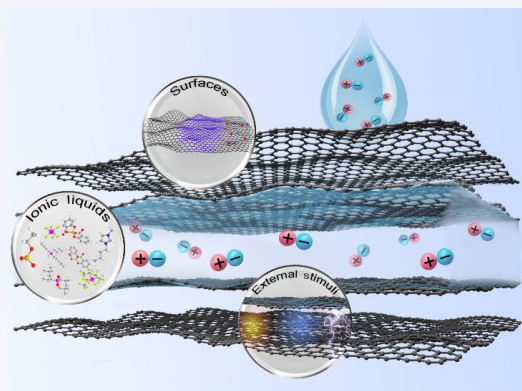


Advances in nanofriction of ionic liquids confined at graphene-based materials

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ABSTRACT: Graphene and graphene-based materials are increasingly integrated into micro- and nanoelectromechanical systems, flexible electronics, and energy devices, wherein interfacial friction poses critical challenges. Ionic liquids (ILs), owing to their negligible volatility, high stability, and structural tunability, are promising lubricants for these systems. When confined on a graphene surface, ILs form layered films that are markedly distinct from their bulk structures and exhibit unique nanofrictional behaviors. This review establishes a mechanism-oriented substrate–IL–stimuli framework to connect interfacial descriptors (the surface properties of graphene, the molecular architecture of ILs, and external stimuli, i.e., humidity, temperature, and electric field) to interfacial ion structuring and nanofrictional outputs. Insights from atomic force microscopy (AFM), surface force apparatus (SFA), and molecular dynamics (MD) simulation studies are integrated to explain how the interfacial ion structure governs nanoscale friction. Transferable structure–property relationships are extracted from recent advances from AFM, SFA, and MD simulations to identify key challenges and propose a roadmap toward programmable graphene–IL lubrication for advanced nanodevices.



KEYWORDS: nanofriction; ionic liquid (IL); graphene; interface; structure

1 Introduction

Graphene and graphene-based materials, such as graphite, are widely used in micro- and nanoelectromechanical systems (MEMS/NEMS) [1–6], flexible electronics [7, 8], and energy devices [9–11]. In these applications, interfacial friction is crucial because device performance is determined by surface processes at the micro- and nanoscales [12]. Ionic liquids (ILs), which are salts with melting points below 100 °C, are promising lubricants for such systems because of their negligible volatility, high thermal and electrochemical stability, and structural tunability [13, 14]. When confined on the surfaces of graphene-based materials, ILs form robust and ordered layers that are markedly different from the bulk, which enables stable and controllable reduction of friction at the nanoscale [15–17].

Under nanoconfinement, these lubricant films approach molecular dimensions [18, 19], and the conventional Amontons' law [20] is no longer valid. Such a breakdown is also quantitatively stark. For example, the macroscopic friction coefficient (μ_M) for bulk IL lubrication typically falls within a relatively narrow range ($\mu_M = 0.05\text{--}0.1$) [14], while the reported AFM-derived nanoscale friction coefficient (μ_N ; hereafter denoted as μ unless otherwise stated) can vary dramatically across systems and measurement protocols, spanning orders of magnitude and in some cases reaching the superlubricity regime [21, 22]. Furthermore, at the nanoscale, friction at graphene–IL interfaces is dictated by the interfacial structure, which is primarily controlled by three interdependent factors (Fig. 1), i.e., (1) the properties of graphene, such as its surface chemistry and roughness, which regulate confinement geometry and ion–substrate interactions [16]; (2) the

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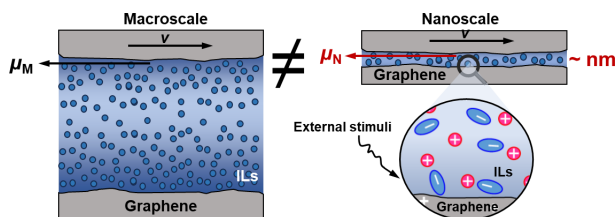


Fig. 1 Schematic of the macroscopic friction of ILs (macroscopic friction coefficient (μ_M)) and nanoscale friction of the ILs (nanoscale friction coefficient (μ_N)) confined by graphene interfaces. Nanofriction under confinement is strongly influenced by the molecular structure of the IL, the properties of the contacting graphene surface, and external stimuli (humidity, electric field, and temperature).

molecular architecture of the IL, including its ion identity and alkyl chain length, which govern interfacial packing and ordering [23]; and (3) external stimuli (e.g., humidity, electric field, and temperature), which can dynamically reorganize interfacial layers [24]. These factors modulate ion-graphene and ion-ion interactions, reshape the interfacial structure, and ultimately determine μ [25].

To understand the processes underlying nanoscale friction and improve the performance of graphene-based IL lubrication systems, researchers have used complementary experimental and computational approaches. In particular, atomic force microscopy (AFM) is used to probe interfacial layering and nanoscale friction using normal force curves and lateral force traces, respectively. The surface force apparatus (SFA) provides molecular resolution measurements of oscillatory forces originating from the sequential squeeze-out of ion pairs. Molecular dynamics (MD) simulations, including equilibrium and nonequilibrium methods, provide atomic-scale insights into structure and dynamics at confined interfaces. Equilibrium MD simulations provide insights into ion distributions, interfacial order, and electroresponsive structuring near graphene surfaces. In contrast, nonequilibrium MD (NEMD) simulations can directly calculate key dynamic properties, such as the interfacial friction coefficient (λ), and investigate flow behavior and frictional response under external force. MD simulation-integrated AFM/SFA studies reveal that graphite and graphene exhibit distinct layering and rupture forces and that charged graphene surfaces induce pronounced cation-anion alternation. These substrate-controlled interfacial structures directly correlate with layered or quantized friction behaviors, thereby explaining how interfacial ordering couples with frictional dissipation pathways and suggesting strategies to tune nanofriction by tuning surface geometry and chemistry.

Many excellent review articles have summarized the progress on the use of ILs as advanced lubricants [14, 26–28], their interfacial structures and interactions on various solids (e.g., metals and oxides) [29–33], the emerging field of electro-tunable friction enabled by ILs [34, 35], and the general behavior of ILs under nanoconfinement [36–39]. However, a unified, mechanism-oriented framework dedicated to the nanofriction of ILs at graphene-based interfaces remains scarce. Furthermore, the available literature does not adequately address the following questions:

(1) How do the surface properties of graphene influence the nanofriction of confined ILs? While previous review articles have comprehensively discussed IL structures on many solids, they have seldom discussed how the atomic flatness, tunable electronics, and chemical groups of graphene, graphene oxide (GO), or reduced GO (rGO) influence the layering, orientation, and shear response of ions [29, 31, 38].

(2) What are the specific friction mechanisms in two-

dimensional (2D) confinement? Although nanoconfined ILs have been widely discussed, the analysis rarely focuses on the well-defined, 2D slit geometry formed by graphene-based layers. In this unique space, competing mechanisms such as ion squeeze-out vs. structural locking and in-plane ordering vs. collective slip become critical, but they have not been clearly defined [30, 37, 38].

(3) What are the mechanisms underlying the observed unique friction phenomena on graphene? Studies often report unique friction phenomena on graphene, such as quantized friction or superlubricity, but they rarely discuss the mechanisms underlying these effects, such as the precise rearrangement of ions, the pathways of energy dissipation, or the specific role of the surface properties of graphene [30, 31, 38].

Note that in this review, graphene-based materials are used as an umbrella term, but graphite, pristine graphene, GO, and rGO are not treated as interchangeable substrates. Graphite and pristine graphene are discussed as conductive, chemically less specific sp^2 carbon reference surfaces; substrate-supported graphene is further regarded as a substrate-coupled interfacial state with wettability and electronic screening depending on the underlying substrate. In contrast, GO is an oxygen-rich, defect-bearing surface with strong hydrogen-bonding capability and stronger molecular pinning, whereas rGO represents an intermediate case with partially restored conductivity but residual oxygen groups and defects. Accordingly, universal trends are discussed in terms of how interfacial layering, ion ordering, and shear-plane location govern nanofriction, while absolute values of slip length, friction coefficients, etc., are interpreted as substrate specific.

This review addresses these three questions by systematically combining recent findings. It addresses the following main questions: (1) How do the surface properties of graphene govern the interfacial ion structure? (2) How does the molecular architecture of ILs influence this structure and friction? (3) How do external stimuli (i.e., humidity, electric field, and temperature) tune friction by altering these structures? By integrating insights from AFM, SFA, and MD simulation studies, we clarify how the interfacial ion structure influences the nanoscale friction of confined ILs. This review provides a clear guide for designing smarter IL-graphene systems for future lubrication applications.

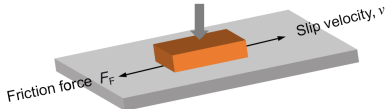
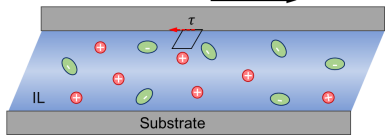
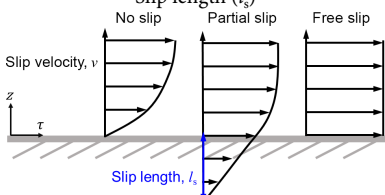
The review is structured as follows. Section 2 introduces key experimental (AFM and SFA) and computational (MD) approaches and the parameters used to quantify nanofriction. Sections 3–5 discuss how graphene surface properties, IL molecular architecture, and external stimuli (humidity, electric fields, and temperature) regulate interfacial structuring and nanofriction, respectively. Finally, Section 6 synthesizes the established structure-property relationships (Section 6.1), summarizes key challenges (Section 6.2), and outlines a roadmap toward programmable graphene-IL lubrication systems (Section 6.3).

