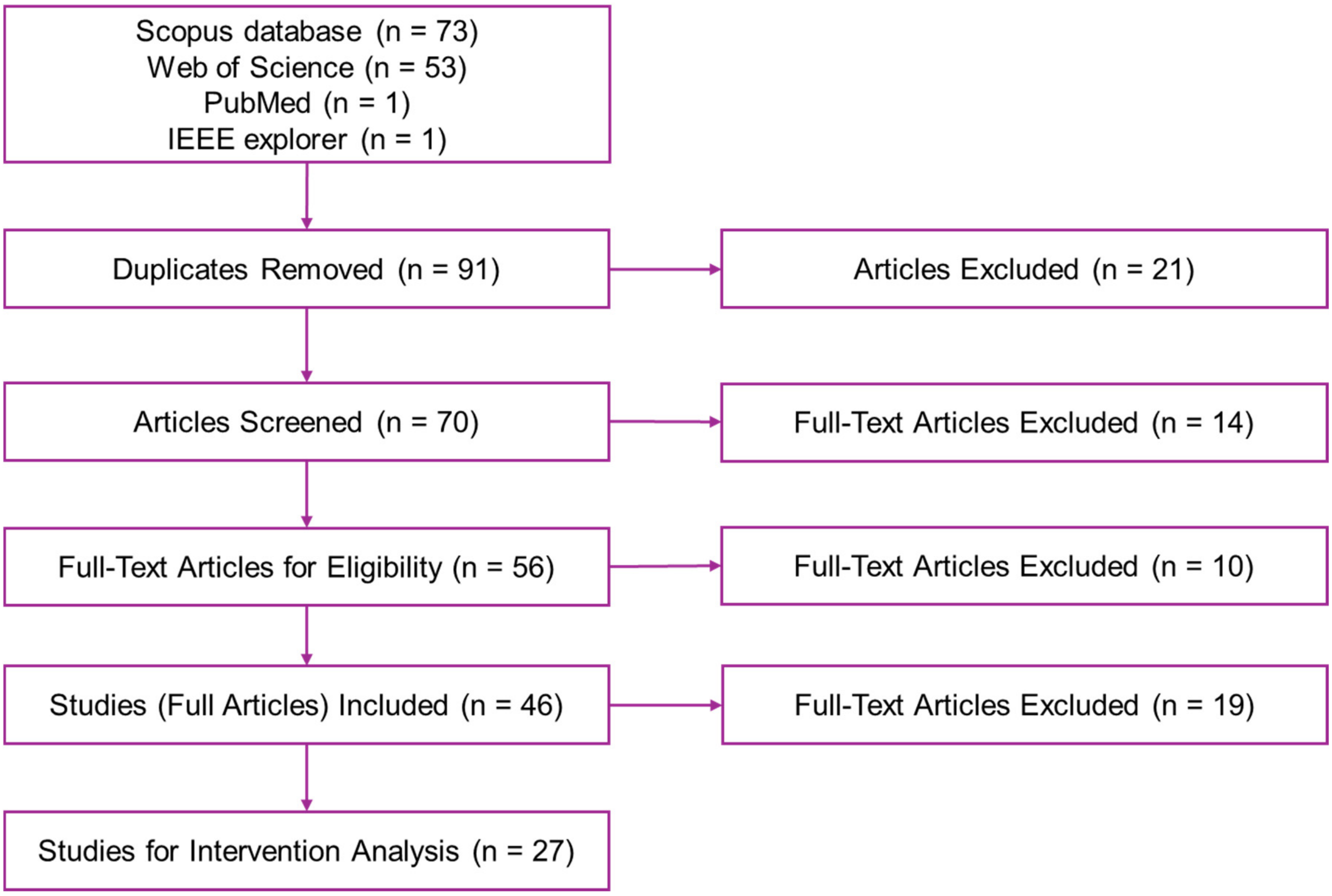

Figure 1. Illustration of the systematic review process.

2.2. *Identification Stage*

As denoted, the PRISMA guidelines help to maintain consistency and rigor in the review process, from the initial search strategy to the final synthesis of data within the timeframe of interest and database sources. The timeframe for this systematic review was set from 2010 to 2023 to capture the most relevant and recent advancements in the field of EDLCs incorporating GO and rGO. The year 2010 marks the beginning of significant research interest and technological advancements in graphene-based materials, making it an appropriate starting point for our review. Extending the search to 2023 ensures the inclusion of the latest studies, providing a comprehensive overview of the current state of research. To ensure a thorough search, four major scientific databases were selected: Scopus, Web of Science, PubMed, and IEEE Xplore. Each of these databases offers unique strengths that contribute to the robustness of the review [51–54].

Lastly, we aimed to identify and analyze studies that explore synthesis methods, electrode fabrication techniques, electrolytes involved, and performance metrics such as capacitance, energy density, and cycling stability. Keywords and Boolean operators were used to ensure a comprehensive search, encompassing all relevant studies within the specified timeframe from 2010 to 2023. The search query results are reported in Table 2, which provides an overview of the articles selected for this research effort.

Table 2. Summary of search queries and results across selected databases.

Database	Query	Results
Scopus	("Graphene Oxide" OR "Reduced Graphene Oxide") AND ("Electric Double Layer Capacitor" OR "Electric Double-Layer Capacitor")	73
Web of Sciences	("Graphene Oxide" OR "Reduced Graphene Oxide") AND ("Electric Double Layer Capacitor" OR "Electric Double-Layer Capacitor")	53
PubMed	("Graphene Oxide" OR "Reduced Graphene Oxide") AND ("Electric Double Layer Capacitor" OR "Electric Double-Layer Capacitor")	1
IEEE Xplore	("Graphene Oxide" OR "Reduced Graphene Oxide") AND ("Electric Double-Layer Capacitor" OR ("Electric Double Layer Capacitor")	1

2.3. Screening Stage

After conducting searches across four different databases, we initially identified a total of 128 articles: 73 from Scopus, 53 from Web of Science, 1 from IEEE Explorer, and 1 from PubMed. Following the removal of duplicates, 91 unique articles remained for further screening based on their titles and abstracts. The screening criteria included the following:

- Inclusion of only review articles, full articles, and proceedings papers;
- Focus on articles specifically about GO and rGO;
- Relevance to EDLC technology;
- Inclusion of articles in any language.

During the screening process, 21 articles were excluded, resulting in 70 articles moving forward to the next stage. The reasons for exclusion were as follows:

- Seventeen articles were not related to oxidized graphenes;
- Three articles did not pertain to EDLC technology;
- One article was not available.

2.4. Eligibility Stage

During the eligibility phase, the articles were randomly distributed among the authors for a full-text analysis. The eligibility criteria were as follows:

- The full text of the article is available in any language;
- The article focuses on the use of GO or rGO in supercapacitor technology;
- The article is centered on EDLC technology based on carbon materials;
- The article specifically discusses the application of GO or rGO in EDLC technology.

At this stage, 14 articles were excluded as ineligible, resulting in 56 articles being deemed suitable for inclusion and data extraction. The reasons for exclusion were as follows:

- Eight articles did not focus on EDLC technology and oxidized graphenes.
- Six articles were narrative review papers on the general topic of graphene-based supercapacitors.

2.5. Included Stage

Finally, the eligible articles were processed to extract all interventions that impact the performance of EDLC metrics. In this phase, 56 articles underwent thorough analysis. These works were evaluated based on the following metrics:

- Energy density;
- Capacitance;
- Power density;
- Cycling stability;
- Efficiency;
- Performance enhancement.

At this stage, 10 articles were taken as ineligible, resulting in 46 articles being deemed suitable for intervention extraction. The reasons for exclusion were as follows:

- Nine articles did not report capacitance or energy density;
- One article contained only theoretical predictions about electrode types with no reported metrics.

Additionally, out of these 46 articles, 19 were excluded because they did not report conclusive metrics such as capacitance, cycling stability, and efficiency. This resulted in 27 articles being selected for in-depth study to contextualize the role of GO or rGO in EDLCs.

3. Results

3.1. Summary of Search Results

Initially, a total of 128 articles were identified. After removing duplicates, 91 articles remained for further screening. Following the screening process, 70 articles were selected, and after the eligibility stage, 56 articles were considered suitable. Then, 27 review articles were included in the final data extraction.

From these 27 articles, parameters and related interventions were extracted to contextualize the role of GO and rGO in EDLCs, as well as their measured impacts on various performance metrics (see Tables S1–S3). The details of the interventions were categorized into several key areas, as follows:

- Type of intervention describes the specific approach or modification applied;
- Preparation method details the synthesis and preparation techniques used for GO and rGO;
- Electrode fabrication discusses the methods and materials used in the creation of electrodes;
- Electrolyte used specifies the types of electrolytes employed in the studies;
- Comparison details provide a comparison of the methods and materials used across different studies;
- Results and findings summarize key results, including statistical significance and cycling stability.

On the other hand, the impact on performance metrics refers to observed changes or no changes in the following:

- Capacitance is the measurement of the ability to store charge;
- Energy density is the amount of energy stored per unit volume or mass;
- Power density is the rate at which energy can be delivered;
- Efficiency is the overall performance efficiency of the EDLCs.

This detailed extraction and categorization provides a comprehensive understanding of how GO and rGO influence EDLC performance, offering insights into the most effective approaches and highlighting areas for future research.

3.2. Interventions Outcomes

Tables 3 and S1–S3 report the complete information of 27 articles considered for the intervention stage. In particular, Table 3 provides a comprehensive summary of various interventions and their impact on performance metrics in the development of supercapacitors based on GO or rGO. The interventions include the use of different materials and synthesis methods to enhance key metrics such as capacitance, energy density, power density, and efficiency. Several studies highlighted the potential of graphene-based composites in improving supercapacitor performance. For instance, the development of polymer/graphite oxide composites and superparamagnetic magnetite nanocrystals combined with graphene oxide demonstrated notable enhancements in capacitance and charge–discharge capabilities. The incorporation of nitrogen doping and other functional groups into graphene materials significantly improved capacitance, energy, and power densities while maintaining high retention rates after numerous cycles.

Table 3. Summary of interventions and the impact on performance metrics.

