

Graphene Oxide and Halide Perovskite Nanocomposites with Gel Polymer Electrolytes for Electrochemical Applications: A Review

Abstract

Graphene oxide (GO) and halide perovskites have separately emerged as two of the most versatile materials in modern electrochemical energy science, and their integration with gel polymer electrolytes (GPEs) is increasingly recognized as a route toward efficient, flexible, and durable devices. This review synthesizes recent literature on the structural chemistry, synthesis, and electrochemical roles of GO and halide perovskites, with particular attention to their combination with GPEs in photovoltaic, supercapacitor, and battery systems. Graphene oxide is discussed as a crystallization additive and interfacial layer in perovskite solar cells, and as a conductivity-enhancing filler that suppresses polymer crystallinity in gel and solid polymer electrolytes for lithium batteries and dye-sensitized solar cells. Halide perovskites are examined both as light-harvesting and pseudocapacitive electrode materials, with emphasis on the electrolyte-compatibility challenges created by their intrinsic moisture and ion-migration instability. The review then considers how gel polymer electrolytes, and GO-modified variants in particular, can mitigate leakage, improve ionic conductivity, and extend device lifetimes. Key performance metrics reported in the literature, including power conversion efficiency, ionic conductivity, and capacitance retention, are compared, and outstanding challenges relating to long-term stability, scalability, and toxicity are outlined. The review concludes that rationally engineered GO-perovskite-GPE architectures represent a promising but still-maturing direction for next-generation flexible electrochemical devices.

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Introduction

The last two decades have seen parallel breakthroughs in two classes of functional materials for energy conversion and storage: graphene-based carbon nanomaterials and metal halide perovskites. Graphene oxide, an oxygenated derivative of graphene bearing hydroxyl, epoxide, and carboxyl functional groups, combines solution processability with a high surface area and tunable electronic structure, making it attractive as an electrode additive, interfacial layer, and electrolyte filler. Organic-inorganic and all-inorganic halide perovskites, typified by methylammonium lead iodide and cesium lead halides, have achieved remarkable power conversion efficiencies in photovoltaics and have more recently been explored as redox-active electrodes in supercapacitors and batteries owing to their tunable bandgaps and mixed ionic-electronic conductivity.

A persistent obstacle for both material classes is device-level stability. Perovskite solar cells (PSCs) suffer from ion migration, moisture sensitivity, and thermal degradation, while conventional liquid electrolytes used in dye-sensitized and related devices are prone to leakage and solvent evaporation. Gel polymer electrolytes, which combine the mechanical robustness of a solid polymer matrix with the ionic transport properties of a liquid electrolyte immobilized within it, have been proposed as a quasi-solid-state solution to these problems. Incorporating GO into such gels, or coupling halide perovskite electrodes with GPEs, has been reported to improve ionic conductivity, suppress leakage, and extend operational lifetimes across several device types. This review brings together these strands of research, organizing the discussion around (i) the fundamental properties of GO and halide perovskites, (ii) the design principles of gel polymer electrolytes, and (iii) the reported performance of composite systems in photovoltaic and energy-storage applications.