2 Experimental and computational methods

2.1 Experimental parameters

Well-defined parameters and appropriate techniques are required to characterize the nanofriction of ILs confined on graphene-based materials. To this end, three parameters are most commonly employed, namely, friction coefficient (μ), interfacial friction coefficient (λ), and slip length (l_s). They are described below and summarized in Table 1. These parameters are complementary rather than numerically interchangeable. In this review, μ_M is used only as a macroscopic reference, whereas μ denotes a study-specific nanoscale friction coefficient extracted

Table 1 Key parameters for quantifying nanofriction in confined ILs

Parameter	Mathematical definition	Primary measurement method
Friction coefficient (μ) 	$\mu = dF_F / dF_N$	AFM friction–load curves
Interfacial friction coefficient (λ) 	$\lambda = \tau / v$	MD simulations and indirect derivation from $\lambda = \eta / l_s$
Slip length (l_s) 	$l_s = \eta / \lambda$	SFA/AFM hydrodynamic fitting and MD simulations

under a defined AFM tip radius, load range, sliding velocity, and environment. The interfacial friction coefficient λ is an intrinsic solid–liquid drag coefficient for idealized smooth interfaces and is usually obtained from molecular dynamics simulations; it should therefore not be numerically equated with μ . The slip length l_s is a hydrodynamic descriptor that is linked to λ through $\lambda = \eta / l_s$ only when the viscosity, boundary condition, and interface definition are consistent. Accordingly, throughout this review, we compare absolute values only within the same metric and methodological context, while cross-method synthesis is based on common structural trends, such as layering, ion composition, and shear-plane location, rather than on direct numerical equivalence.

(1) Friction coefficient (μ): It is derived from the equation $\mu = dF_F / dF_N$, where F_F and F_N are the friction force and normal force, respectively. μ describes the linear relationship between the friction and applied normal load, quantifying the load-dependent resistance to sliding at a macroscopic or asperity level. It is routinely extracted from AFM friction–load curves, providing a direct experimental measure of load-dependent resistance to sliding (Fig. 2(c)) [40].

(2) Interfacial friction coefficient (λ): It is defined via the constitutive relation for an ideal solid–liquid interface as the constant of proportionality between the tangential stress (force per unit area) (τ) and the slip velocity (v), i.e., $\lambda = \tau / v$. λ represents the intrinsic molecular-scale drag (friction) at an ideally smooth solid–liquid interface and is a genuine interfacial property. It can be directly computed via MD simulations [16], commonly using either nonequilibrium shear methods or equilibrium approaches based on the Green–Kubo formalism [41]. On atomically smooth surfaces, λ is also often derived indirectly from equation $\lambda = \eta / l_s$ using the slip length.

(3) Slip length (l_s): Slip length is the ratio of the bulk fluid viscosity (η) to the interfacial friction coefficient (λ), i.e., $l_s = \eta / \lambda$, which originates directly from the Navier boundary condition [42]. Its primary physical significance is geometric: it represents the distance inside the solid to which the linear fluid velocity profile must be extrapolated at zero velocity, thereby quantifying the influence of the slip condition on the flow. l_s is the central measurable parameter for quantifying interfacial slip. It is often determined by fitting hydrodynamic models to drainage

force–distance curves measured using SFA or AFM [43], as illustrated in Fig. 2(d). Using this experimentally accessible parameter, the interfacial friction coefficient (λ) is indirectly obtained.

2.2 Methods

Three methods are commonly used for probing nanoscale friction, namely, AFM, SFA, and MD simulations. AFM is the most widely used technique for probing nanoscale friction at IL-confined graphene interfaces (Fig. 2(a)). Normal force–distance measurements directly reveal oscillatory ion layering near the graphene surface (Fig. 2(b)) [43], while lateral force mapping quantifies the corresponding friction coefficient (μ) with nano-Newton sensitivity (Fig. 2(c)) [40]. Furthermore, colloid probe AFM, which utilizes a micron-sized spherical particle instead of a sharp tip, enables the reliable determination of the l_s at IL–graphene interfaces (Fig. 2(d)). This spherical probe configuration has a well-defined contact geometry and a reduced contact pressure, establishing a direct relationship between the interfacial ion structure, boundary slip, and nanofrictional dissipation. Collectively, these AFM measurements provide fundamental insights into the correlation between the interfacial ion structure, slip length, and nanoscale energy dissipation in IL–graphene systems.

The SFA provides molecular-resolution measurements of normal and shear forces in confined IL films [44–46]. Using the fringes of equal chromatic order interferometry [47–49], the SFA resolves oscillatory force profiles that reflect discrete ion-layer squeeze-out (Fig. 3) [50, 51]. This capability makes the SFA particularly useful for quantifying the layer spacing, shear modulus, and film stability under confinement [52–56]. While traditionally restricted to transparent substrates such as mica, SFA studies have established benchmark evidence for ion layering and frictional transitions [57–59], and comparisons with graphene surfaces highlight the distinct confinement effects of atomically smooth carbon interfaces [60].

MD simulations provide atomistic insights into the structuring and dynamic response of ILs at graphene interfaces (Fig. 4) [61]. Equilibrium MD simulations reveal pronounced layering, with the oscillatory organization of cations and anions extending several

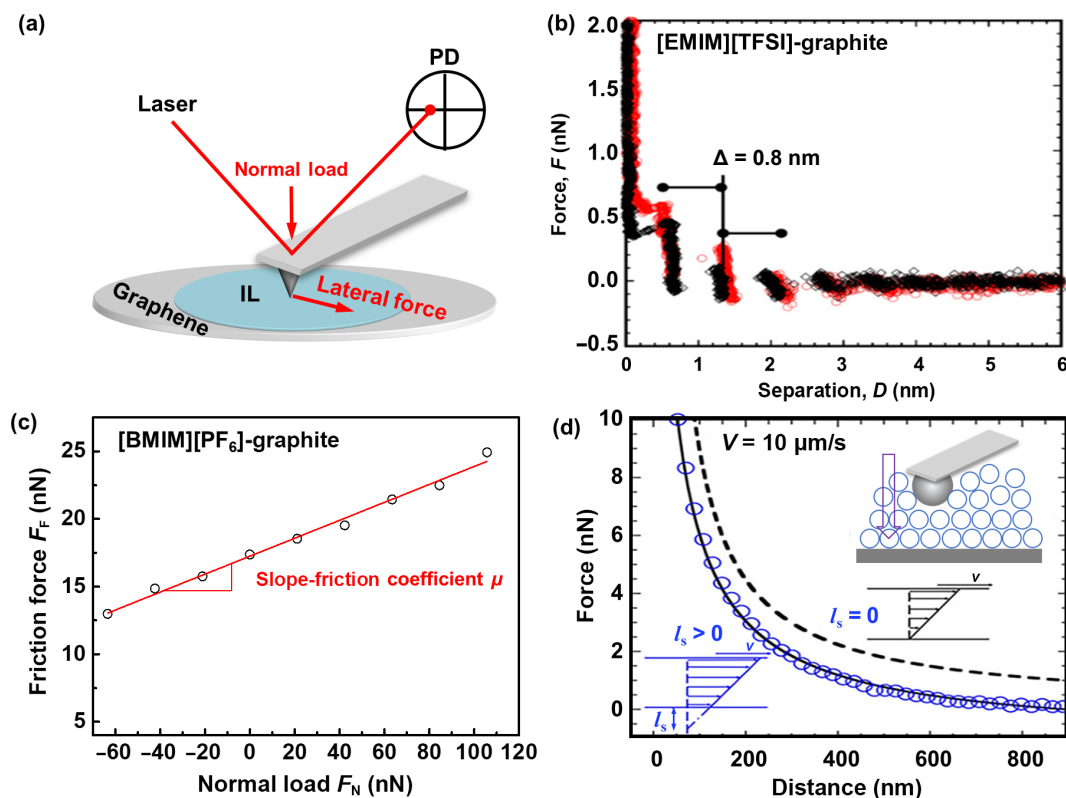


Fig. 2 AFM measurements of confined ILs at graphene interfaces: (a) schematic of an AFM setup, (b) normal force–distance curve of 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([EMIM][TFSI]) on a graphene surface showing ion layering, and (c) lateral force trace used to determine the friction coefficient of 1-butyl-3-methylimidazolium hexafluorophosphate ([BMIM][PF₆]) on a graphite surface using a SiO₂ AFM colloid probe. The fitting slope in (c) represents the friction coefficient (μ), based on Amontons' law of friction. (d) Colloid probe AFM measurement of the slip length (l_s) at an IL–graphene interface. The experimental force–distance curve (spherical line) is compared with theoretical fits (solid and dashed lines) for partial-slip and no-slip boundary conditions, respectively. The insets in (d) illustrate the velocity profiles and experimental configuration. Reproduced with permission from Ref. [43] for (b, d), © American Chemical Society 2021; Ref. [40] for (c), © The Author(s) 2021.

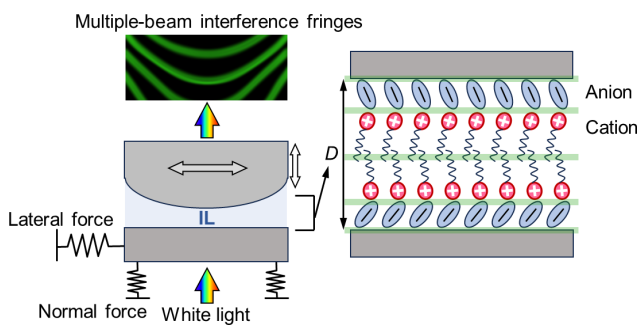


Fig. 3 Schematic of an SFA for measuring normal and lateral forces across confined IL films, in which white-light multiple-beam interferometry is used to resolve the molecular layering [49]. Reproduced with permission from Ref. [54], © The Author(s) 2013.

nanometers from the surface. Notably, variations in the alkyl chain length of the imidazolium cation systematically modulate the interfacial structure: short alkyl chains permit anions to approach the graphene surface more closely, whereas long alkyl chains (e.g., [C₈MIM]⁺) spread laterally, forming a shielding layer that pushes anions away from the interface and promotes more continuous multilayer ordering. Complementing these structural insights, NEMD simulations, by mimicking sliding through applied shear, allow the direct computation of the interfacial friction coefficient, providing a molecular counterpart to experimental friction–force measurements. This capability is exemplified by studies that have connected interfacial structural rearrangements, such as the load-induced reorientation of long