Reference	Type of Intervention:	Capacitance	Energy Density	Power Density	Efficiency
Gutierreza et al., 2013 [55]	Development of a scalable prototype supercapacitor using metal oxides and graphene oxide with an ionic liquid electrolyte to combine pseudocapacitance and electric double-layer capacitance.	0.356 F/g, 117 F/g	Not reported	Not reported	Not reported
Lee et al., 2012 [56]	Research on supercapacitors, specifically focusing on the synergy between graphene nanosheets and the conducting polymer polyaniline.	402.5–219.4 F/g (PG10), 489.0–304.8 F/g (PG10-H), 240.1–103.5 F/g (PG10-HC), 24.6–10.9 F/g (GG10-H)	18.92 Wh/kg (PG10), 30.34 Wh/kg (PG10-H)	1.0 kW/kg	90.7% retention of initial capacitance after 500 cycles (PG10-H)
Tran et al., 2013 [57]	Development of new poly sodium 4-styrenesulfonate intercalated graphene oxide electrodes for supercapacitors	88 F/g (new composite), 109 F/g (PSSGO5)	Not reported	Not reported	94% retention of initial capacitance after 3000 cycles
Huang et al., 2014 [58]	Development of new gel electrolyte for EDLCs	141.8 F/g	Not reported	Not reported	Not reported
Gao et al., 2015 [59]	Development of a high-power density electric double-layer capacitor using porous multi-walled carbon nanotube microspheres as a local electrolyte micro-reservoir	118 mF/cm^2, 136 mF/cm^2	9 µWh/cm^2	1540 mW/cm^2	98% retention after 5000 cycles.
Gupta et al., 2015 [60]	Development of graphene/polymer hybrid thin films as supercapacitors, utilizing physical–chemical interfacial processes for enhanced electrochemical performance.	270–350 F/g (for hybrids with ErGO), 210–300 F/g (for hybrids with GO), 10–20 F/g (for PPy and PAni), 70–80 F/g (for GO and ErGO)	Not reported	Not reported	Not reported
Haque et al., 2015 [61]	Development of nitrogen-doped graphene via thermal treatment of graphene oxide and aminoterephthalic acid for use in supercapacitors	210 F/g (at 1 A/g), 226 F/g (from CV)	Not reported	Not reported	>90% capacity retention after 5000 cycles.
Ming Li et al., 2015 [62]	Development of N-doped reduced graphene oxide (N-rGO) using a novel method for enhanced performance in supercapacitors	234.3 F/g (in 0.5 M H_2SO_4), 187.8 F/g (in 1 M TEABF4/PC)	25.1 Wh/kg	10 kW/kg	97.9% capacitance retention after 730 cycles
Suleman et al., 2015 [63]	Development of high-rate supercapacitive EDLCs using GO and r-GO electrodes interfaced with a plastic-crystal-based gel polymer electrolyte (GPE)	66 F/g (GO), 60 F/g (r-GO)	18 Wh/kg (GO), 15.6 Wh/kg (r-GO)	33.3 kW/kg (GO), 54.9 kW/kg (r-GO)	Stable up to 11,000–13,500 charge–discharge cycles
Tran et al., 2015 [64]	Development of a one-pot synthesis method for a graphene/glucose/nickel oxide composite for use in supercapacitors	Up to five times higher than that of reduced GO, about 300 F/g	Not reported	Not reported	Maintains the same specific capacitance up to 5000 cycles
Youn et al., 2015 [65]	Development of high-surface-area N-RGO for use in EDLCs	291 F/g (at 1 A/g), 261 F/g (at 50 A/g)	91.0 Wh/kg	75.0 kW/kg	96% capacitance retention after 100,000 cycles

Table 3. *Cont.*

Reference	Type of Intervention:	Capacitance	Energy Density	Power Density	Efficiency
Muhammed et al., 2016 [66]	Development of graphene oxide–MnO_2 nanocomposite for use in supercapacitors	305 F/g for GO, 545 F/g for GO-MnO_2 composite	91.0 Wh/kg	75.0 kW/kg	96% capacitance retention after 100,000 cycles
Wang et al., 2017 [67]	Development of a graphene-based lithium-ion capacitor (LIC) with enhanced gravimetric energy and power densities	Not reported	Up to 200 Wh/kg.	10 kW/kg	70% capacity retention after 5000 cycles.
Ma et al., 2019 [68]	Development and electrochemical characterization of graphene-based electrodes with different fabrication parameters for supercapacitors	2.12 F/g	244.46 mWh/kg	54.6 W/kg	Not reported
Sengottaiyan et al., 2017 [69]	Development of cobalt oxide/reduced graphene oxide (Co_3O_4/rGO) composites for supercapacitors using a one-pot hydrothermal synthetic route without surfactants	487 F/g	Not reported	Not reported	96.6% capacitance retention after 2000 cycles
Zhang et al., 2017 [70]	Synthesis of a ternary composite of graphene oxide/carbon dots/polypyrrole (GO/CDs/PPy) for supercapacitor applications	576 F/g	30.1 Wh/kg	250 W/kg	92.9% capacitance retention after 5000 cycles
Hu et al., 2018 [71]	Development of a 3D hierarchical porous $V_3O_7 \cdot H_2O$ nanobelts/CNT/reduced graphene oxide composite for supercapacitors	657 F/g	34.3 Wh/kg	3000 W/kg	99.7% capacitance retention after 10,000 cycles
Noh, et al., 2019 [72]	Development and investigation of SnO_2 nanospacer-incorporated reduced graphene oxide electrodes for enhanced supercapacitor performance	138.4 F/g for RGO–SnO_2–NR, 78.4 F/g for RGO–SnO_2–NP, 70.0 F/g for bare RGO	4.17 Wh/kg for RGO–SnO_2–NR.	123.0 W/kg for RGO–SnO_2–NR	Not reported
Li et al., 2019 [73]	Development of SnO_2 nanospacer-incorporated reduced graphene oxide electrodes for improved capacitive energy storage	6.12 F/g	0.48 Wh/kg	371 W/kg	99% capacitance retention after 2400 cycles
Bagher et al., 2020 [74]	Functionalization of highly rGO with butyl methyl imidazolium ionic liquid to enhance supercapacitive features	436.7 F/g for IFG, 262.5 F/g for RGO	Not reported	Not reported	Not reported
Kil et al., 2020 [75]	Development of rGO electrodes for supercapacitors using a solution-processed method with low-temperature thermal reduction	Not reported	186 Wh/kg at 142 W/kg, 100 Wh/kg at 10 kW/kg.	10 kW/kg	70% capacity retention after 5000 cycles

Table 3. *Cont.*

Reference	Type of Intervention:	Capacitance	Energy Density	Power Density	Efficiency
Maphiri, et al., 2021 [76]	Development of thermally reduced graphene oxide microsupercapacitors using a mask-free AxiDraw direct writing technique	The highest areal capacitance of 0.5421 mF/cm^2 at 10 mV/s for TRGO-200.	Volumetric energy density of 14.61 mWh/cm^3 for TRGO-500.	Volumetric power density of 142.67 mW/cm^3 for TRGO-500.	Capacitance retention and coulombic efficiency of 95% and 100%, respectively, at a current density of 0.83 µA/cm^2 for 4000 cycles.
Lun Wu et al., 2021 [77]	Development of rGO/CoSx-rGO/rGO hybrid films using a novel method involving co-assembly and sulfidation of 2D metal organic framework (MOF) nanoflakes and graphene oxide	375 F/g	31.68 Wh/kg	6750 W/kg	96.6%
Zhao et al., 2021 [78]	Development of nanocarved vanadium nitride (VN) nanowires encapsulated in lamellar graphene layers for supercapacitor electrodes	222 F/g at 0.5 A/g, 65 F/g at 10 A/g	Not reported	Not reported	Not reported
Qiu et al., 2015 [79]	Development of gel polymer electrolytes (GPEs) using GO-decorated polymers for enhanced performance in EDLCs under extreme temperature conditions	40–75 F/g (from CV curves), ~100 F/g at a current density of 0.1 A/g	3.52 Wh/kg	0.44 kW/kg	Not reported
Ngamjumrus, et al., 2023 [80]	Development of a supercapacitor using hydrothermal and 3-D ball milling processes to improve the properties of reduced graphene oxide activated durian shell carbon composites	545.78 F/g (highest), 65.585 F/g (coin cell)	60.834 Wh/kg (highest), 5.123 Wh/kg (coin cell)	260.834 W/kg (highest), 47.286 W/kg (coin cell)	Not reported
Qu et al., 2023 [81]	Development of hybrid supercapacitor electrodes using an optimized synthetic route for $Cu_{0.33}Co_{0.67}Se_2$ nanorods on Ni foam integrated with N, S co-doped porous carbon (NSPC)	~458 mAh/g at 1 A/g	~41.5 Wh/kg	~801.5 W/kg	Not reported

Several fabrication techniques such as laser holography reduction of graphene oxide and the use of fractal designs in laser-scribed graphene for micro-supercapacitors have been shown to increase the electrochemically active surface area, thereby enhancing capacitance and energy density. Furthermore, the integration of graphene with materials like nickel hydroxide, molybdenum disulfide, and vanadium nitride has led to substantial improvements in electrochemical performance, demonstrating the versatility and potential of graphene-based materials in supercapacitor applications. Moreover, advancements in the development of high-power density electric double-layer capacitors using porous carbon nanotubes and the creation of gel polymer electrolytes decorated with GO for extreme temperature conditions underscore the importance of material selection and processing techniques. These studies have shown that specific combinations of materials and innovative synthesis methods can lead to significant improvements in performance metrics such as capacitance, energy density, and power density.

Sustainability has also been a focus in several interventions, such as using agricultural wastes for synthesizing rGO and developing graphene-based lithium-ion capacitors with enhanced energy and power densities. These approaches not only improve the performance of supercapacitors but also align with sustainable manufacturing practices, highlighting the potential for environmentally friendly energy storage solutions. In general, Table 3 highlights that various materials and synthesis methods can lead to significant enhancements in supercapacitor performance. The detailed discussion in Section 4 will delve deeper into these findings, providing a thorough analysis of the observed trends and their implications for future research and applications in energy storage technologies.

3.3. Data Analysis

The analysis of data from the 27 selected works in Tables 3 and S1–S3 reveals a varied consideration of performance metrics, specifically capacitance, energy density, power density, and efficiency. Notably, not all works simultaneously consider all four metrics, as illustrated in Figure 2. Indeed, Figure 2 shows the number of papers that reported (black bars) versus those that did not report (gray bars) each of the four key performance metrics. The observed results suggest a pattern where capacitance (23 articles) and efficiency (25 articles) are the most frequently reported metrics among the reviewed works. This can be attributed to the direct relationship of capacitance with the fundamental charge storage capability of the supercapacitors, making it a critical parameter for initial characterization. Efficiency, on the other hand, is crucial for evaluating the overall energy utilization and performance stability of the devices.