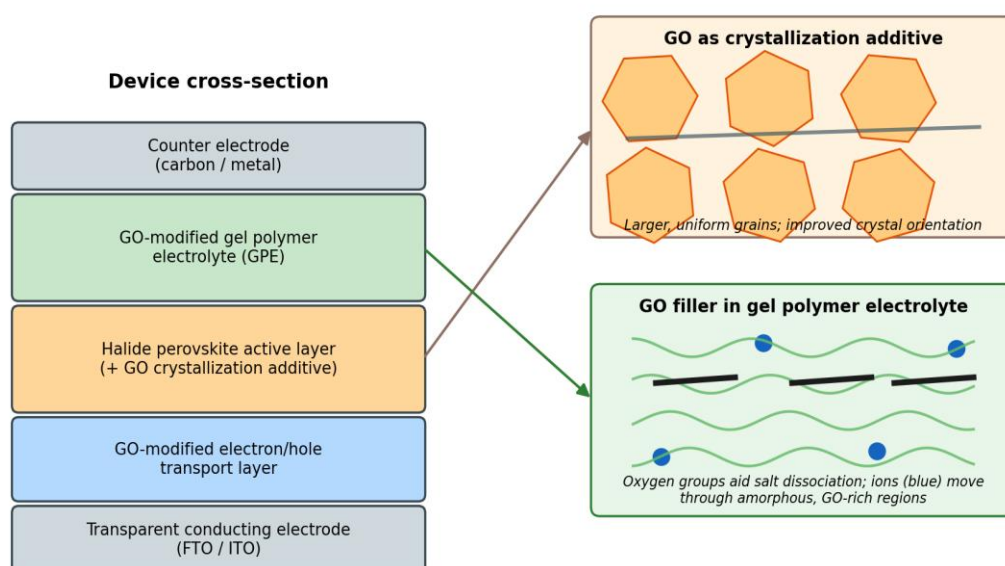


Figure 1. Schematic device cross-section showing the two principal points of GO integration discussed in this review: as a crystallization additive within the halide perovskite active layer, and as a conductivity-enhancing filler dispersed within the gel polymer electrolyte.

Graphene Oxide: Structure, Properties, and Electrochemical Roles

Graphene oxide is typically produced by oxidative exfoliation of graphite, most commonly via the Hummers method, in which graphite is treated with concentrated sulfuric acid, sodium nitrate, and potassium permanganate to yield a highly oxidized graphite product that exfoliates into single- or few-layer sheets (Hummers & Offeman, 1958). These sheets are decorated with oxygen-containing functional groups that disrupt the sp^2 lattice, rendering GO electrically insulating relative to pristine graphene, but they also provide reactive sites for functionalization and strong interactions with polar polymer chains and ionic species. In electrolyte systems, these interactions are exploited directly: GO can assist dissociation of lithium salts and increase the amorphous fraction of semicrystalline host polymers such as poly(ethylene oxide) (PEO).

Several studies illustrate this effect quantitatively. In a PEO-based solid polymer electrolyte for lithium-metal batteries, the incorporation of only 1 wt% GO increased ionic conductivity roughly sevenfold relative to the GO-free electrolyte while lowering the activation energy for ion transport, an effect attributed to GO's suppression of PEO crystalline-nuclei formation and the resulting increase in amorphous, ion-conducting regions; the same GO-modified electrolyte widened the electrochemical stability window to about 5 V and supported stable lithium symmetric-cell cycling for 600 hours (Wen et al., 2021). In aqueous acid-doped gel electrolytes for flexible supercapacitors, GO addition simultaneously improved ionic conductivity, tensile strength, and cycling stability, with an optimum GO loading beyond which further addition became counter-productive; the best-performing GO/poly(vinyl alcohol)/phosphoric-acid gel retained about 89% of its initial capacitance after 2000 cycles (Alipoori et al., 2019).

Beyond bulk electrolytes, GO and its functionalized derivatives have also been dispersed directly into gel polymer electrolytes for dye-sensitized solar cells (DSSCs). Because unmodified graphene nanoplatelets tend to aggregate in polymer gels, chemical functionalization with polyethylene glycol has been used to improve dispersion within a PVDF-HFP/PEO blended gel, yielding a maximum ionic conductivity of about 4.11×10^{-3} S/cm, among the higher values reported for graphene-modified quasi-solid electrolytes in DSSCs (Manafi et al., 2020). Collectively, these findings establish GO not merely as a passive filler but as an active structural and chemical modifier of gel polymer electrolytes.