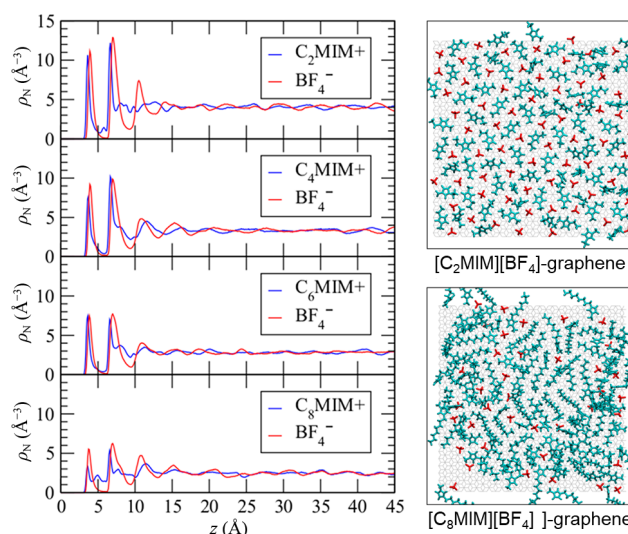


Fig. 4 MD simulations of chain length-dependent IL structuring at a graphene interface. (a) Number density profiles of cations (blue) and anions (red) for [C_nMIM][BF₄] ILs with different alkyl chain lengths ($n = 2, 4, 6, 8$), showing oscillatory multilayer organization. (b) The corresponding interfacial configurations of [C₂MIM][BF₄] and [C₈MIM][BF₄] within 6 Å above the graphene surface, illustrating that longer alkyl chains promote more continuous layering and push anions away from the graphene surface. Reproduced with permission from Ref. [61], © American Chemical Society 2019.

alkyl chains triggering superlubricity [62] or the negative friction–load dependence in IL mixtures [63], to the exceptional

frictional response of graphene-confined IL films. Moreover, MD enables the systematic exploration of variables that are difficult to experimentally decouple, such as surface charge density [64], ion symmetry, and external stimuli. Although the predictive accuracy of MD simulations depends on the force field selection and simulation timescale, they remain indispensable for linking experimental observations (e.g., AFM force mappings and spectroscopic signatures) with molecular-level mechanisms, providing a microscopic picture of how the IL composition and ion architecture govern graphene/IL interfacial properties.

In summary, AFM, SFA, and MD simulations constitute a complementary toolbox for probing IL–graphene nanofriction. Their respective strengths and limitations are summarized in Table 2 [65–67]. Despite individual constraints, combining these methods has enabled key discoveries (e.g., substrate-induced ion ordering), providing a foundation for rationally designing IL–graphene lubrication systems.

3 Influence of the surface properties of graphene on the nanofriction of confined ILs

The interfacial structure and nanofrictional properties of ILs confined on graphene are strongly governed by the nature of the graphene substrate. According to recent MD simulation studies,

the nanofrictional behavior of ILs confined in two-dimensional graphene and GO nanochannels, which is one representative form of graphene surface nanostructuring, is critically determined by geometric confinement and the surface chemistry of the confining walls [15, 16]. These characteristics of the substrate and ion structures of ILs coregulate ion–substrate and ion–ion interactions, restructuring interfacial ion arrangements and ultimately governing frictional dissipation. Specifically, the channel width and surface chemistry jointly modulate the dynamic friction coefficient and slip length, defining the nanofrictional response of confined ILs.

ILs confined within graphene- or GO-based nanochannels exhibit interfacial structures that are markedly different from their bulk counterparts. As illustrated by Wang et al. [15] (Fig. 5), the channel width influences the degree of layering and mobility of ILs: when the channel width is comparable to the ion pair dimension, only a few layers are formed, resulting in strong oscillatory ordering and restricted mobility. As the channel widens, ILs exhibit bulk-like packing and mobility, leading to reduced confinement effects. Therefore, geometric confinement establishes the baseline interfacial structure and frictional response of confined ILs. In contrast, surface chemistry exerts a more decisive influence. GO, containing abundant hydroxyl, epoxy, and carbonyl groups, forms heterogeneous interactions with ILs.

Table 2 Comparison of AFM, SFA, and MD simulation techniques for probing nanofriction and interfacial structuring of graphene-confined ILs

Technique	Strength	Limitation
AFM	(1) Nanoscale spatial resolution: It is capable of friction mapping and imaging with a nanometer-scale lateral resolution. (2) Broad material compatibility: It can be used to study diverse, opaque, and rough real-world materials in various environments. (3) Study of practical interfaces: It is ideal for investigating boundary lubrication on real, heterogeneous surfaces.	(1) Challenging absolute separation determination: The tip geometry significantly influences AFM results. (2) Potential sample alteration: The scanning tip can disrupt or displace the native lubricant layer. (3) Limited to near-surface phenomena: It is not well-suited for studying thick, continuous fluid films.
SFA	(1) Precise absolute distance measurement: It directly measures the absolute thickness of lubricant films with subnanometer accuracy. (2) Controlled high-load testing: Owing to its high load capacity, film failure and pressure limits can be studied. (3) Study of continuum rheology: It allows the probing of the rheology (e.g., viscosity and shear response) of confined, continuous fluid films.	(1) Stringent sample requirement: Its use is generally limited to atomically smooth, transparent surfaces (e.g., mica). (2) Lacks spatial resolution: It can only measure average friction and force properties over a large contact area. (3) Low experimental throughput: Experiments are time-consuming and have low efficiency.
MD simulation	(1) Atomic-scale resolution: It provides fundamental, atomic-level mechanistic insights into molecular interactions and dynamics. (2) Perfect variable control: It enables precise control of all system variables (chemistry, temperature, and pressure). (3) Calculation of fundamental properties: It can calculate intrinsic properties (e.g., theoretical interfacial shear strength).	(1) Extremely limited scales: Simulations are restricted to nanosecond timescales and nanometer-length scales, falling short of most experimental conditions. (2) Accuracy dependency: Results are highly dependent on the quality and applicability of the chosen force field and interatomic potential. (3) High resource demand: Simulations require significant computational resources (i.e., processing power and time).

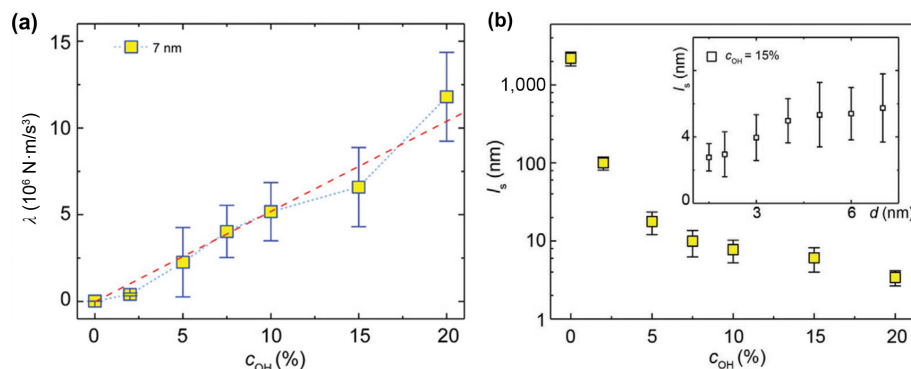


Fig. 5 Effect of the surface chemistry and nanochannel size on confined ILs. (a) Plot of the interfacial friction coefficient (λ) as a function of the hydroxyl concentration on GO walls. (b) Plot of the slip length (l_s) as a function of the hydroxyl concentration. The inset in (b) shows the dependence of the l_s on the nanochannel width. Reproduced with permission from Ref. [15], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2019.

Specifically, hydroxylation disrupts Coulombic ordering, strengthens hydrogen-bond networks, and enhances solid-liquid coupling, amplifying friction. These combined effects are quantified by the interfacial friction coefficient (λ) and slip length (l_s), as shown in Fig. 5. Unlike the conventional friction coefficient (μ), which relates the friction force to the applied normal load, the λ reported in Refs. [15, 16] characterizes energy dissipation at the nanoscale. Figure 5(a) shows that increasing the hydroxyl content in GO nanochannels sharply increases λ , reflecting increased solid-liquid coupling. Conversely, Fig. 5(b) shows that l_s considerably decreases with hydroxylation, indicating the stronger molecular pinning of IL molecules and reduced interfacial slip. Importantly, these trends are further modulated by the channel width: narrower channels intensify ion-wall interactions, magnifying the increase in λ and the reduction in l_s . Wider channels mitigate such effects by allowing more bulk-like ion organization. The coupled variation in λ , l_s , and channel width paints a coherent mechanistic picture: narrow channels with hydroxyl-rich GO walls maximize interfacial ordering and hydrogen-bond anchoring, resulting in high λ and low l_s values, implying strong friction. In contrast, wider and less-functionalized channels yield low λ and high l_s values, corresponding to smoother slip and reduced dissipation. This highlights how channel geometry and surface chemistry act synergistically to restructure confined ILs and govern their nanofrictional behavior.

The substrate supporting the graphene layer also plays a decisive role in regulating nanofriction. AFM-based experimental studies have shown that the supporting substrate can effectively dope graphene: Cu substrates induce *n*-doping, whereas SiO₂ substrates induce *p*-doping, thereby modulating its charge carrier density and surface wettability [43]. This doping-dependent wettability governs the balance between IL-graphene and IL-IL interactions at the interface [18, 68, 69]. Specifically, *p*-doped graphene on SiO₂ disrupts interfacial ordering and promotes heterogeneous ion adsorption (Fig. 6(a)), whereas *n*-doped graphene on Cu improves dispersion interactions and stabilizes ordered multilayer structures (Fig. 6(b)) [43]. As illustrated in Fig. 6(c), these substrate-induced variations in interfacial layering directly affect the slip length: ordered, well-correlated IL layers on graphene/Cu result in larger slip lengths and lower energy dissipation, whereas disordered structures on graphene/SiO₂ result in reduced slip lengths and higher friction.