Energy density, while important (18 articles), may be less frequently reported due to the complexity involved in its accurate measurement and the additional experimental setups required. Furthermore, some studies might focus primarily on enhancing capacitance or efficiency, leaving energy density as a secondary concern. Power density is reported in fewer studies (13 articles), potentially due to the specific application focus of some research. High power density is essential for applications requiring rapid energy discharge, but it might not be a primary objective for studies aimed at optimizing other aspects like cycling stability or electrode material properties.

Figure 3 reveals that not all studies included both materials in their experimentation. Specifically, 4 articles focused on GO, 22 articles focused on rGO, and only 1 article used both GO and rGO for comparison. The observed results suggest a clear preference for rGO over GO in EDLC research. This preference can be attributed to several factors:

- rGO typically offers higher electrical conductivity than GO, making it more suitable for applications requiring efficient charge and discharge cycles, which is critical for supercapacitors;
- rGO tends to have a suitable surface area compared to GO, which enhances its ability to store charge, thus improving the capacitance and overall performance of supercapacitors;
- rGO generally shows better electrochemical stability, making it more reliable for long-term applications in energy storage devices.

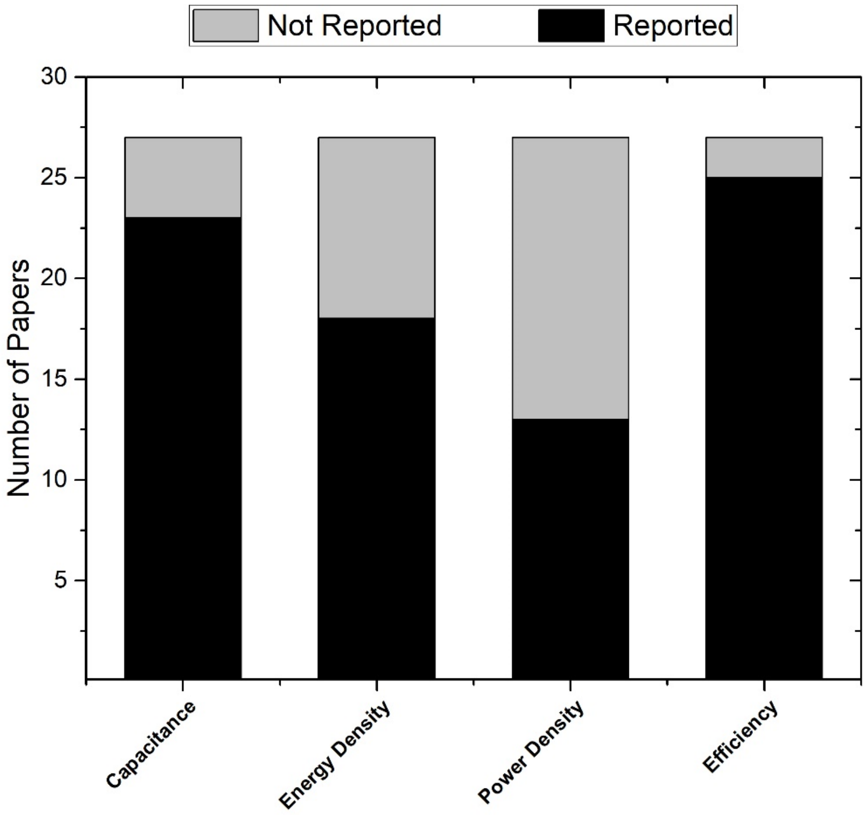

Figure 2. Analysis of reported and non-reported metrics in research papers on the application of GO and rGO in EDLCs.

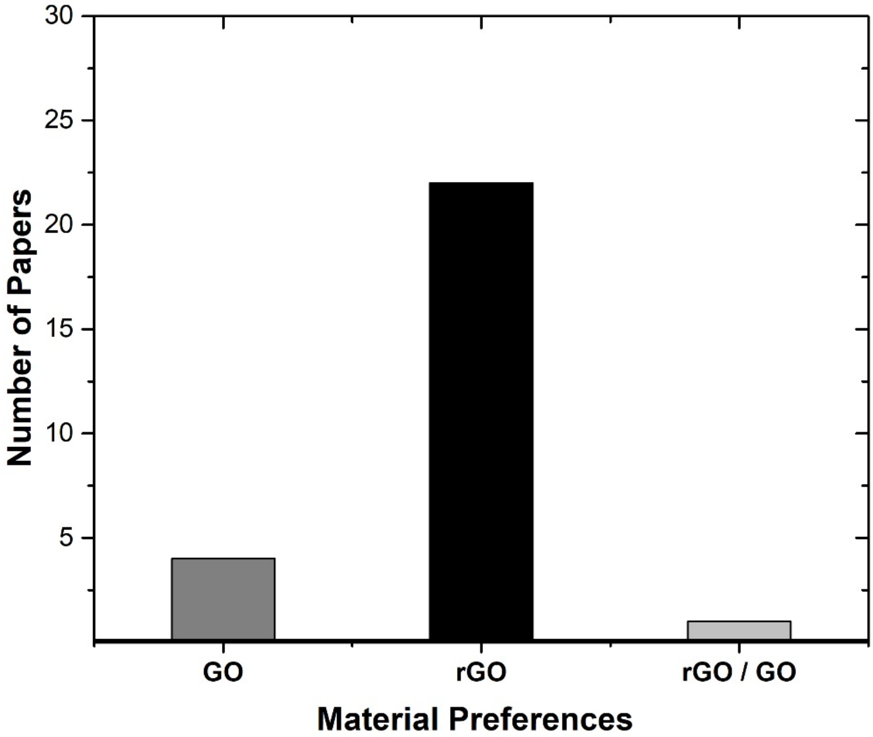

Figure 3. Prevalence of studies focusing on the use of GO vs. rGO.

The single study that used both GO and rGO for comparison aimed to directly evaluate the performance differences between these two materials. This approach provides valuable insights but is less common, likely due to the additional complexity and resources required for such comparative studies. The relatively lower number of studies focusing on GO alone might be due to its lower conductivity and stability compared to rGO.

3.4. Generated Data for PICOS PRISMA Process

To ensure transparency and facilitate further research, we have carefully documented every step of our data generation process in this systematic review. This detailed documentation is illustrated in Figure 4, which maps out the entire review process from the initial identification of articles to the final data extraction. The step-by-step flowchart covers the following stages:

- Identification collects initial articles from databases such as Scopus, Web of Science (WoS), IEEE Explorer, and PubMed;
- Screening review titles and abstracts to filter out irrelevant studies;
- Eligibility assesses full-text articles based on specific inclusion criteria;
- Inclusion selects studies that meet all the criteria for the final review;
- Extraction extracts detailed data from the included studies for thorough analysis.

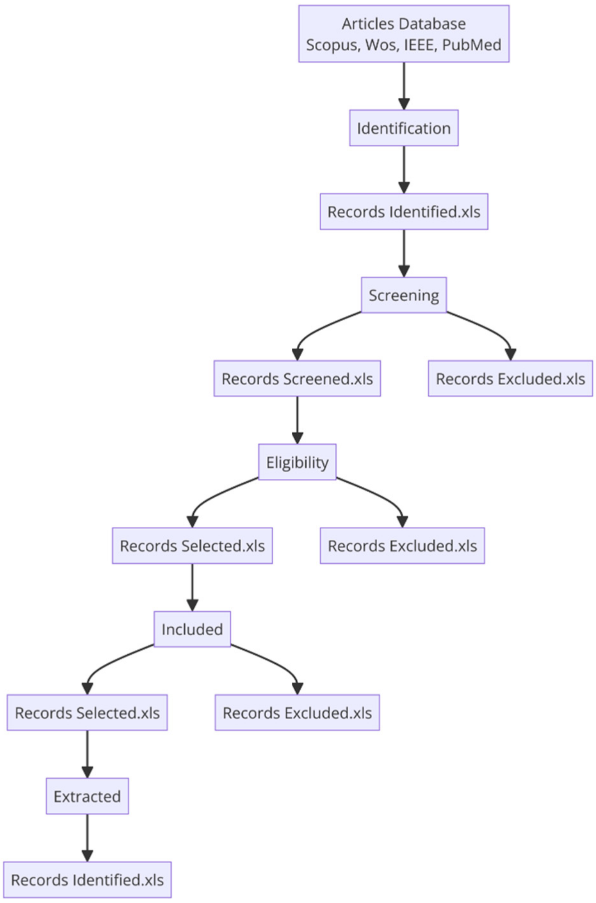

Figure 4. Generated data on the current systematic review process. Available at: https://doi.org/10.17605/OSF.IO/HRKVD (accessed on 13 July 2024).

Each stage includes tasks like identifying records, screening for relevance, selecting eligible studies, and excluding those that do not meet the criteria. This thorough approach ensures a comprehensive and rigorous review process. To make our methods transparent and reproducible, we have made all this information accessible for detailed examination at the following link: https://doi.org/10.17605/OSF.IO/HRKVD (accessed on 13 July 2024).

4. Discussion

4.1. Preparation Methods Suitable in EDLCs

EDLCs are promising energy storage devices due to their high-power density, rapid charge/discharge cycles, and long cycle life. The integration of GO and rGO into EDLCs has garnered significant attention for enhancing their performance metrics. We discuss the most promising preparation strategies, focusing on scalable preparation methods and their implications for realistic applications. Gutierrez et al. (2013) [55] developed a scalable prototype supercapacitor that combines pseudocapacitance and electric double-layer capacitance using metal oxides and graphene oxide. The preparation method involved the modified Hummers' method for synthesizing graphene oxide, ammonium evaporation for Co_3O_4 nanorods, and acid digestion for MnO_2 nanowires. This approach offers a balanced enhancement in capacitance and energy density, making it a viable option for large-scale production. The combination of GO with metal oxides enables the dual benefits of high energy storage capacity and stability, crucial for industrial applications. Lee et al. (2012) [56] explored the synergy between graphene nanosheets and the conducting polymer polyaniline (PANi) for supercapacitors. They employed a layer-by-layer (LbL) assembly method to create multilayer thin films through electrostatic interactions between negatively charged GO nanosheets and positively charged PANi. This method is relatively simple and scalable, offering a pathway to fabricate high-performance electrodes with enhanced electrochemical properties. The LbL assembly's adaptability to various materials further supports its potential for industrial-scale production.

Tran et al. (2013) [57] focused on the development of new poly sodium 4-styrenesulfonate intercalated graphene oxide electrodes for supercapacitors. By controlling the oxidation time during the synthesis of graphite oxide, they could modify the oxygen content in GO, directly affecting the electric double-layer capacitance. This controlled synthesis process is scalable and allows for the customization of electrode properties to meet specific application needs, enhancing the practical applicability of GO-based EDLCs. Gao et al. (2015) [59] developed a high-power density EDLC using porous multi-walled carbon nanotube (MWCNT) microspheres as a local electrolyte micro-reservoir. The spray drying method used to prepare these microspheres is highly scalable and capable of producing large quantities of consistent material. The resulting microspheres provide a stable porous structure, beneficial for high-current density operations. This innovation not only enhances the power density but also supports the feasibility of mass production, making it suitable for commercial energy storage solutions.