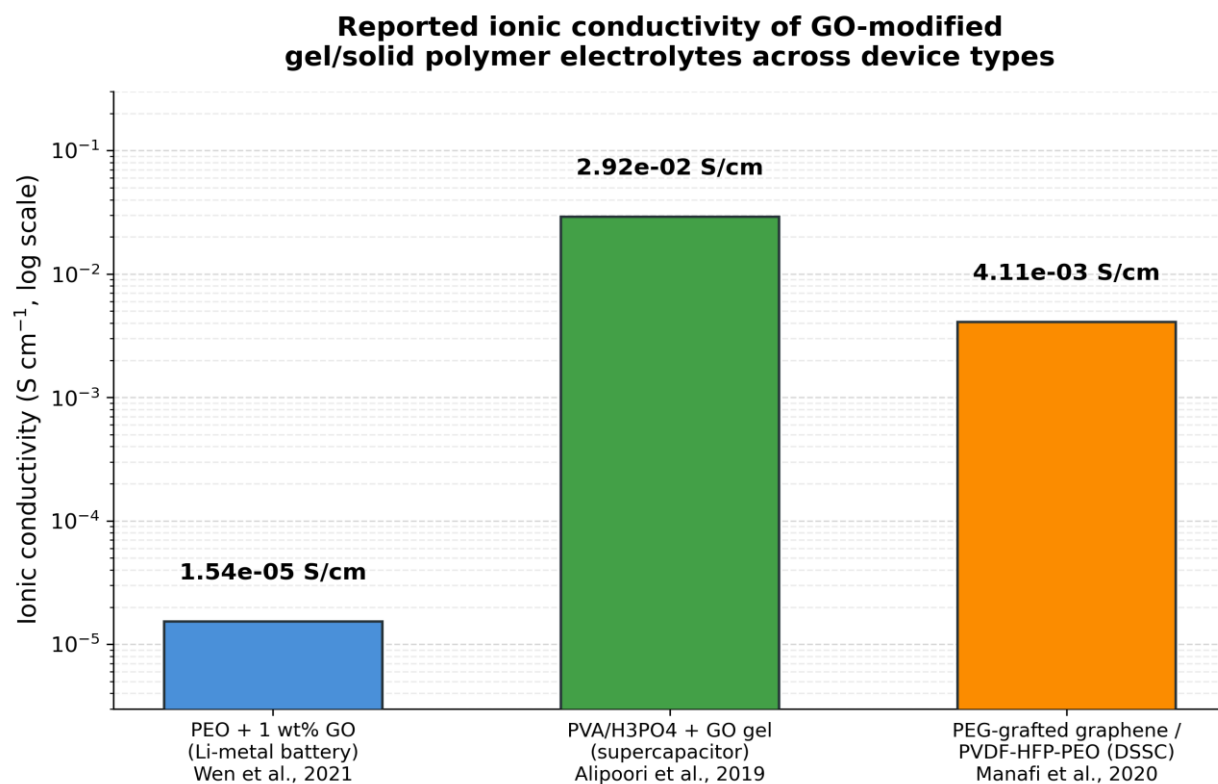


Figure 2. Reported room-temperature ionic conductivity of GO-modified gel and solid polymer electrolytes across three device contexts (note the logarithmic y-axis). Values vary by more than three orders of magnitude depending on the host polymer, GO loading/functionalization, and electrolyte chemistry.

Halide Perovskites: Structure, Properties, and Electrochemical Behavior

Halide perovskites adopt the general ABX_3 structure, where A is an organic or inorganic monovalent cation (methylammonium, formamidinium, or cesium), B is typically Pb^{2+} or Sn^{2+} , and X is a halide (I-, Br-, or Cl-). Their appeal in optoelectronics stems from tunable direct bandgaps, high absorption coefficients, long carrier diffusion lengths, and defect tolerance, which together have driven a rapid rise in perovskite solar cell power conversion efficiency over the past decade and a half, from the original 3.8% efficiency demonstrated for a methylammonium lead halide-sensitized photoelectrochemical cell (Kojima et al., 2009) to well over 25% in current record devices.

More recently, halide perovskites have been investigated directly as electrode materials for supercapacitors and batteries, exploiting redox-active B-site cations and mixed ionic-electronic conduction to store charge pseudocapacitively. A recent review of halide perovskite supercapacitors summarizes synthesis routes, electrode architectures, and electrolyte-selection strategies for this emerging application, while highlighting that reproducibility and long-term cycling stability remain the key open problems (Popoola & Qahtan, 2025). A central complication is that most halide perovskites are chemically incompatible with the aqueous alkaline or acidic electrolytes conventionally used in supercapacitors, since exposure to water or strong acids/bases readily decomposes the perovskite lattice. A comprehensive review of the chemical reactions perovskite films undergo under moisture, oxygen, and light, and at interfaces with charge-transport materials and metal electrodes, similarly identifies these degradation pathways as the central obstacle to long-term device stability, whether in photovoltaic

or energy-storage contexts (Zhuang et al., 2023). This incompatibility is a major driver of interest in non-aqueous, quasi-solid-state, and gel polymer electrolytes for perovskite-based energy-storage devices, discussed further in Section 6 (Popoola et al., 2020).

Gel Polymer Electrolytes: Design Principles

A gel polymer electrolyte is formed by swelling a polymer host (e.g., PEO, PVDF-HFP, polyurethane, or PVA) with a liquid electrolyte solution, or by in-situ polymerizing a monomer around a salt-containing solvent. The resulting quasi-solid-state material retains much of the ionic conductivity of the liquid phase while gaining the leakage resistance, mechanical integrity, and processability of a solid film. Polymer choice, molecular weight, and cross-linking chemistry strongly influence gel performance, and redox-shuttle chemistry within the gel can also be engineered to raise open-circuit voltage and reduce charge recombination in dye-sensitized architectures. Graphene-family fillers, discussed in Section 2, are one of the most effective and widely studied strategies for simultaneously improving the ionic conductivity and mechanical robustness of these gels.