In addition, recent AFM experiments have shown that local surface corrugation can also markedly alter the interfacial structure and nanofriction of ILs on graphene. Zheng et al. [70]

investigated 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([EMIM][TFSI]) on single-layer graphene transferred onto 20 nm silica nanoparticle films, where the graphene formed convex peaks and concave valleys, the latter corresponding to locally suspended regions. They found that the friction coefficient depends strongly on the local curvature and that the electro-tunability of friction at low loads is significantly enhanced on peaks and valleys compared with smooth graphene. Normal force measurements further suggested that both convex and concave interfaces support thicker boundary IL films, although through different mechanisms: promoted overscreening on convex regions and steric hindrance on concave regions. These results indicate that graphene roughness or corrugation is not merely a perturbation to the flat interface case but can fundamentally modify interfacial layering and frictional dissipation. To the best of our knowledge, however, direct IL nanofriction studies on moiré-patterned graphene remain scarce. Existing work on these systems has mainly focused on related but nontribological topics, such as interfacial water friction on moiré-patterned graphene [71]. This highlights surface corrugation and nonideal graphene topographies as an important open direction for future graphene-IL nanotribology.

MD simulations of 1-ethyl-3-methylimidazolium tetrafluoroborate ([EMIM][BF₄]) in graphene-coated silica nanochannels show that graphene promotes the layering effect of ILs, producing sharper density oscillations and more ordered interfacial arrangements [72]. By screening the strong hydroxyl anion interactions between silica and ILs, graphene also homogenizes the charge distribution and reduces the asymmetry between cation and anion adsorption. Such structural reinforcement can suppress ion diffusion in the near-wall region while simultaneously promoting positive slippage at the wall, thereby facilitating faster overall transport in graphene-coated nanochannels. Therefore, graphene not only strengthens interfacial layering but also tunes the charge ordering and dynamic properties of confined ILs.

The surface charge density of graphite also has a considerable influence on the interfacial structure and nanofriction of confined ILs [64]. MD simulations and AFM experiments show that changing the surface charge density controls the ionic composition and orientation at the graphite interface. Specifically, negative charges enrich cations, positive charges enrich anions, and intermediate states yield mixed layers. These charge-induced structural changes shift the position of the slip plane and change the dominant pathways for energy dissipation. As shown in Fig. 7,

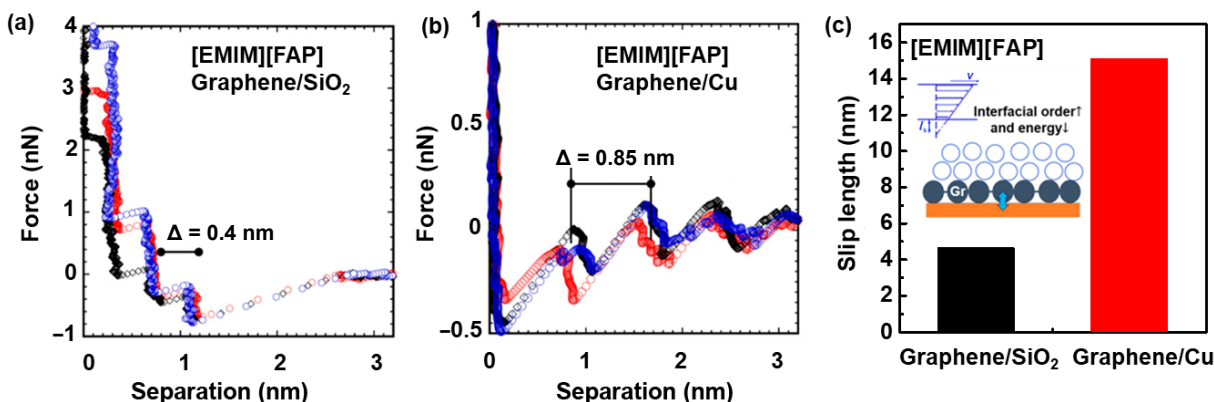


Fig. 6 Structural forces and slip lengths of 1-ethyl-3-methylimidazolium tris(pentafluoroethyl)trifluorophosphate ([EMIM][FAP]) confined on graphene supported by (a) Gr/SiO₂ and (b) Gr/Cu substrates. (c) Comparison of the slip length for the two cases. Reproduced with permission from Ref. [43], © American Chemical Society 2021.

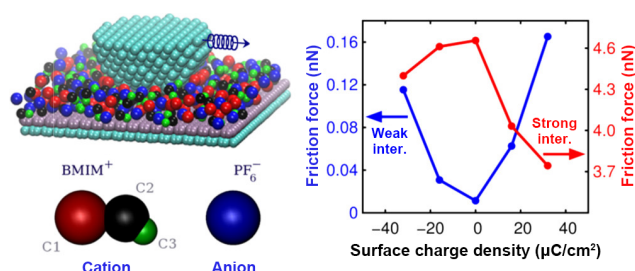


Fig. 7 MD simulations of [BMIM][PF₆] on a graphite substrate. (a) Schematic of the simulated system. (b) Friction force as a function of surface charge density under weak and strong ion–substrate interactions, highlighting the charge-dependent modulation of interfacial friction. Reproduced with permission from Ref. [64], © American Chemical Society 2018.

under weak ion–substrate coupling, friction exhibits a minimum at zero charge and increases parabolically with the surface charge density due to enhanced ion adsorption and higher sliding barriers. In this regime, the applied charge primarily strengthens the interfacial layer, making it more resistant to shear. In contrast, under strong ion–substrate coupling, friction peaks at zero surface charge density but decreases as the magnitude of the surface charge density increases (regardless of sign), reflecting a loss in commensurability between the IL structure and the tip lattice. Here, the initial neutral interface is already in a high-friction, locked state due to strong specific interactions, and introducing a charge disrupts this optimal locking, thereby facilitating slip. Therefore, the surface charge density is a powerful tool for reversibly tuning the interfacial ordering and frictional dissipation at IL–graphite interfaces, where the direction of frictional change is determined by the initial ion–substrate coupling strength.

In contrast, the effect of graphene thickness appears limited under the conditions studied. Recent AFM force–separation studies revealed that the number of graphene layers (from single- to multilayer) exerts only a negligible influence on the interfacial structuring of confined ILs. For 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([EMIM][TFSI]), oscillatory layering transitions and the overall attractive interaction between the tip and graphene were essentially independent of whether the substrate was single-layer graphene, bilayer graphene, few-layer graphene, or multilayer graphene. This indicates that the nanostructure of ILs at the graphene interface is primarily governed by the outermost graphene layer, with deeper layers playing little role [73]. The persistence of the confined IL film between the probe and the graphene surface can suppress direct tip–surface contact, therefore minimizing thickness-dependent puckering effects.

It is noteworthy that graphene differs from other frequently studied solid surfaces (e.g., mica, Au, and silica) in IL nanotribology. On layered, atomically smooth, hydrophilic, and

negatively charged mica, SFA studies have revealed pronounced oscillatory structural forces, discrete ion-pair squeeze-out, and even quantized friction, reflecting strong electrostatic adsorption and mechanically robust ionic layering between atomically smooth, hydrophilic sheets [44, 56]. On nonlayered metallic Au(111), AFM studies have shown potential-dependent multilayer electrical double layers and nanoscale friction control, with more near-surface layers detected at higher potentials [74, 75]. On a nonlayered oxide silica surface, by contrast, strong polarity and nanoscale roughness substantially modify ionic layering and lubricity [76, 77]. Relative to these surfaces, pristine graphene lacks a fixed surface charge and abundant hydrogen-bonding sites, so its interfacial organization is more strongly governed by dispersion and π -related interactions, while its frictional response is additionally shaped by substrate-induced doping, wettability, and relatively large interfacial slip. Thus, graphene is distinctive by combining atomic flatness with electronically tunable yet chemically less specific interfacial interactions.

4 Effect of the molecular architecture of ILs on nanofriction in confined ILs

The nanofrictional response of ILs confined on graphene-based surfaces is not only determined by the geometry and chemistry of the substrate but also strongly governed by the molecular architecture of the ILs. Variations in the cation and anion dictate how ILs interact with graphene and with each other at the interface. These changes dictate the ion–graphene interactions and the resulting layered structure of the confined IL film, which ultimately governs the friction on graphene-based surfaces. For example, aromatic cations (including imidazolium-type species) can interact with graphene via π – π interactions, which may stabilize interfacial adsorption and ordering [78], whereas bulky phosphonium cations can introduce steric hindrance and inefficient packing, leading to higher shear resistance [67]. Similarly, the alkyl side-chain length can markedly influence lubricity, but its effect is strongly system dependent. In some ILs, increasing alkyl chain length has been associated with higher viscosity and, under specific test conditions, higher friction coefficients [79]. However, no universal conclusion can yet be drawn. Depending on the cation family, accompanying anion, surface potential, and confinement conditions, longer alkyl chains may either stabilize ordered boundary layers and lower friction or introduce steric frustration and greater dissipation. One illustrative case in which longer alkyl chains reduce friction is the comparison between trihexyl(tetradecyl) phosphonium bis(trifluoromethylsulfonyl)imide ([P_{6,6,6,14}][TFSI]) and triethyl(butyl)phosphonium bis(trifluoromethylsulfonyl)imide ([P_{4,4,4,1}][TFSI]) on graphite under open circuit potential

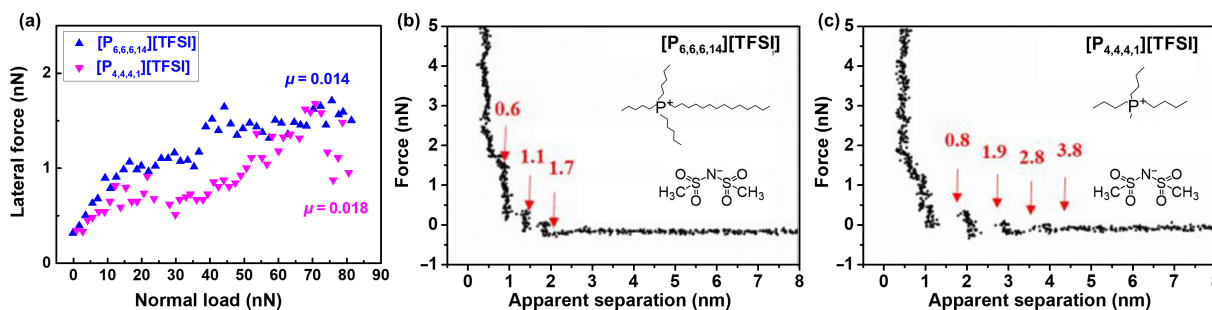


Fig. 8 Friction and force–distance measurements of two phosphonium ILs on graphite surfaces. (a) Friction force vs. load curves of [P_{6,6,6,14}][TFSI] and [P_{4,4,4,1}][TFSI]. Force–distance curves of (b) [P_{6,6,6,14}][TFSI] and (c) [P_{4,4,4,1}][TFSI]. Reproduced with permission from Ref. [80], © American Chemical Society 2021.