Haque et al. (2015) [61] introduced nitrogen-doped graphene for supercapacitors through the thermal treatment of graphene oxide with aminoterephthalic acid at 750 °C under argon flow. Nitrogen doping significantly improves the electrical conductivity and electrochemical performance of graphene, making it highly efficient for energy storage applications. The thermal treatment process is scalable and can be integrated into existing manufacturing setups, ensuring the practical implementation of this approach in commercial supercapacitor production. Suleman et al. (2015) [63] developed high-rate supercapacitive EDLCs using GO and rGO electrodes interfaced with a plastic-crystal-based gel polymer electrolyte (GPE). The GPE film fabrication involved lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) in succinonitrile (SN) within a polyvinylidene fluoride-co-hexafluoropropylene (PVdF-HFP) matrix. This approach enhances the interface stability and performance of GO and rGO electrodes. The scalability of this fabrication process, coupled with the improved electrochemical performance, makes it a promising candidate for developing flexible and high-performance energy storage devices.

Youn et al. (2015) [65] developed high-surface-area N-RGO for EDLCs through a two-step method involving solid-state microwave irradiation followed by heat treatment under NH_3 gas. This combination allows for efficient nitrogen doping and surface area enhancement, crucial for high-performance supercapacitors. These scalable methods improve the material's suitability for real-world energy storage applications, offering both high energy density and durability. Muhammed et al. (2016) [66] synthesized a graphene oxide–MnO_2 nanocomposite for supercapacitors using Hummer's method for GO and a soft chemical route for MnO_2. The nanocomposite leverages the high conductivity of graphene oxide and the high capacitance of MnO_2, offering superior energy storage capabilities. The scalable synthesis methods employed ensure the feasibility of large-scale production, supporting the practical implementation of these high-performance materials in commercial supercapacitors.

Wang et al. (2017) [67] developed a graphene-based lithium-ion capacitor (LIC) with enhanced gravimetric energy and power densities. The synthesis involved creating a 3D macroporous foam from reduced rGO decorated with tin oxide nanoparticles for the anode, and thermally expanded and physically activated rGO (a-TEGO) for the cathode. This 3D macroporous structure enhances ion transport and electrode stability, making it suitable for high-performance LICs. The preparation methods are scalable, supporting potential industrial applications and offering a robust solution for advanced energy storage systems. Zhao et al. (2021) [78] developed nanocarved vanadium nitride (VN) nanowires encapsulated in lamellar graphene layers for supercapacitor electrodes. The hierarchical porous structure and encapsulation of VN nanowires within graphene layers improve electrochemical performance. Synthesized via a freeze-casting process followed by NH_3 nitridation, these materials demonstrate significant potential for scalability and large-scale production. This innovation enhances both energy density and cycling stability, addressing key requirements for practical supercapacitor applications.

To further emphasize these opportunities, Table 4 provides an overview of the key studies. It highlights various innovative approaches, including scalable supercapacitor prototypes, layer-by-layer assembly, controlled oxidation processes, and advanced thermal treatments. Each method is evaluated based on its potential for scalability and practical applications, demonstrating improvements in performance metrics such as energy density, stability, and conductivity, thereby supporting the feasibility of these techniques for industrial-scale production and realistic energy storage solutions.

Table 4. Key studies on preparation methods of GO and rGO for EDLCs.

Reference	Intervention Type	Preparation Method	Scalability and Application
Gutierrez et al. (2013) [55]	Scalable supercapacitor	Modified Hummers' method, ammonium evaporation, acid digestion	Mass production
Lee et al. (2012) [56]	Synergy in supercapacitors	Layer-by-layer assembly	Industrial scale
Tran et al. (2013) [57]	GO electrodes for supercapacitors	Controlled oxidation time	Customizable, practical
Gao et al. (2015) [59]	High-power density EDLC	Spray drying of MWCNT microspheres	Mass production
Haque et al. (2015) [61]	Nitrogen-doped graphene	Thermal treatment at 750 °C	Improved performance, existing setups
Suleman et al. (2015) [63]	High-rate EDLCs	Fabrication of GPE film	Flexible, high-performance
Youn et al. (2015) [65]	High-surface-area N-doped rGO	Microwave irradiation, NH_3 heat treatment	High-performance
Muhammed et al. (2016) [66]	GO-MnO_2 nanocomposite	Hummers' method for GO, soft chemical route for MnO_2	Superior energy storage
Wang et al. (2017) [67]	Graphene-based LIC	3D macroporous foam from rGO, SnO_2 nanoparticles	Industrial applications
Ma et al. (2019) [68]	VN nanowires in graphene layers	Freeze-casting, NH_3 nitridation	Practical supercapacitor

Additionally, we provide a heatmap score analysis in Figure 5. Data were extracted from Table 4. Each study was evaluated for its scalability and application potential, and these evaluations were translated into scores on a scale from 1 to 10. The scores were assigned based on a qualitative and quantitative analysis of each study. Scalability scores reflected the feasibility of mass production and industrial-scale implementation, considering factors such as method simplicity, material availability, cost efficiency, and integration with existing processes. Scores ranged from 7 to 9, reflecting relative strengths and limitations. No study received a perfect score of 10, as all methods had some limitations. Similarly, no study scored below 7, as all included studies demonstrated a reasonable level of promise for scalability and application. Specifically, Figure 5 shows the comparative analysis of various preparation methods for GO and rGO in enhancing EDLCs. By providing a clear visual summary of scalability and application scores, the heatmap accentuates the findings discussed earlier. Studies with higher scores, such as those by Gutierrez et al. (2013) [55] and Gao et al. (2015) [59], illustrate the significant potential of scalable methods that leverage straightforward, cost-effective processes. These methods not only facilitate mass production but also demonstrate substantial improvements in energy density and cycling stability, making them suitable for practical applications. The clustering of scores around the higher end of the spectrum (7–9) indicates a general trend towards effective and scalable preparation techniques. This graphical representation reinforces the discussion that innovative yet simple methods hold the key to advancing the practical deployment of EDLCs, bridging the gap between laboratory research and industrial-scale implementation.

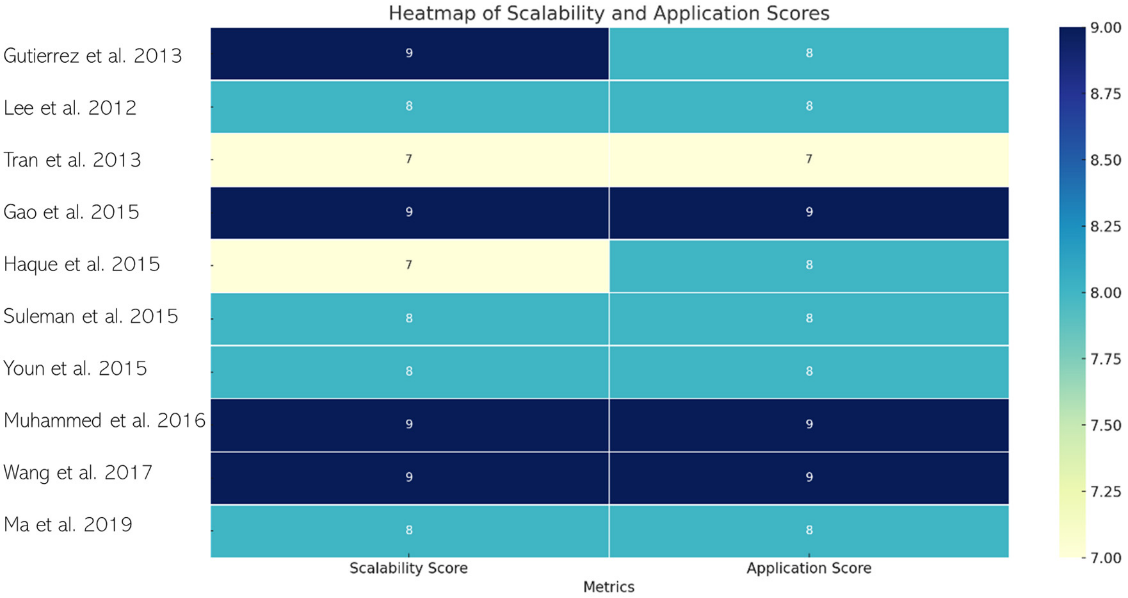

Figure 5. Heatmap of scalability and application scores for preparation methods.

4.2. Comparison of Electrodes and Electrolytes for EDLCs

The analysis of electrode fabrication and electrolytes used in various studies provides deeper insights into the most promising methods for enhancing EDLCs. Gutierrez et al. (2013) [55] employed a tri-layered composite electrode comprising Co_3O_4 nanorods, MnO_2 nanowires, and electrophoretically deposited graphene oxide reduced to rGO. The electrolyte used, a combination of KOH, acetonitrile, and ionic liquid EMIm-BF4, supports high ionic conductivity and stability, making it suitable for scalable and practical applications in supercapacitors. Lee et al. (2012) [56] fabricated electrodes using a combination of GO and PANi in a multilayer structure through layer-by-layer (LbL) assembly. Sulfuric acid (H_2SO_4) was used as an electrolyte, known for its high ionic conductivity. The multilayer structure enhances the electrochemical performance of the electrode, and the use of H_2SO_4 ensures efficient charge transfer.

Tran et al. (2013) [57] developed electrodes from a new composite with reduced oxygen content in graphene oxide. While the specific composition of the aqueous electrolyte was not detailed, the use of an aqueous medium indicates a focus on practicality and safety. Gao et al. (2015) [59] utilized multi-walled carbon nanotube (MWCNT) microspheres as micro-reservoirs for electrode fabrication. The robust and stable porous structure is beneficial for high-current-density operations. The KOH aqueous electrolyte used in their tests provides excellent ionic conductivity and stability, supporting the high scalability and application scores attributed to this study.

Haque et al. (2015) [61] developed nitrogen-doped graphene electrodes prepared by mixing the active material with PTFE binder and applying it to gold electrodes. The use of 0.5 M H_2SO_4 as an electrolyte significantly improves electrochemical performance and conductivity. The combination of nitrogen doping and a strong acid electrolyte enhances the effectiveness of the electrode, supporting its high scalability and practical application potential. Suleman et al. (2015) [63] created high-rate supercapacitive EDLCs using GO and r-GO electrodes interfaced with a plastic-crystal-based gel polymer electrolyte (GPE). The GPE, comprising LiTFSI in SN entrapped in PVdF-HFP, enhances interface stability and performance. This innovative approach to electrode–electrolyte interfacing supports its high scalability and application scores, indicating strong potential for flexible, high-performance energy storage devices.