Integrating Graphene Oxide with Perovskites: Photovoltaic and Interfacial Applications

The most extensively documented convergence of GO and halide perovskites occurs in the perovskite active layer and its interfaces, rather than in the electrolyte itself. Adding a small volume fraction of GO to the perovskite precursor solution has been shown to template larger, more uniform grains and improve preferential crystal orientation, which in turn enhances charge extraction and optical absorption relative to GO-free films. In one representative study, an optimized 1 vol% GO addition to a methylammonium lead iodide precursor increased and homogenized grain size and improved the out-of-plane crystalline orientation of the resulting perovskite film, as evidenced by electron microscopy and grazing-incidence X-ray diffraction (Zhang et al., 2018). GO has also been used as an additive in inverted planar perovskite/graphene-oxide hybrid solar cells, where hybridization with the perovskite layer was shown to improve device performance (Chung et al., 2017).

GO and its doped derivatives have also been deployed as interfacial and hole-transport layer components. At the highest-performing end of this approach, doping the hole-injection layer of a carbon-electrode perovskite solar cell with graphene oxide was recently reported to enable a certified efficiency of 23.6%, illustrating that GO's role has expanded from a laboratory-scale crystallization additive to a component of some of the highest-performing low-cost device architectures reported to date (Wang et al., 2025). Separately, a monolithic single-layer graphene interface coupled with a polymer coating has been used to mechanically reinforce perovskite thin films, producing a twofold enhancement in modulus and hardness, restricting photoinduced lattice expansion, and allowing devices to retain more than 97% of their initial power conversion efficiency after over 3,670 hours of maximum-power-point tracking under simulated sunlight at 90 degrees Celsius (Li et al., 2025). Together, these studies show that graphene oxide's role in perovskite photovoltaics spans crystallization control, charge-transport-layer engineering, and mechanical stabilization, three functions that are conceptually distinct from, but complementary to, its role as a gel-electrolyte filler.

Halide Perovskites and Graphene Oxide in Electrochemical Energy Storage

Beyond photovoltaics, halide perovskites have been explored directly as pseudocapacitive electrodes for supercapacitors. Organometallic halide perovskite electrodes have been fabricated and paired with quasi-solid-state electrolytes specifically to avoid the rapid aqueous decomposition that halide perovskites otherwise undergo,

with one-step and two-step solution-processed perovskite electrodes compared for their resulting morphology and electrochemical performance (Popoola et al., 2020). Broader reviews of halide perovskite supercapacitors likewise emphasize that realizing the potential of these electrodes depends on solving electrode-electrolyte compatibility problems, since aqueous KOH, H₃PO₄, and related conventional supercapacitor electrolytes tend to be incompatible with halide perovskite lattices (Popoola & Qahtan, 2025).

On the electrolyte side, GO-doped gel polymer and ion-gel electrolytes have already demonstrated strong performance in solid-state supercapacitors and lithium batteries built around conventional carbon and metal-oxide electrodes (Alipoori et al., 2019; Wen et al., 2021), establishing a template that could plausibly be extended to halide perovskite electrodes once their electrolyte-compatibility constraints are better understood. Taken together, the literature suggests a clear but still largely unrealized opportunity: combining GO-modified gel polymer electrolytes, which offer improved conductivity, mechanical robustness, and leakage resistance, with halide perovskite electrodes, which offer high theoretical capacitance but poor aqueous stability, to produce composite electrochemical energy-storage devices that draw on the complementary strengths of both material families. Table 1 summarizes the representative material systems, applications, and key reported metrics discussed across Sections 2, 3, 5, and 6.

Table 1. Summary of representative material systems, applications, and key reported electrochemical or photovoltaic metrics discussed in this review.