(OCP) conditions (Fig. 8) [80]. Both ILs have the same [TFSI][−] anion, yet they exhibit different frictional responses due to the different lengths of their phosphonium cations (Fig. 8(a)). The force–distance profiles in Figs. 8(b) and 8(c) show step widths of ~ 1.1 and ~ 0.9 nm for [P_{6,6,6,14}][TFSI] and [P_{4,4,4,1}][TFSI], respectively, consistent with ion-pair dimensions assuming cubic packing. The long alkyl substituents in [P_{6,6,6,14}]⁺ enhance lateral cohesive interactions among confined ions, creating a shielding layer that pushes anions away from the interface and facilitates the formation of a more continuous multilayer ordering [61]. This structural reinforcement directly translates into a lower friction coefficient in [P_{6,6,6,14}][TFSI] than in [P_{4,4,4,1}][TFSI]. These observations demonstrate that even when the anion is kept constant, variations in their cation chain length can decisively tune interfacial layering [81–83] and energy dissipation, offering a handle for molecular design to optimize IL lubricity on graphene-based surfaces.

Beyond these tetraalkylphosphonium systems, imidazolium-based ILs with even longer alkyl substituents provide further insights into how the cation chain length stabilizes layering and lowers friction. Lu et al. [84] demonstrated that the chain length of the cation plays a critical role in determining the interfacial structure of ILs confined on graphene surfaces, which in turn significantly influences their nanofrictional properties. As shown in Fig. 9, longer alkyl chains promote more ordered and compact layering at the IL–graphene interface due to enhanced lateral cohesive forces between ions. These well-organized boundary layers, especially those formed by ILs with longer alkyl chains (e.g., 1-octadecyl-3-methylimidazolium bis(trifluoromethylsulfonyl) imide, ([C₁₈mim][NTf₂)]), facilitate smooth interfacial sliding and reduce energy dissipation during shear, lowering the friction coefficient. In contrast, shorter alkyl chains yield less ordered interfacial structures, resulting in higher friction and greater

energy dissipation due to weaker ion–ion interactions. Increasing the chain length stabilizes a robust ordered structure; longer alkyl tails enhance dispersion-driven tail aggregation and packing, which helps sustain friction-reducing boundary ordering [85]. These findings highlight the importance of the chain length as a key design parameter for optimizing IL lubricity on graphene-based surfaces [86]. The structure–property relationships observed in the ILs with varying cation alkyl chain lengths underscore the potential of tailoring the molecular structures of ILs to achieve desirable nanofrictional performance.

Interestingly, conflicting trends have been reported regarding the effect of the cation alkyl chain length on interfacial ordering. While some studies have concluded that longer chains promote more ordered and compact layering at the IL–graphene interface, others have reported the opposite. MD simulations of [C_nmim][BF₄] ($n = 2–8$) show that shorter chains enable denser packing of ions at the graphene surface, whereas longer chains introduce steric hindrance and unfavorable orientations, which weaken interlayer interactions [87]. These apparent contradictions imply that the effect of alkyl chain length is not universal but depends on a complex interplay between intrinsic IL properties and external conditions. An illustrative example of the coupled effect is the interplay between ionic size matching and the applied potential. As demonstrated by Zhang et al. [80], an applied potential on a graphite surface electrostatically switches the boundary layer composition between cation- and anion-enriched states. However, the lubricity of each state is critically governed by the ionic size complementarity. For example, at the OCP, where ions are mixed, a mismatch between a large cation and a small anion leads to a rough, disordered interface and high friction. Conversely, superlubricity is achieved only when a specific potential (e.g., -1.0 V) enriches a well-matched, structurally coherent layer (e.g., long-chain cations forming a robust film).

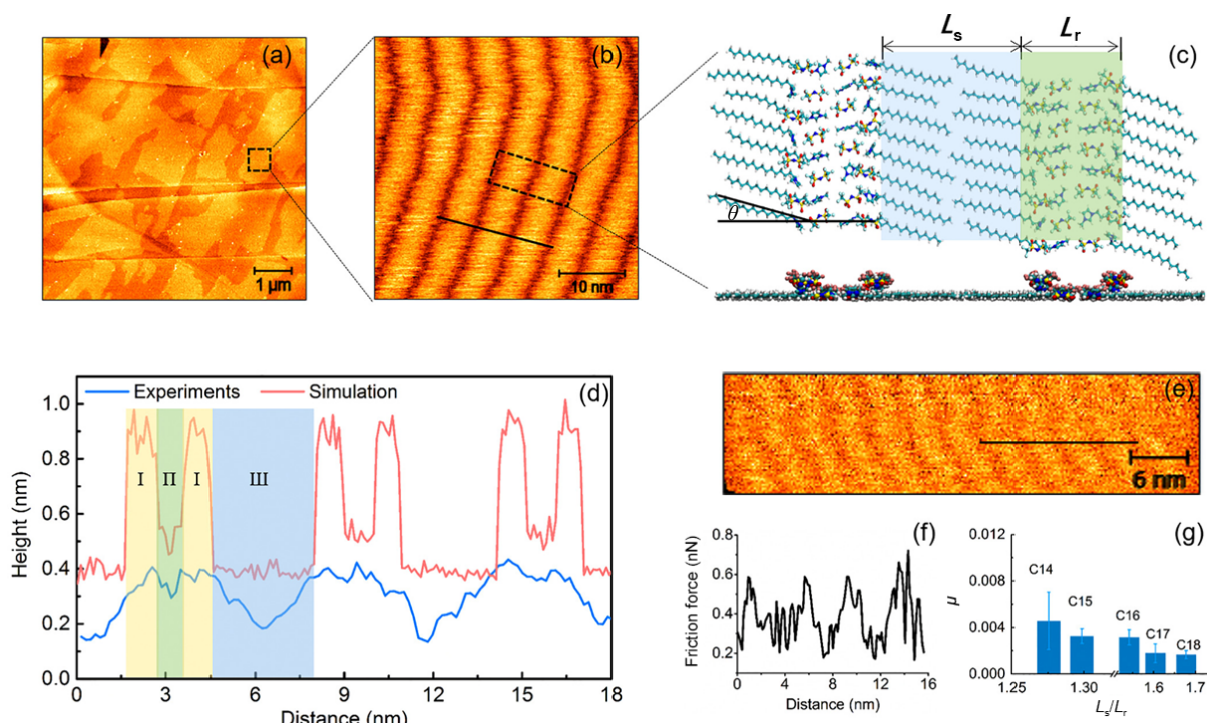


Fig. 9 Multiscale characterization of the self-assembled structure and nanofrictional properties of [C₁₈mim][NTf₂] on highly oriented pyrolytic graphite. (a, b) AFM images showing the large- and atomic-scale striped ordering. (c, d) Simulation and experimental results revealing the ordered alkyl chain regions (L_s) and disordered imidazolium/anion regions (L_r). (e, f) Friction force mapping, indicating the periodic variations correlated with the striped structure. (g) The L_r/L_s ratio shows a strong negative correlation with the friction coefficient in ILs with different alkyl chain lengths. Reproduced with permission from Ref. [84], © American Chemical Society 2022.

This shows that while the surface charge can reconfigure the interface, the final performance is dictated by the inherent structural compatibility of the involved ions. Therefore, resolving discrepancies requires decoupling intertwined factors (e.g., ion symmetry, anion type, potential, and confinement conditions) to formulate more general design rules.

While the cation structure has received much attention, the type and size of the anion are equally decisive in shaping interfacial ordering and nanofriction. For example, among ILs, 1-ethyl-3-methylimidazolium chloride ([EMIM][Cl]), an imidazolium-based IL, exhibits the strongest adsorption and most stable layered structure at the graphene interface, followed by 1-ethyl-3-methylimidazolium nitrate ([EMIM][NO₃]) and 1-ethyl-3-methylimidazolium thiocyanate ([EMIM][SCN]). In contrast, [EMIM][BF₄] forms weaker, more diffuse layers because it has a bulky and charge-delocalized anion [88]. Long-chain anions, such as dioctyl sulfosuccinate ([AOT][−]) [72], have a polar headgroup and extended alkyl tails, which allow them to form highly ordered stratified arrangements at the interface. In these systems, the polar headgroups remain adsorbed at the graphene surface, while the extended tails align laterally and promote the development of well-defined ... -anion-cation-anion-cation-... multilayered

structures (Figs. 10(a)–10(c)). In contrast, short anions such as [BF₄][−] lack long hydrophobic substituents and instead form simpler alternating...-cation-anion-... monolayers (Figs. 10(d)–10(f)), leading to less pronounced ordering and weaker interfacial cohesion [89]. From a nanofriction perspective, these differences in interfacial structuring imply that long-chain anions stabilize more robust boundary layers, which resist shear and enhance slip by maintaining distinct multilayer organization [31, 90, 91]. In contrast, short anions produce less stable monolayer arrangements, which are more easily disrupted during sliding, resulting in higher friction coefficients [92]. In the case of amino acid-based ILs on graphene surfaces, long alkyl side chains of the anion have a negative effect on interfacial structuring. Short, compact anions, such as [PF₆][−], enable multilayered, well-defined ordering, extending nearly 20 Å from the graphene surface. Long amino acid anions such as alaninate and isoleucinate anions yield progressively thinner ordered regions, with the isoleucinate anion producing only a single dense layer before bulk-like disorder emerges. This trend is accompanied by reduced ion adsorption in the innermost layer, a lower charge concentration, and slower ion diffusion, all arising from stronger van der Waals side-side interactions among bulky anions. Consequently, increasing the