Youn et al. (2015) [65] prepared high-surface-area N-rGO electrodes using solid-state microwave irradiation followed by NH_3 gas heat treatment. The use of tetraethylammonium tetrafluoroborate in acetonitrile as the electrolyte improves electrochemical performance by enhancing ionic conductivity and stability. Muhammed et al. (2016) [66] synthesized a graphene oxide–MnO_2 nanocomposite using a soft chemical route, providing a novel approach to combining these materials for enhanced performance. The electrodes, tested with 1 M Na_2SO_4 in aqueous solution, demonstrate significant improvements in energy storage capabilities. The combination of a high-capacity nanocomposite electrode and a stable aqueous electrolyte supports its promise for scalable and practical supercapacitor applications.

Wang et al. (2017) [67] developed a graphene-based lithium-ion capacitor (LIC) with anodes made from SnO_2–rGO and cathodes from a-TEGO, incorporating advanced graphene technology. The use of 1 M LiPF6 in ethylene carbonate and dimethyl carbonate (EC) as the electrolyte enhances the performance metrics, supporting the high scalability and application scores. Zhao et al. (2021) [78] developed electrodes with nanocarved vanadium nitride (VN) nanowires encapsulated in lamellar graphene layers, enhancing structural integrity and electrochemical performance. Using 1 M KOH as the electrolyte, this study demonstrates excellent practical applicability. The scalable freeze-casting process and significant performance improvements justify its high scores.

To remark on these outcomes, Table S1 consolidates the key aspects of electrode fabrication and electrolyte use across all works analyzed. This comparison strengthens the discussion by highlighting the diverse yet effective approaches employed. The consistent use of high-conductivity electrolytes, such as H_2SO_4 and KOH, further emphasizes the focus on achieving superior energy storage capabilities, making these methods promising for realistic deployment. On the other hand, Figure 6 provides a clear visual representation of the scalability and application scores, emphasizing why these methods are considered the most promising (Figure 5) now in the context of electrodes and electrolytes used. Similarly, the clustering of scores around the higher end of the spectrum (7–9) highlights the overall effectiveness. This graphical representation shows again that advanced electrode fabrication techniques combined with high-conductivity electrolytes are key to advancing EDLC technology, bridging the gap between research and industrial-scale implementation.

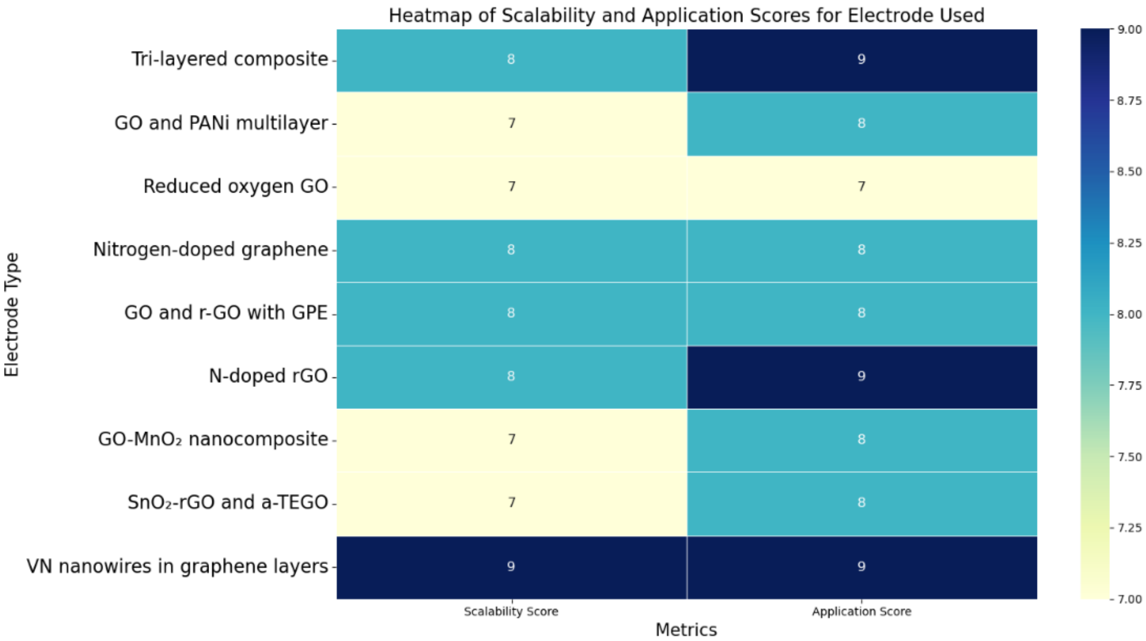

Figure 6. Heatmap of scalability and application scores for electrodes used in EDLCs.

4.3. Commonly Materials and Methods Used

The comparative analysis presented in (Tables 5 and S2) offers a comprehensive overview of various studies examining the role of GO and rGO in EDLCs. The recurring use of cyclic voltammetry, galvanostatic charge–discharge, and impedance spectroscopy across numerous studies highlights these techniques as standard methodologies for evaluating the electrochemical performance of GO and rGO-based materials. Several studies, such as Lee et al. (2012) [56], Tran et al. (2013) [57], and Noh et al. (2019) [72], have demonstrated significant improvements in capacitance and stability when using GO and rGO composites. The incorporation of these materials often leads to higher specific capacitance and better retention over extended cycles.

Table 5. Summary of comparison materials and methods for supercapacitors based on GO and rGO.

Reference	Comparison Materials	Comparison Method
Lee et al., 2012 [56]	GO, PANi, combinations	Cyclic voltammetry, galvanostatic charge–discharge, impedance spectroscopy
Tran et al., 2013 [57]	GO, PSSGO, modified PSSGO	Cyclic voltammetry, galvanostatic charge–discharge, impedance spectroscopy
Suleman et al., 2015 [63]	GO, r-GO electrodes	Cyclic voltammetry, galvanostatic charge–discharge, EIS
Tran et al., 2015 [64]	RGO, RGO/nickel oxide	Cyclic voltammetry, charge/discharge cycling, impedance spectroscopy
Youn et al., 2015 [65]	rGO, other graphene materials	Cyclic voltammetry, galvanostatic charge–discharge, impedance spectroscopy
Muhammed et al., 2016 [66]	GO, MnO_2 composite	Cyclic voltammetry, galvanostatic charge–discharge
Noh et al., 2019 [72]	SnO_2 nanorod bundles, SnO_2 nanoparticles, rGO	Cyclic voltammetry, galvanostatic charge–discharge, impedance spectroscopy
Li et al., 2019 [73]	rGO electrodes without SnO_2	Cyclic voltammetry, galvanostatic charge–discharge, impedance spectroscopy

Table 5. *Cont.*

Reference	Comparison Materials	Comparison Method
Bagher et al., 2020 [74]	Functionalized rGO, non-functionalized rGO	Cyclic voltammetry, impedance spectroscopy
Kil et al., 2020 [75]	rGO preparation methods	Cyclic voltammetry, galvanostatic charge–discharge, impedance spectroscopy
Maphiri et al., 2021 [76]	Microsupercapacitors at different levels of thermal reduction	Cyclic voltammetry, galvanostatic charge–discharge, impedance spectroscopy

The combination of GO or rGO with other materials, as seen in studies by Tran et al. (2015) [64] and Muhammed et al. (2016) [66], suggests that synergistic effects can further enhance the electrochemical properties. For instance, the use of rGO with nickel oxide or MnO_2 composites not only improves the capacitance but also the cycling stability, demonstrating the versatility of these materials in forming hybrid structures. Studies such as those by Bagher et al. (2020) [74] and Kil et al. (2020) [75] explore the impact of functionalization and processing techniques on rGO. Functionalized rGO shows significant improvements in electrochemical characteristics, indicating that surface modifications can play a critical role in optimizing performance. The durability of these materials is also a focal point, with studies such as Youn et al. (2015) [65] and Maphiri et al. (2021) [76] reporting excellent cycling stability over thousands of cycles. These results are crucial for the long-term applicability of GO and rGO in commercial supercapacitors as they ensure consistent performance over prolonged periods.

Figure 7 illustrates the most common comparison methods used across the reviewed studies. Cyclic voltammetry is the predominant method, featured in 11 of the studies, followed closely by galvanostatic charge–discharge, used in 8 studies, and impedance spectroscopy, used in 8 studies. These methods are crucial for assessing the electrochemical performance of supercapacitors, including their specific capacitance, energy density, and power density. Cyclic voltammetry is a vital technique that allows for the rapid assessment of the electrochemical behavior of materials by measuring the current response of an electrode to a linearly changing voltage. By employing cyclic voltammetry, one can obtain valuable information about the redox processes, electrode kinetics, and overall electrochemical stability of the materials.

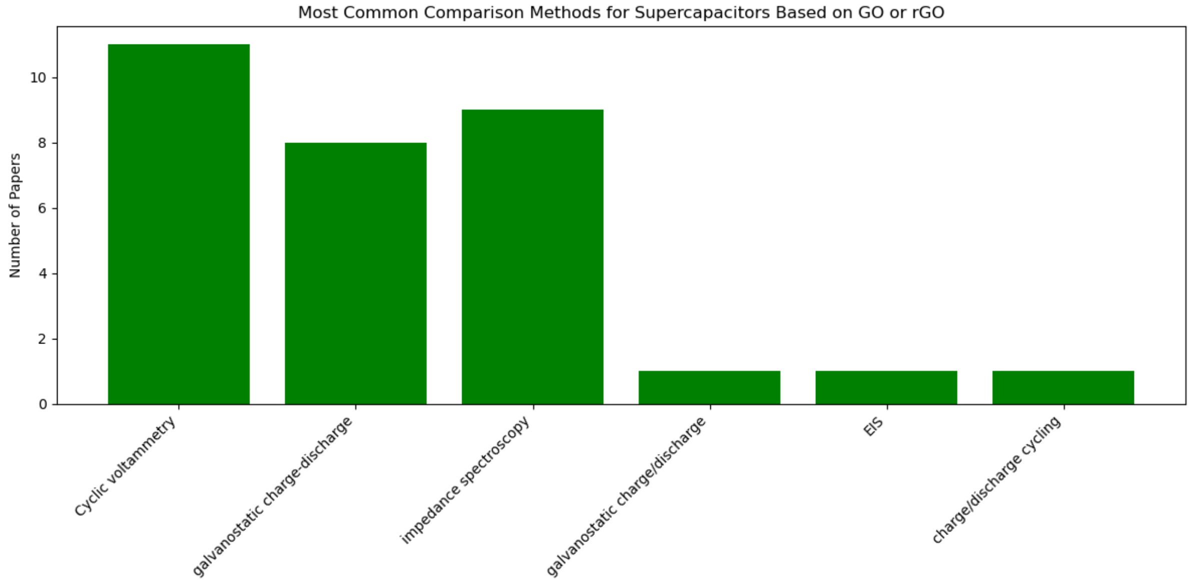

Figure 7. The most common comparison methods for EDLCs based on GO or rGO.