Material system	Application	Key electrochemical metric	Notable finding	Reference
GO (1 wt%) / PEO solid polymer electrolyte	Li-metal battery	$\sigma = 1.54 \times 10^{-5}$ S/cm at 24 °C (7× increase)	GO suppresses PEO crystallinity; widens electrochemical window to ~5 V	Wen et al. (2021)
GO / PVA / H ₃ PO ₄ gel electrolyte	Flexible supercapacitor	σ up to 2.92×10^{-2} S/cm	89% capacitance retention after 2000 cycles	Alipoori et al. (2019)
PEG-functionalized graphene / PVDF-HFP-PEO gel	Dye-sensitized solar cell	$\sigma = 4.11 \times 10^{-3}$ S/cm	Functionalization prevents graphene aggregation in the gel	Manafi et al. (2020)
GO (1 vol%) additive in MAPbI ₃ precursor	Perovskite solar cell (active layer)	Improved grain size/uniformity; enhanced PL and absorption	Out-of-plane crystal orientation confirmed by GIXRD	Zhang et al. (2018)
Perovskite / graphene oxide hybrid composite	Inverted planar perovskite solar cell	Improved device efficiency vs. non-hybrid control	GO hybridization used at the perovskite interface	Chung et al. (2017)
GO-doped hole-injection layer	Carbon-electrode perovskite solar cell	Certified PCE = 23.6%	GO doping enables high efficiency without noble-metal electrodes	Wang et al. (2025)
Single-layer graphene / polymer interface	Perovskite solar cell (mechanical durability)	>97% PCE retention after 3,670 h at 90 °C (MPPT)	2× increase in modulus/hardness;	Li et al. (2025)

Material system	Application	Key electrochemical metric	Notable finding	Reference
			deformation ratio cut from 0.31% to 0.08%	
Organometallic halide perovskite electrode + quasi-solid electrolyte	Electrochemical supercapacitor	Device-level performance under quasi-solid electrolyte	One- vs two-step perovskite processing compared for electrode quality	Popoola et al. (2020)

Challenges and Future Outlook

Several challenges recur across the reviewed literature. First, moisture and ion migration remain the dominant degradation pathways for halide perovskites, whether used as photoactive layers or electrode materials, and gel polymer electrolytes must be formulated to minimize residual water and halide-reactive species. Second, while GO reliably improves ionic conductivity and mechanical strength in gel and solid polymer electrolytes, its effect is generally non-monotonic, with performance peaking at a low optimal loading before aggregation or excessive interaction with the polymer host degrades properties; identifying this optimum is formulation-specific and not always transferable between systems. Third, most reported halide perovskite electrochemical energy-storage devices remain at a proof-of-concept, laboratory scale, with limited data on long-term cycling under realistic operating conditions, encapsulation strategies, and scalable, low-toxicity lead-free alternatives. Fourth, the environmental and toxicological profile of lead-based perovskites continues to motivate research into tin-based and other lead-free halide perovskites; however, tin-based perovskites in particular still exhibit lower operational stability and efficiency than their lead-based counterparts because of the oxidation susceptibility of Sn^{2+} , volatile halides, and inherent structural instability, while non-tin lead-free alternatives based on bismuth, antimony, or germanium offer better ambient stability at the cost of substantially lower efficiency (Aftab & Ahmad, 2021; Barua et al., 2026). The stability and performance of these lead-free compositions in GPE-based devices remain comparatively underexplored.

Looking forward, promising directions include the co-design of GO-modified gel polymer electrolytes specifically tailored to the chemical sensitivities of halide perovskite electrodes; the use of GO not only as a filler but as a chemically functionalized, ion-selective interfacial layer between perovskite electrodes and gel electrolytes; and the extension of mechanical-reinforcement strategies, already demonstrated for graphene-perovskite photovoltaic films, to perovskite-based supercapacitor and battery electrodes that must withstand repeated charge-discharge cycling. Systematic studies that vary GO loading, functionalization chemistry, and polymer host in tandem with perovskite composition would help establish general design rules rather than system-specific optima.

Conclusion

Graphene oxide and halide perovskites each bring distinct, complementary strengths to electrochemical materials science: GO offers tunable surface chemistry, mechanical reinforcement, and conductivity enhancement, while halide perovskites offer tunable optoelectronic and redox properties at low processing cost. Gel polymer electrolytes provide a practical bridge between these materials and durable, leakage-resistant devices, and GO-modified GPEs in particular have already demonstrated measurable gains in ionic conductivity, mechanical strength, and cycling stability across dye-sensitized solar cells, lithium batteries, and supercapacitors. Direct

integration of halide perovskite electrodes with GO-modified gel polymer electrolytes remains comparatively nascent, constrained chiefly by the moisture and chemical sensitivity of the perovskite phase, but the individual advances reviewed here suggest that such integration is a technically feasible and promising direction for future flexible, stable, high-performance electrochemical energy devices.

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