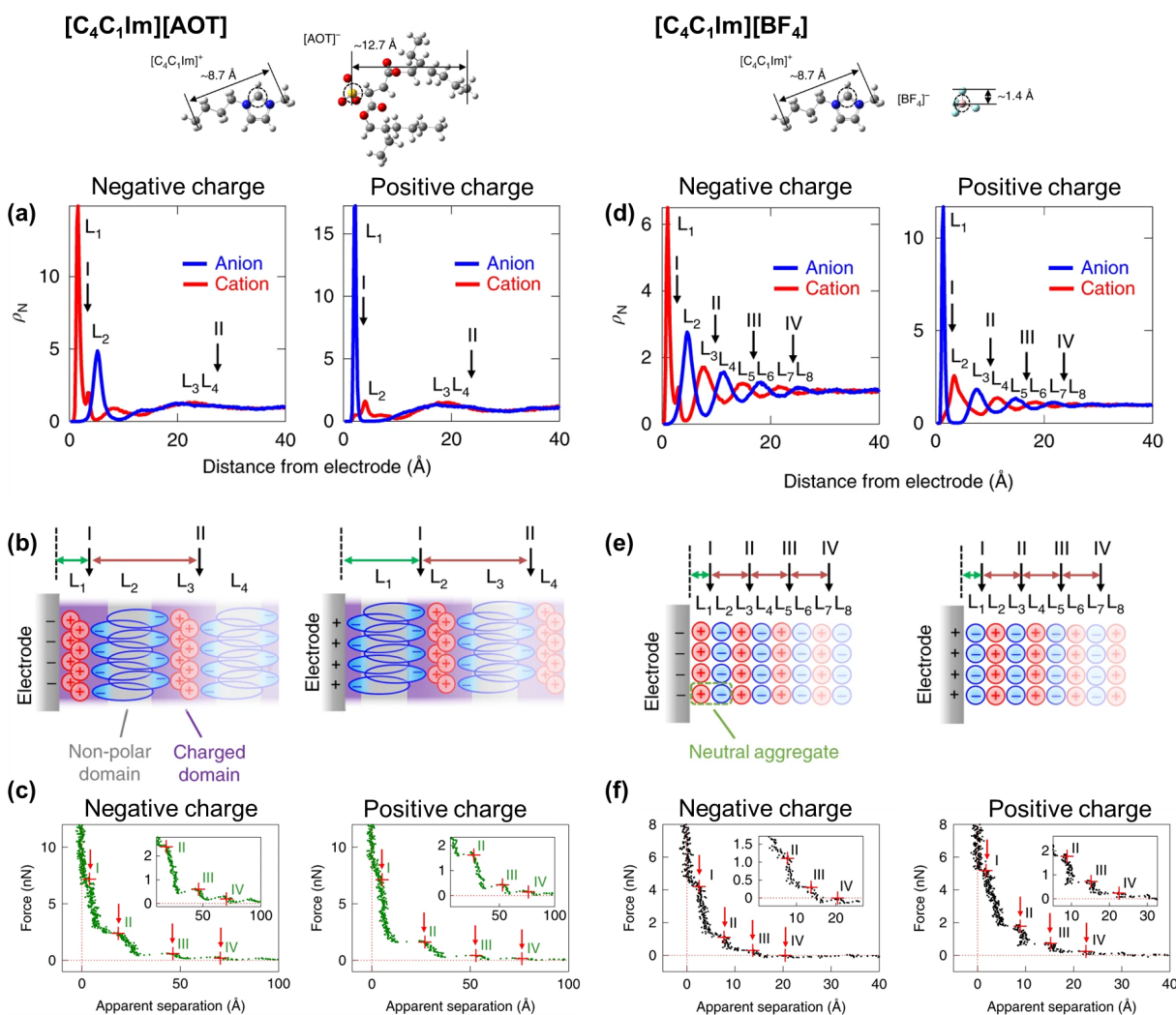


Fig. 10 Interfacial layering of ILs at charged graphite surfaces. (a–c) 1-Butyl-3-methylimidazolium tetrafluoroborate ([C₄C₁Im][BF₄]) and (d–f) 1-butyl-3-methylimidazolium 1,4-bis(2-ethylhexoxy)-1,4-dioxobutane-2-sulfonate ([C₄C₁Im][72]). (a, d) Number density profiles (ρ_N) of cations and anions from MD simulations. (b, e) Schematic representations of ion arrangements at negatively (left) and positively charged (right) electrodes. (c, f) Force–distance curves measured with a silica colloid probe on graphite at negative (left) and positive (right) potentials. The insets in (c, f) show low-force regimes. Red crosses (I–IV) denote discrete layering steps and the corresponding rupture forces. Reproduced with permission from Ref. [89], © The Author(s), under exclusive licence to Springer Nature Limited 2019.

anion side-chain length weakens interfacial layering and destabilizes the boundary film [93]. These findings underscore the crucial roles of the size and structure of anions in interfacial ordering and, consequently, the nanofrictional performance of ILs confined on graphene-based surfaces.

These findings demonstrate that the molecular architecture of ILs plays a decisive role in determining nanofriction on graphene surfaces. Long alkyl chains in cations enhance lateral cohesive forces and promote compact, well-ordered layering, thereby stabilizing boundary films and lowering friction. Likewise, long-chain anions can assemble into multilayered stratified structures that resist shear, whereas short-chain anions produce simpler monolayers that are more easily disrupted, resulting in higher friction coefficients. In contrast, bulky asymmetric cations could break interfacial ion ordering and increase shear resistance, whereas symmetric aromatic cations, such as imidazolium, favor π - π interactions with graphene and stabilize ordered layers. Therefore, no universal rule can yet be drawn for alkyl chain length alone. The nanofrictional response emerges from the balance between lateral cohesion, steric hindrance, ion size matching, and operating conditions.

5 Effect of external stimuli on the nanofriction of graphene-confined ILs

Beyond substrate properties (Section 3) and the molecular architecture of ILs (Section 4), external stimuli provide additional degrees of freedom to actively regulate friction at IL-graphene interfaces. The nanofrictional behavior of confined ILs is not fixed but highly tunable using factors such as humidity, electric field, and temperature. These perturbations reshape interfacial ion layering, orientation, and cohesion, which in turn affect the slip length, shear resistance, and overall frictional dissipation. This section analyzes the effect of three external stimuli, i.e., humidity, electric fields, and temperature, on the nanofriction of graphene-confined ILs.

5.1 Effect of humidity on the friction of confined ILs

Humidity introduces competitive interactions at the IL-graphene interface. The presence of water considerably modifies the interfacial structure of ILs confined on graphene and graphite

[94]. Water molecules exhibit a strong preference to adsorb on the graphene surface, particularly under positive polarization, where they compete directly with anions for adsorption sites. This competitive adsorption partially replaces IL ions at the interface, disrupting the well-defined multilayered ionic arrangement and diminishing long-range order [46, 95, 96]. Moreover, water forms hydrogen bonds with interfacial ions (preferentially with anions), which promotes ion-water coadsorption and disrupts the well-defined ionic layering. As illustrated in Fig. 11, in hydrophobic ILs such as 1-butyl-1-methylpyrrolidinium bis(trifluoromethylsulfonyl) imide ([pyr14][TFSI]), water molecules are readily electrosorbed on polarized graphene; specifically, under a positive bias, they form hydrogen bonds with anions and partially replace the first ionic layers. This process disrupts ion layering, reduces long-range order, and weakens the stability of the interfacial ion layer. In contrast, hydrophilic ILs such as [BMIM][BF₄] absorb more water in the bulk but suppress its accumulation on negatively charged graphene, maintaining more stable interfacial structures [97].

Zheng et al. [98] recently illustrated how water uptake modifies the interfacial structure of ILs on graphene (Fig. 12). In hydrophobic ILs such as 1-ethyl-3-methyl imidazolium bis(trifluoromethylsulfonyl)imide ([C₂C₁Im][TFSI]), even trace water preferentially accumulates on the positively polarized graphene surface, displacing anions and reducing the thickness of the first adsorbed layers. This leads to a shift in the potential of zero charge and a weakening of well-defined multilayer ordering. In contrast, while the more hydrophilic 1-ethyl-3-methyl imidazolium ethyl sulfate ([C₂C₁Im][EtSO₄]) incorporates water throughout the interfacial region, the layering remains relatively stable due to strong hydrogen bond networks. Therefore, water disrupts cation-anion correlations, alters the ion orientation, and diminishes long-range order, with the extent of structural change considerably dependent on the IL type [46]. Recent electrochemical studies on graphene foam electrodes with 1-butyl-3-methylimidazolium tetrafluoroborate ([BMIM][BF₄]) further confirm that humidity is a decisive factor of the interfacial structure and interfacial reactivity. Low water levels promote cation intercalation into graphene layers within the graphene foam electrode, whereas high water levels accelerate exfoliation of graphene layers and reduce the reversibility of the intercalation-deintercalation process. Simultaneously, water

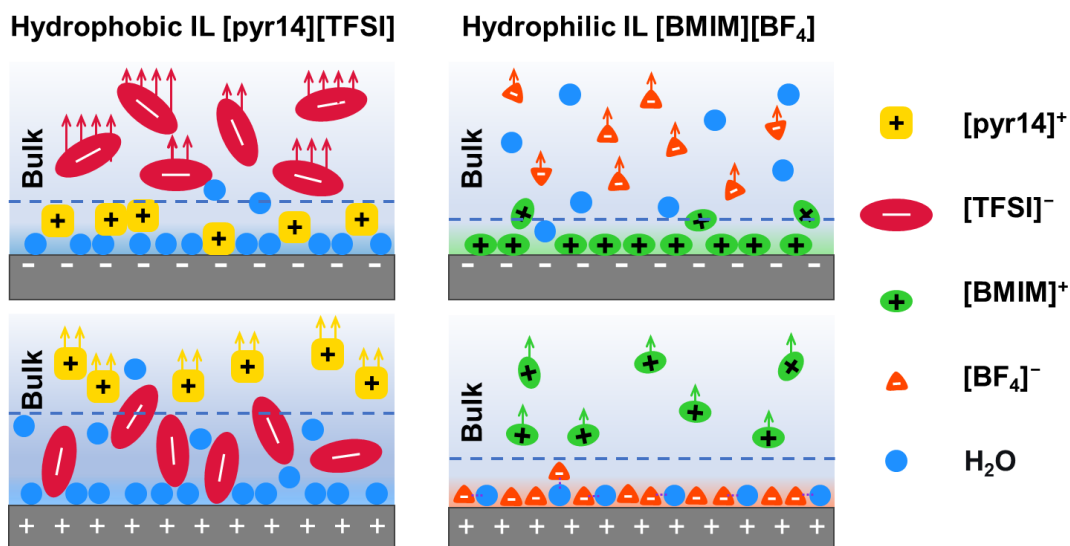


Fig. 11 Schematic comparing the interfacial responses of hydrophobic [pyr14][TFSI] and hydrophilic [BMIM][BF₄] to trace water under confinement. Drawn based on the mechanism described in Ref. [97].