Galvanostatic charge–discharge testing, another widely used method, provides essential insights into the charge storage and release processes by applying a constant current to the supercapacitor and measuring the voltage response. The ability to repeatedly charge and discharge the supercapacitor under controlled conditions allows for the assessment of long-term performance and the identification of potential degradation mechanisms. Impedance spectroscopy, a technique that analyzes the frequency response of a system, is frequently used to understand the resistive and capacitive behavior of the electrode materials. By examining the impedance spectra, it is possible to gain insights into the charge transfer resistance, ion diffusion processes, and capacitive behavior of the electrode–electrolyte interface, which are critical for optimizing the design and performance of supercapacitors.

The less commonly used methods, such as electrochemical impedance spectroscopy (EIS) and charge/discharge cycling, are also highlighted in the studies. EIS is used in two of the studies, providing additional layers of understanding regarding the dynamic processes occurring within the supercapacitors. Charge/discharge cycling tests, featured in two studies, further validate the long-term stability and reliability of the materials under practical operating conditions. EIS, in particular, allows for the separation of different contributions to the overall impedance, enabling a more detailed analysis of the electrochemical processes.

4.4. Performance Metrics: Capacitance

In Figure 8, Tables 3 and S3, the analysis of specific capacitance values for GO- and rGO-based EDLCs reveals significant insights into the performance enhancements through various modifications and composites. The study by Gutierrez et al. [55] demonstrates the high specific capacitance values of 0.356 F/g and 117 F/g achieved with a tri-layered electrode incorporating rGO, MnO_2 nanowires, and Co_3O_4 nanorods. This result underscores the effectiveness of combining rGO with metal oxides to enhance capacitive performance. Similarly, Lee et al. [56] reported specific capacitance values ranging from 24.6 to 489 F/g for various PANi and GO hybrid electrodes. The significant enhancement in capacitance values, particularly for PG10-H (489 F/g), emphasizes the synergistic effects of PANi and GO, which combine pseudocapacitive behavior with a high surface area.

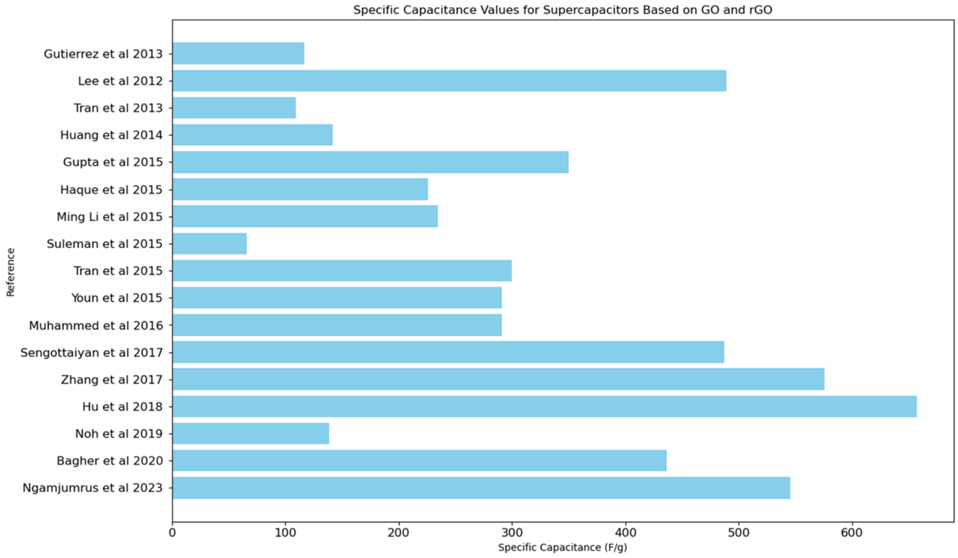

Figure 8. Specific capacitance values for EDLCs based on GO and rGO.

Tran et al. [57] achieved specific capacitance values of 88 F/g and 109 F/g for new composites and PSSGO5, respectively, by integrating poly sodium 4-styrenesulfonate with GO. This study highlights the potential of polymer intercalation to improve the electrochemical performance of GO-based electrodes. In a related study, Huang et al. [58] reported a specific capacitance of 141.8 F/g using a GO-B-PVA/KOH gel electrolyte, demonstrating the benefits of gel electrolytes over traditional aqueous solutions in enhancing capacitance. Gupta et al. [60] provided a comprehensive analysis of hybrid films involving GO and ErGO with polyaniline (PAni) and polypyrrole (PPy), showing specific capacitance values ranging from 70 to 350 F/g. The layer-by-layer assembly of these hybrid films offers a clear path for achieving high-performance supercapacitors through strategic material combinations. Haque et al. [61] further supported this by demonstrating significant capacitance improvements with nitrogen-doped graphene, achieving 226 F/g from CV measurements, which outperformed undoped graphene and GO. Ming Li et al. [62] showed specific capacitance values of 234.3 F/g in 0.5 M H_2SO_4 and 187.8 F/g in 1 M TEABF4/PC for nitrogen-doped reduced graphene oxide (N-rGO), highlighting the role of nitrogen doping in enhancing the capacitive performance. Suleman et al. [63] reported specific capacitance values of 66 F/g for GO and 60 F/g for r-GO when interfaced with a plastic-crystal-based flexible gel polymer electrolyte, emphasizing the superior performance of GO in flexible energy storage applications.

The study by Tran et al. [64] demonstrated that functionalizing rGO with glucose and nickel oxide could increase specific capacitance up to five times higher than that of reduced GO alone, reaching about 300 F/g. This significant enhancement points to the importance of functionalization in improving electrochemical properties. Youn et al. [65] achieved a specific capacitance of 291 F/g at a current density of 1 A/g with reduced graphene oxide, illustrating the high capacitance and rate capability of rGO. Muhammed et al. [66] reported specific capacitance values of 291 F/g at 1 A/g and 261 F/g at 50 A/g for nitrogen-doped reduced graphene oxide synthesized via solid-state microwave irradiation, showcasing an effective method for producing high-performance rGO. Sengottaiyan et al. [69] and Zhang et al. [70] further pushed the boundaries, reporting specific capacitance values of 487 F/g and 576 F/g, respectively, with Co_3O_4 nanocrystals embedded in rGO and a ternary composite of GO, carbon dots, and polypyrrole. These studies stress the potential of advanced composites to achieve superior capacitive performance.

Hu et al. [71] achieved a remarkable specific capacitance of 657 F/g with a $V_3O_7{\cdot}H_2O$ nanobelt/CNT/reduced graphene oxide composite, highlighting the significant gains possible through innovative material combinations. Noh et al. [72] demonstrated the benefits of rGO–SnO_2 nanorod bundles, achieving a specific capacitance of 138.4 F/g, which outperformed other configurations like rGO–SnO_2 nanoparticles and bare rGO. Bagher et al. [74] highlighted the advantages of ionic liquid-functionalized graphene, reporting specific capacitance values of 436.7 F/g for IFG and 262.5 F/g for rGO, indicating the effectiveness of functionalization in enhancing performance. Finally, Ngamjumrus et al. [80] reported high specific capacitance values of 545.78 F/g for rGO/AC–HDC–3D15M samples, emphasizing the role of innovative composite structures in achieving superior energy storage capabilities.

These studies provide compelling evidence of the advancements in specific capacitance values for GO and rGO-based EDLCs through various strategies, including hybridization, functionalization, and innovative composite designs. These findings collectively contribute to a deeper understanding of the material properties and offer pathways for developing high-performance energy storage devices.

4.5. Performance Metrics: Energy Density

The energy density of supercapacitors is a critical parameter as it determines the amount of energy that can be stored per unit mass, which is essential for applications requiring high-energy storage. Table 3 and Figure 9 collectively present the energy density values reported across multiple studies, highlighting the significant variability in performance due to different material compositions and synthesis methods. The study by Lee et al. [56] reported energy densities of 18.92 Wh/kg and 30.34 Wh/kg for hybrid electrodes of PANi and GO. This enhancement can be attributed to the combination of the pseudocapacitive behavior of PANi with the high surface area of GO, which facilitates better charge storage. Likewise, Ming Li et al. [62] achieved an energy density of 25.1 Wh/kg for N-rGO, emphasizing the role of nitrogen doping in enhancing the electrochemical performance by introducing additional active sites for charge storage.

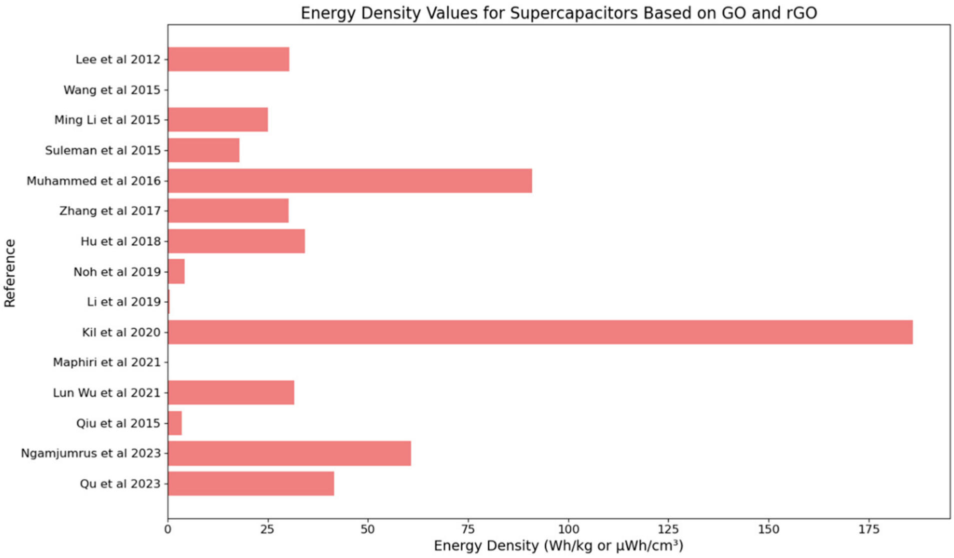

Figure 9. Energy density values for EDLCS based on GO and rGO.