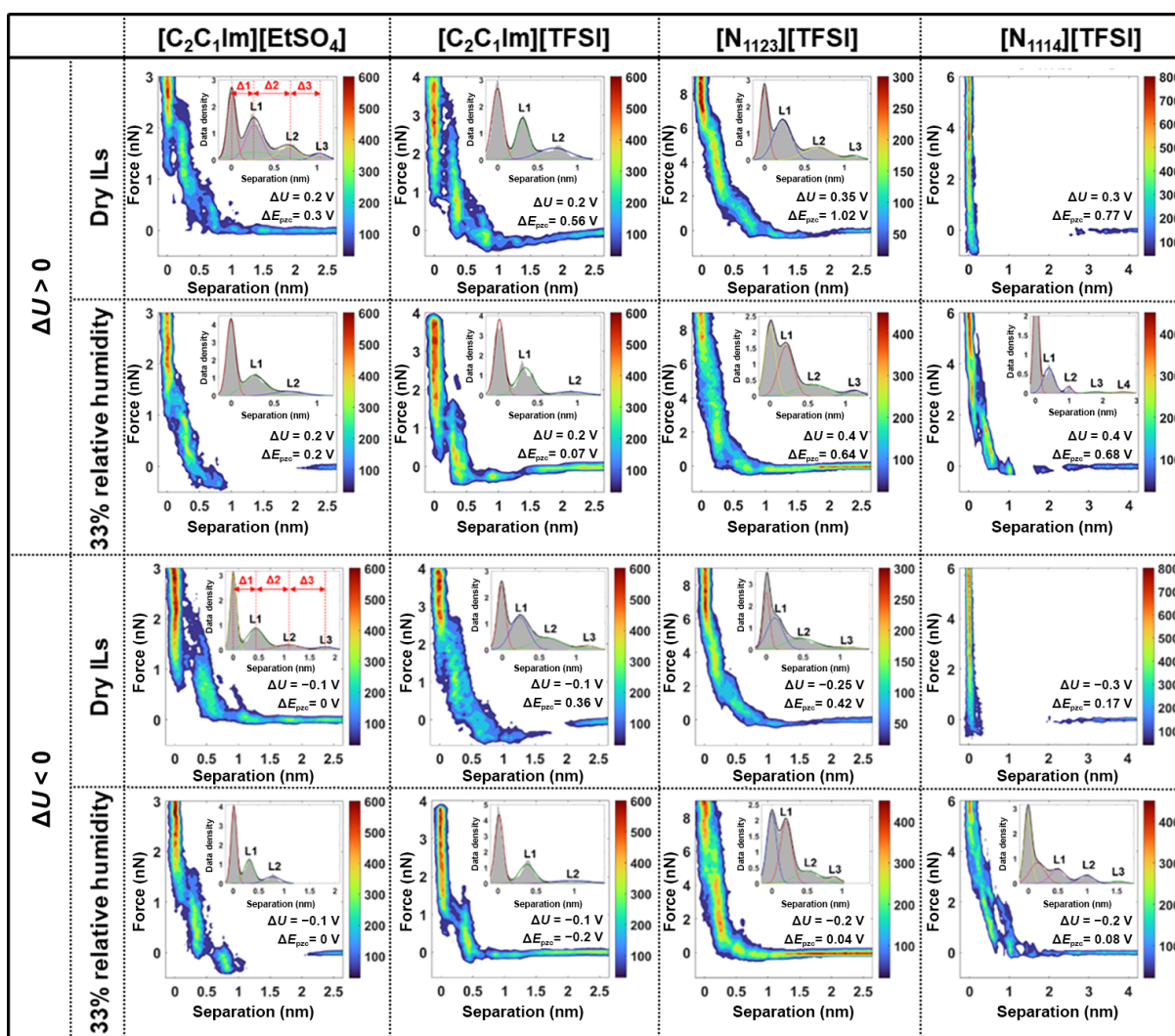


Fig. 12 Heatmaps of force-distance curves for $[C_2C_1Im][EtSO_4]$, $[C_2C_1Im][TFSI]$, ethyldimethylpropylammonium bis(trifluoromethylsulfonyl)imide ($[N_{1123}][TFSI]$), and butyltrimethylammonium bis(trifluoromethylsulfonyl)imide ($[N_{1114}][TFSI]$) measured at 0% (dry) and 33% relative humidity. Data are shown under positive ($\Delta U > 0$) and negative ($\Delta U < 0$) applied potentials, and the insets highlight discrete layering steps (L1, L2, and L3) and the corresponding layer size ($\Delta 1$, $\Delta 2$, and $\Delta 3$). Reproduced with permission from Ref. [98], © The Author(s) 2023.

modifies the diffusion and redox kinetics of molecular redox probes, highlighting its role in tuning the structural stability of the electrode-electrolyte interface and the transport (diffusion) of redox-active species across IL-graphene junctions [99]. These structural perturbations induced by water increase shear resistance and suppress interfacial slip [100]. However, their exact dependence on water concentration and graphene surface chemistry remains insufficiently quantified.

The extent of such a disruption considerably depends on the water concentration. Even trace amounts of water can strongly perturb the layering of ILs. AFM and MD studies demonstrate that water molecules adsorbed on graphene weaken Coulombic interactions and disrupt the well-defined cation-anion layering within the confined IL film at the IL-graphene interface, leading to broader density peaks and reduced structural order [98]. At low humidity, hydrogen in water preferentially bonds with anions such as $[FAP]^-$, transforming a cation-rich surface layer into a mixed ion-water layer. This rearrangement reduces the slip length and promotes stick-slip dynamics, thereby increasing the friction coefficient [16, 43, 101, 102]. Espinosa-Marzal et al. [45] further revealed that water suppresses avalanche-like microslip events at the IL-graphene interface, which usually occur under dry

conditions, resulting in smoother but overall higher friction. At higher humidity, water adsorption on graphene is more pronounced, forming hydration layers that partially displace IL ions from the IL-graphene interface. Owing to their stronger polarity and affinity to hydrophilic sites compared to IL ions, water molecules can even replace IL ions and preferentially adsorb on the solid substrate [30]. This substitution decreases the density of ordered IL layers at the interface, diminishing long-range structural correlations and destabilizing robust boundary lubrication. Although film swelling at a high water content may relieve shear stress [100, 103, 104], overall competition between water and IL ions for adsorption at graphene disrupts cohesive ion structuring and generally impairs lubricity [45, 46, 95, 105].

5.2 Influence of the electric field on the friction of confined ILs

Electric fields are the most versatile and direct means of tuning IL nanofriction. Owing to their ionic nature, the boundary layers of ILs reorganize under an applied bias [30, 106–109] on a graphene substrate. Positive potentials attract anions to the surface, whereas negative potentials enrich cations, dynamically switching the composition and orientation of the confined layers. This

redistribution shifts shear planes and alters stick-slip events, leading to tunable friction. Importantly, under certain potentials, interfacial layering is so well ordered that friction decreases to immeasurable values, achieving potential-dependent superlubricity.

Li et al. [110] demonstrated electrically controlled nanofriction using 1-hexyl-3-methylimidazolium tris(pentafluoroethyl) trifluorophosphate ([HMIIm][FAP]) on graphite (Fig. 13). Their AFM measurements revealed that the frictional state of the IL-graphite interface can be reversibly switched by tuning the applied potential. At negative potentials, the interfacial layer is cation-rich, and friction remains relatively high due to the continuous reorientation of cations under shear. In contrast, at positive potentials, the enrichment of anions forms a smoother,

more homogeneous boundary layer, progressively suppressing stick-slip events. At +1.5 V, the interfacial layer is almost entirely anion-dominated, leading to a friction coefficient of 0.001, well within the superlubricity regime ($\ll 0.01$) [110, 123]. This highlights that electric fields not only reorganize interfacial ion layering but also act as an on/off switch for achieving superlubricity at IL-graphite interfaces.

AFM combined with quantum chemical simulations has revealed a distinct frictional response to electrical fields at the propyl ammonium nitrate (PAN)-graphite interface [111]. At OCP, the boundary layer is cation-rich, with alkyl chains lying flat on graphene, giving rise to strong solvophobic attractions and high friction (Figs. 14(a) and 14(b)). Applying positive potentials increases the anion concentration at the PAN-graphite interface

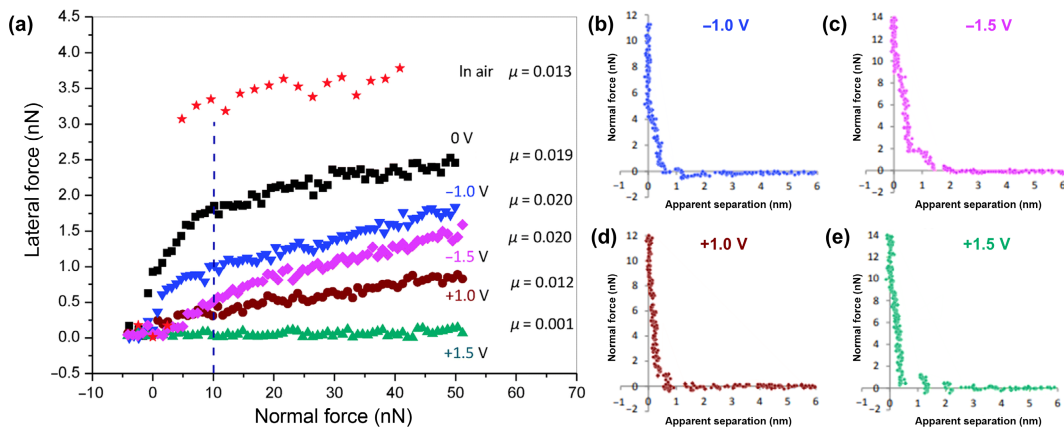


Fig. 13 (a) Lateral force vs. normal load curves of an AFM tip ($r \approx 5$ nm) sliding on graphite in air and [HMIIm][FAP] under different applied potentials, with friction coefficients (μ) extracted from the slopes at normal force > 10 nN. (b–e) Force vs. distance curves recorded in [HMIIm][FAP] at -1.0 V, -1.5 V, +1.0 V, and +1.5 V, showing potential-dependent layering at the IL-graphite interface. Reproduced with permission from Ref. [110] for (a), © The Royal Society of Chemistry 2014.