Suleman et al. [63] reported energy densities of 18 Wh/kg for GO and 15.6 Wh/kg for r-GO when interfaced with a plastic-crystal-based flexible gel polymer electrolyte. This study features the superior performance of GO over r-GO in flexible energy storage applications, which is crucial for developing flexible and wearable electronic devices. Muhammed et al. [66] achieved a remarkable energy density of 91.0 Wh/kg with nitrogen-doped reduced graphene oxide synthesized via solid-state microwave irradiation. The high energy density is likely due to the increased electrical conductivity and surface area resulting from the nitrogen doping and the synthesis method used.

The studies by Zhang et al. [70] and Hu et al. [71] reported energy densities of 30.1 Wh/kg and 34.3 Wh/kg, respectively, for their advanced composites involving GO. In Ref. [70], the ternary composite of GO, carbon dots, and polypyrrole achieved this high energy density due to the synergistic effects of the materials, while in Ref. [71], $V_3O_7 \cdot H_2O$ nanobelts/CNT/reduced graphene oxide composite demonstrated superior charge storage capability, indicating the potential of innovative material combinations. In contrast, Noh et al. [72] reported a lower energy density of 4.17 Wh/kg for rGO–SnO_2 nanorod bundles, which, while lower compared to other configurations, still indicates the potential for further optimization of metal oxide and graphene composites. Li et al. [73] presented an energy density of 0.48 Wh/kg, which is relatively low, indicating that the specific synthesis or material combination used might require further optimization to enhance performance.

Kil et al. [75] reported exceptionally high energy densities of 186 Wh/kg at 142 W/kg and 100 Wh/kg at 10 kW/kg, demonstrating the significant potential of their material design for high-performance energy storage devices. This result sets a benchmark for future research in developing high-energy-density supercapacitors. Maphiri et al. [76] achieved a volumetric energy density of 14.61 mWh/cm^3 for TRGO-500, emphasizing the importance of considering volumetric performance, particularly for applications where space is a limiting factor. Lun Wu et al. [77] reported an energy density of 31.68 Wh/kg, further supporting the potential of innovative composite structures to enhance energy storage capabilities.

Qiu et al. [79] and Ngamjumrus et al. [80] also provided valuable insights, with the first case reporting an energy density of 3.52 Wh/kg and the second case achieving an energy density of 60.83 Wh/kg. These findings illustrate the significant advancements in material design and synthesis methods that can lead to substantial improvements in energy storage performance.

The energy density findings highlight the diverse approaches and material modifications used to enhance the performance of GO and rGO-based supercapacitors. The significant variability in reported energy densities underscores the potential for further optimization and innovation in material design and synthesis methods to achieve even higher performance in future energy storage devices. Indeed, this comprehensive analysis provides a clear direction for future research focused on maximizing the energy density of supercapacitors through advanced material engineering.

4.6. Performance Metrics: Power Density

The power density is a critical parameter that determines the rapid energy delivery capabilities of these devices. As shown in Table 3 and Figure 10, there is significant variability in the reported power densities across different studies, reflecting the impact of diverse material modifications and synthesis techniques. Lee et al. [56] reported a power density of 1.0 kW/kg for hybrid electrodes composed of polyaniline and GO. This finding underscores the potential of combining GO with conductive polymers to enhance the power delivery of EDLCs. The synergistic effect of polyaniline and GO results in improved electrical conductivity and efficient charge transport, making this combination suitable for high-power applications. Gao et al. [59] achieved a power density of 1540 mW/cm^2, translating to substantial power output on a per-area basis. This result is particularly significant for applications where space is limited, such as in compact electronic devices. In addition, the high-power density achieved demonstrates the effectiveness of optimizing electrode surface area and electrolyte interactions. Ming Li et al. [62] reported a power density of 10 kW/kg for nitrogen-doped reduced graphene oxide. The introduction of nitrogen atoms into the graphene lattice enhances the electrical conductivity and electrochemical activity, facilitating rapid charge–discharge cycles. This high power density highlights the importance of doping strategies in developing advanced electrode materials for EDLCs.

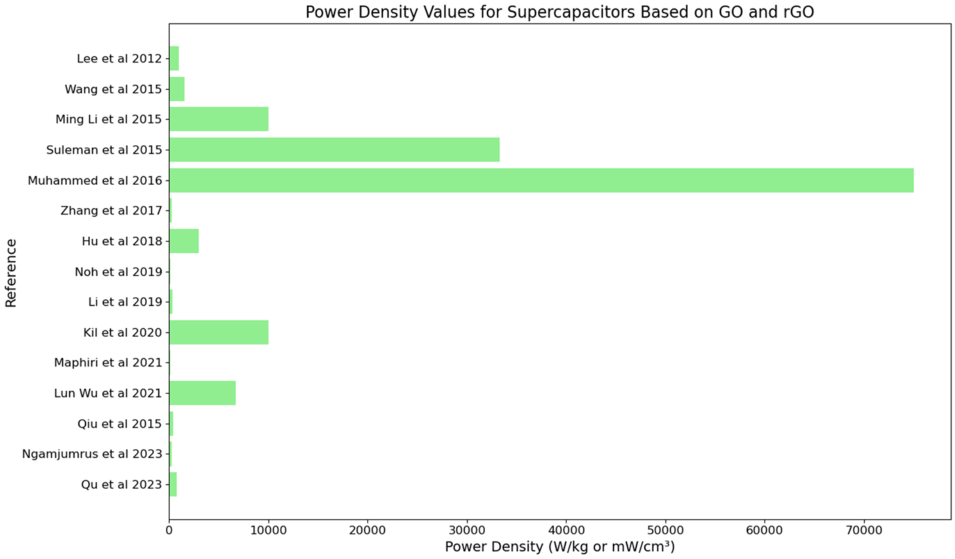

Figure 10. Power density values for EDLCS based on GO and rGO.

Suleman et al. [63] showed the superior performance of rGO with a power density of 54.9 kW/kg, compared to 33.3 kW/kg for GO. The reduced form of GO exhibits enhanced electrical conductivity and lower internal resistance, enabling faster energy transfer and higher power output. This study emphasizes the advantages of reduction processes in improving the performance of GO-based supercapacitors. Muhammed et al. [66] achieved an extraordinary power density of 75.0 kW/kg with nitrogen-doped reduced graphene oxide synthesized via solid-state microwave irradiation. This method yields a highly conductive and porous material, resulting in ultra-high power densities. Such high-performance electrodes are crucial for applications requiring rapid energy delivery, such as in power tools and electric vehicles. Zhang et al. [70] and Hu et al. [71] reported power densities of 250 W/kg and 3000 W/kg, respectively. These studies highlight the potential of innovative material combinations in achieving high-performance supercapacitors. Noh et al. [72] and Li et al. [73] reported lower power densities of 123 W/kg and 371 W/kg for RGO–SnO_2 nanorod bundles and other configurations. While these values are modest, they suggest room for optimization in these composites to enhance power performance. Kil et al. [75] achieved a power density of 10 kW/kg, aligning with their high energy density results and indicating a balanced approach to achieving both high energy and power densities.

Maphiri et al. [76] reported a volumetric power density of 142.67 mW/cm^3, underscoring the importance of considering volumetric performance for applications with space constraints. Lun Wu et al. [77] achieved a high power density of 6750 W/kg, evidencing the potential of innovative composite structures in enhancing power delivery capabilities. Qiu et al. [79] and Ngamjumrus et al. [80] provided valuable insights with power densities of 0.44 kW/kg and 260.834 W/kg, respectively. These findings illustrate the advancements in material design and synthesis methods, leading to significant improvements in power density. Qu et al. [81] reported a power density of approximately 801.5 W/kg, demonstrating the effectiveness of their material synthesis in achieving high power delivery. As noted, the variability in reported power densities emphasizes the potential for further optimization in material design and synthesis methods. The advancements in power density demonstrated by these studies are promising for the development of high-performance energy storage devices capable of rapid energy delivery, which are essential for a wide range of applications.

4.7. Performance Metrics: Efficiency

Efficiency is essential for their long-term stability and practical application. Efficiency, often measured in terms of capacitance retention after multiple charge–discharge cycles, provides insights into the durability and reliability of these materials under repeated usage. Table 3 and Figure 11 illustrate the capacitance retention percentages and the number of cycles for various studies, highlighting the performance and stability of GO and rGO-based supercapacitors.

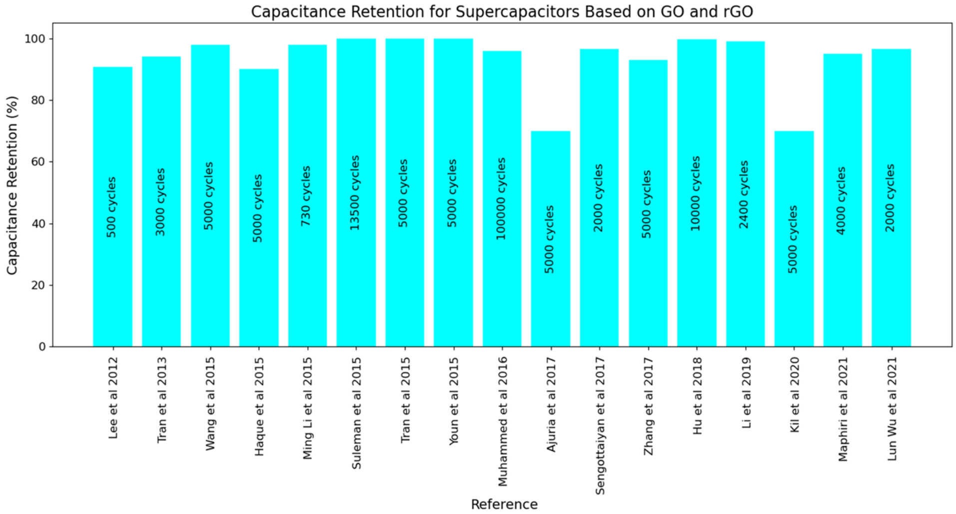

Figure 11. Capacitance retention for EDLCs based on GO and rGO.

Various studies report high capacitance retention, indicating excellent stability and efficiency. For example, Gao et al. [59] achieved 98% retention after 5000 cycles, demonstrating the effectiveness of their material design. Similarly, Hu et al. [71] reported an impressive 99.7% retention after 10,000 cycles, highlighting the potential of their composite material for long-term applications. Muhammed et al. [66] achieved 96% retention after 100,000 cycles, confirming the extraordinary durability of nitrogen-doped reduced graphene oxide synthesized via solid-state microwave irradiation. This result sets a high benchmark for future research aiming to develop ultra-durable supercapacitors.

Other studies also report high efficiency, such as Suleman et al. [63], who achieved stable performance up to 13,500 cycles, and Li et al. [73], who reported 99% retention after 2400 cycles. These findings stress the potential of various modifications and composite designs to enhance the longevity and reliability of GO and rGO-based supercapacitors. In contrast, some studies report lower capacitance retention, suggesting room for improvement. For example, Wang et al. [67] reported 70% retention after 5000 cycles, and Kil et al. [75] achieved the same retention after 5000 cycles. These results indicate that while the materials show promise, further optimization is needed to enhance their efficiency and stability.

The variability in reported efficiencies underscores the importance of material design and synthesis methods in determining the performance of GO and rGO-based supercapacitors. High-efficiency materials typically involve advanced composites, doping strategies, or innovative synthesis techniques that enhance the structural integrity of the material and related electrochemical properties.

5. Limitations and Recommendations

5.1. Limitations of the Current Systematic Review

Despite the comprehensive approach employed in this systematic review, some limitations need to be accepted, which are ascribed as follows:

Population (P):

- The review focuses solely on studies involving EDLCs incorporating GO and rGO. This narrow focus could exclude other types of capacitors and energy storage devices that might benefit from similar materials.

Intervention (I):

- There is considerable variability in the synthesis methods and conditions for producing GO and rGO across different studies. This heterogeneity makes it challenging to directly compare results and draw generalized conclusions about the effectiveness of specific interventions.
- The review includes various interventions (e.g., doping, composite formation, etc.) combined with GO and rGO, leading to diverse performance outcomes. The interactions between different interventions and their combined effects on EDLC performance are complex and not fully understood.

Comparison (C):

- Many studies included in the review lack standardized comparison or control materials, making it difficult to isolate the specific contributions of GO and rGO to performance improvements.
- The performance metrics of EDLCs vary widely depending on the baseline materials and conditions used in the comparative studies, further complicating the comparison of results.

Outcomes (O):

- There are inconsistencies in how performance metrics such as capacitance, energy density, and cycling stability are measured and reported across studies. This lack of uniformity hampers the ability to perform meta-analyses and aggregate data effectively.
- Many studies focus on short-term performance metrics, while long-term stability and degradation behavior, which are critical for practical applications, are less frequently reported.

Study Design (S):

- There is a potential for publication bias, as studies with positive results are more likely to be published than those with negative or inconclusive findings. This bias can skew the overall understanding of the effectiveness of GO and rGO in EDLCs.

While this systematic review provides valuable insights into the role of GO and rGO in EDLCs, these limitations highlight the need for more standardized, high-quality research to validate and expand upon the findings. Future reviews should aim to address these limitations by incorporating more specific inclusion criteria, standardized measurement protocols, and comprehensive reporting practices.

5.2. Recommendations for Future Research

Based on the findings and limitations of this systematic review, several recommendations for future research can be proposed:

Standardization of Synthesis Methods:

- Standardized protocols for the synthesis of GO and rGO should be developed and adopted to ensure consistency in material properties across different studies. This standardization will facilitate more reliable comparisons and meta-analyses of research findings.
- Scalable synthesis methods that can be implemented at an industrial level should be a focus of study. Research should aim to optimize these methods to maintain the high performance of GO and rGO while ensuring economic feasibility and environmental sustainability.

Comprehensive Performance Metrics:

- Future studies should include long-term performance metrics, such as cycling stability over extended periods and under various operating conditions. Understanding the degradation mechanisms of GO and rGO-based EDLCs is crucial for their practical application.
- Standardized testing conditions and protocols for measuring key performance metrics, such as capacitance, energy density, power density, and efficiency, should be established. Consistent reporting of these metrics will enable more accurate comparisons between studies.

Environmental and Sustainable Approaches:

- Investment should be made in developing environmentally friendly synthesis processes for GO and rGO that minimize the use of toxic chemicals and reduce the overall environmental impact. Utilizing renewable resources and waste materials for the synthesis of graphene derivatives can also contribute to sustainability.
- Lifecycle analyses of GO and rGO-based EDLCs should be conducted to evaluate their environmental impact from production to disposal. This analysis would help identify areas for improvement in sustainability.

Material Optimization:

- The effects of various doping elements and functional groups on the electrochemical performance of GO and rGO should be further explored. Tailoring the oxygen content and introducing heteroatoms or other functional groups can optimize the properties of these materials for specific applications.
- The potential of hybrid materials that combine GO and rGO with other high-performance materials, such as metal oxides, conductive polymers, and carbon nanotubes, should be investigated. These hybrid materials can synergistically enhance the performance of EDLCs.

Interfacial Engineering:

- Research should focus on innovative electrode designs that enhance the interfacial contact between GO/rGO and the electrolyte. Improved electrode architecture can lead to better ion transport, increased surface area, and enhanced overall performance.
- The compatibility of GO and rGO with various electrolytes, including aqueous, organic, and ionic liquid electrolytes, should be explored. Understanding the interactions between the electrode material and the electrolyte will help optimize the performance of EDLCs.

Application-Specific Research:

- Application-specific research should be conducted to tailor GO and rGO-based EDLCs for targeted applications, such as portable electronics, electric vehicles, and grid energy storage. Each application may have unique requirements for performance metrics and material properties.
- Researchers should move beyond laboratory-scale experiments to develop and test prototypes of GO and rGO-based EDLCs in real-world scenarios. Prototyping will provide practical insights into the challenges and potential of these materials for commercial applications.

6. Potential Challenges and Solutions for Implementing GO and rGO in Realistic Applications

6.1. Challenges and Proposed Solutions

Scalability and Production Costs

- Challenge: The synthesis of high-quality GO and rGO on a large scale remains a significant challenge. The methods commonly used in laboratories, such as Hummers' method for GO and chemical reduction for rGO, are often not cost-effective or scalable.
- Solution: The development of scalable production techniques, such as continuous flow processes and green synthesis methods, can help in reducing costs and improving yield. Collaborations between academia and industry could foster innovations in manufacturing technologies, making large-scale production more feasible.

Material Consistency and Quality Control

- Challenge: Ensuring consistency in the quality of GO and rGO is critical for their performance in applications. Variations in the degree of oxidation, reduction, and functionalization can lead to significant differences in material properties.
- Solution: Implementing standardized protocols for the synthesis and characterization of GO and rGO can help in maintaining consistency. Advanced characterization techniques and real-time monitoring systems can be employed to ensure high-quality production.

Integration with Existing Technologies

- Challenge: Integrating GO and rGO into existing technologies and systems can be challenging due to compatibility issues with other materials and components.
- Solution: Research on hybrid materials and composites that combine GO and rGO with conventional materials can enhance compatibility. Developing adhesive interfaces and surface treatments can also facilitate better integration.

Performance Stability and Durability

- Challenge: Maintaining the stability and durability of GO and rGO-based devices under operational conditions is crucial. Issues such as electrode degradation and loss of capacitance over time need to be addressed.
- Solution: Enhancing the structural integrity of GO and rGO through doping, cross-linking, and encapsulation techniques can improve their stability. Conducting long-term performance studies and accelerated aging tests can provide insights into improving durability.

6.2. Environmental Sustainability of GO and rGO

Chemical Synthesis and Waste Management

- Impact: The chemical processes involved in the synthesis of GO and rGO often generate hazardous waste, including strong acids, oxidizing agents, and reducing agents. Improper disposal of these chemicals can lead to environmental pollution and health risks.
- Solution: Developing greener synthesis methods that minimize the use of hazardous chemicals and generate less waste is essential. Techniques such as electrochemical

exfoliation and biomass-derived precursors offer more environmentally friendly alternatives. Implementing strict waste management protocols and recycling processes can also mitigate environmental impact.

Toxicity and Biocompatibility

- Impact: The potential toxicity of GO and rGO to humans and the environment is a concern, particularly for applications involving direct human contact or disposal in natural ecosystems.
- Solution: Conducting comprehensive toxicity and biocompatibility studies is necessary to understand the potential risks. Functionalizing GO and rGO with biocompatible molecules and coatings can reduce toxicity. Regulations and guidelines for safe handling, usage, and disposal should be established and followed.

Energy Consumption and Carbon Footprint

- Impact: The energy-intensive nature of GO and rGO production can contribute to a high carbon footprint, which is counterproductive to the goal of sustainable energy storage solutions.
- Solution: Optimizing production processes to reduce energy consumption and adopting renewable energy sources for manufacturing can lower the carbon footprint. Life cycle assessments (LCA) can help identify areas for improvement and track the environmental impact of GO and rGO production.

7. Conclusions

This systematic review comprehensively explored the advancements and applications of GO and rGO in EDLCs, focusing on their impact on capacitance, energy density, power density, and efficiency. Through an in-depth analysis of multiple studies, this review highlights the significant variability in performance due to diverse material modifications and synthesis techniques.

- In terms of capacitance, the studies reviewed demonstrate that both GO and rGO exhibit high specific capacitance values, which are further enhanced through hybridization with conductive polymers and metal oxides. The introduction of nitrogen doping and functionalization with other materials significantly boosts electrochemical performance, making these materials promising for high-capacity energy storage devices.
- In terms of energy density, GO and rGO-based EDLCs show considerable potential in achieving high energy densities, especially when combined with innovative composite structures. The introduction of nitrogen atoms and the development of advanced hybrid materials result in substantial improvements in energy storage capabilities, making these materials suitable for a wide range of applications requiring high energy density.
- In terms of power density, the review highlights the remarkable power densities achieved with GO and rGO-based supercapacitors, particularly those utilizing nitrogen-doped reduced graphene oxide and advanced composites. These materials exhibit excellent power delivery capabilities, essential for applications requiring rapid energy discharge.
- The efficiency of GO and rGO-based EDLCs, measured in terms of capacitance retention after multiple charge–discharge cycles, varies significantly across different studies. High-efficiency materials typically involve advanced composites, doping strategies, and innovative synthesis techniques that enhance structural integrity and electrochemical properties. The durability and long-term stability of these materials are promising for practical applications.

Finally, GO and rGO hold significant promise for the development of high-performance EDLCs. These findings provide a clear direction for future research and development efforts aimed at maximizing the potential of graphene-based materials in energy storage applications.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/batteries10070256/s1, Table S1. Interventions considering the preparation method, electron, and electrolyte. Table S2. Interventions considering the material and method for comparison and cyclin stability. Table S3. Performance metrics.

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