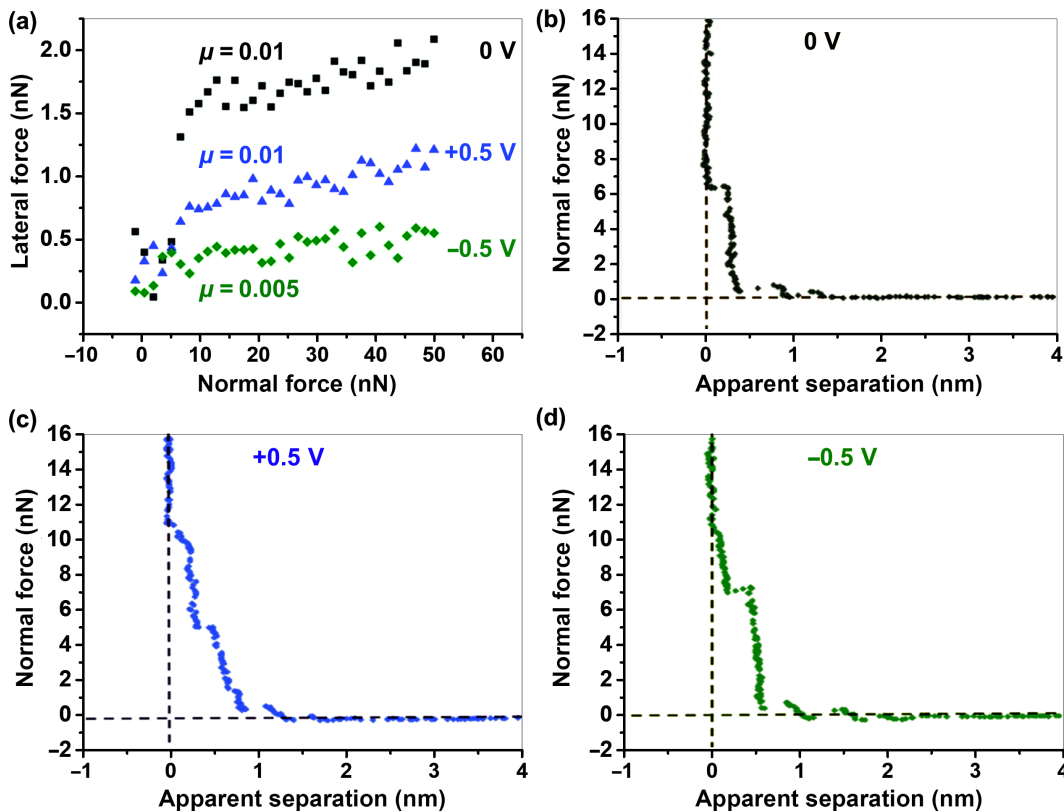


Fig. 14 (a) Lateral force vs. normal load curves of an AFM tip ($r \approx 8$ nm) sliding on graphite in propyl ammonium nitrate (PAN) at different applied potentials, with the friction coefficients (μ) extracted from the boundary regime (10–50 nN). (b–d) Normal force vs. distance curves in PAN at 0 V, +0.5 V, and -0.5 V, showing potential-dependent layering at the IL-graphene interface. Reproduced with permission from Ref. [111], © The Author(s) 2015.

and drives cations to adopt more upright conformations, reducing interlayer coupling and lowering friction by $\sim 50\%$ (Figs. 14(a) and 14(c)). Under negative potentials, the boundary layer is further enriched in cation alkyl chains but with weakened interlayer attractions, resulting in the lowest friction coefficients ($\mu \approx 0.005$; Figs. 14(a) and 14(d)). This highlights that electric fields can finely modulate the ion composition and orientation in the boundary layer of the PAN-graphite interface, directly tuning interfacial energy dissipation pathways and enabling controllable nanofriction.

Mechanistically, the electro-tunable nanofriction summarized here reflects a coupled interfacial response rather than a potential effect alone. The applied potential first sets the sign and magnitude of the surface charge and thereby selects the dominant ions in the first adsorbed layer [30, 106–109]. The frictional consequence of this charge selection is then governed by ion-specific adsorption and packing. For [HMIm][FAP] on graphite, anion enrichment at positive potentials produces a smoother and more homogeneous boundary layer and suppresses stick-slip, whereas cation-rich layers at negative potentials remain more dissipative because cations continue to reorient under shear [110]. Likewise, for PAN on graphite, the potential changes not only the interfacial composition but also the cation conformation: at OCP, the cation-rich layer with alkyl chains lying flat gives strong solvophobic coupling and high friction, while positive potentials increase anion concentration and drive cations to adopt more upright conformations, and negative potentials further enrich cation alkyl chains but weaken interlayer attractions, thereby lowering friction [111]. The surface state determines how strongly the potential-selected ions are bound. As discussed in Section 3, substrate-dependent graphene doping modifies wettability and

slip [43], while MD/AFM studies on graphite showed that the sign of the charge dependence changes with ion–substrate coupling strength [64]. Therefore, friction minima, maxima, or switching arise from the combined action of surface charging, ion-specific adsorption, and surface-dependent ion–substrate coupling.

Moreover, electrical control over the interfacial properties of the IL–graphite interface is inherently dynamic, as interfacial restructuring evolves with time even under a constant bias. Time-dependent AFM measurements (Fig. 15) have revealed that the electrical field-induced restructuring of IL boundary layers on graphite is not instantaneous but gradual. For 1-octyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide

([OMIM][TFSI]) under a strong negative potential of -2 V, the measured friction coefficient increases steadily from the initial stage to longer polarization times (20–60 min), even if the potential is held constant [24]. The friction–time profiles display two distinct regimes. Initially, the low-friction regime corresponds to a compact cation monolayer adsorbed on graphite, enabling relatively smooth sliding. However, with prolonged polarization, force–distance measurements reveal enhanced oscillatory layering, indicating the gradual formation of quasicrystalline cation-rich overstructures. This structural aging induces a transition to a higher-friction regime, characterized by increased interfacial roughness and ion–ion interactions that enhance energy dissipation [31, 56, 108, 112]. These two friction regimes reflect the time-dependent reorganization of the IL boundary layer and highlight the intrinsic coupling between interfacial structuring and nanofriction. These findings suggest that electrically controlled nanofriction is inherently time dependent, with structural aging strongly influencing frictional dissipation pathways.

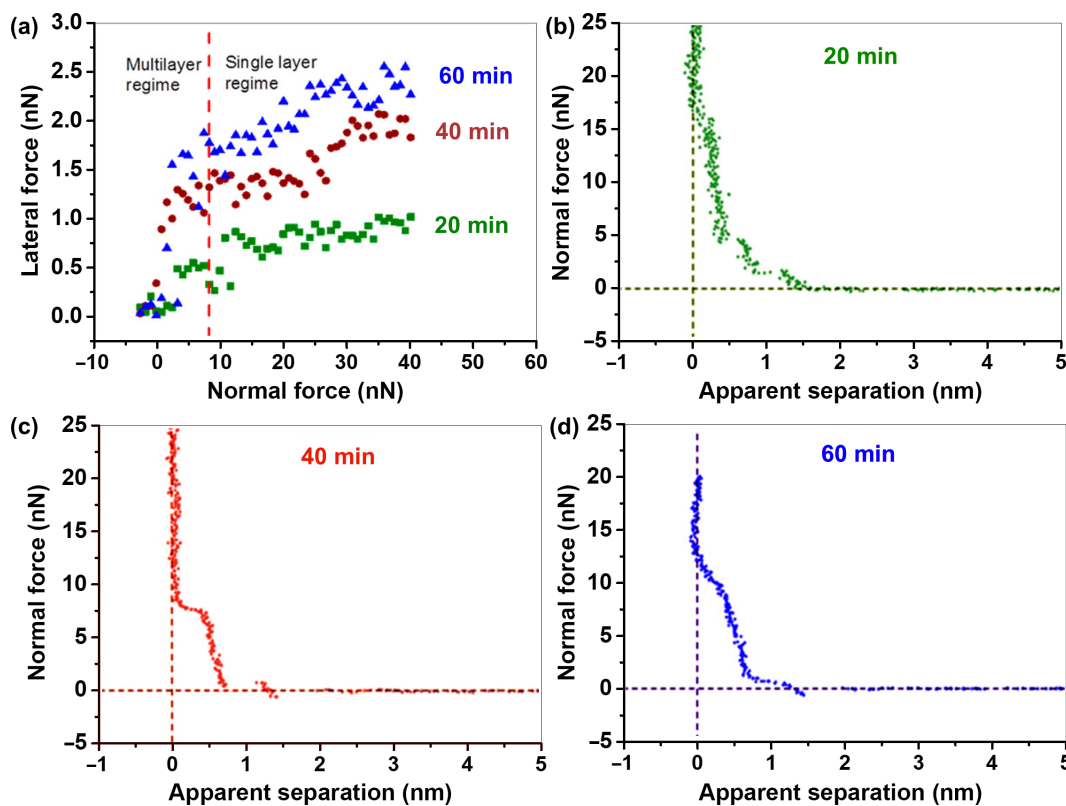


Fig. 15 Time-dependent frictional response of [OMIM][TFSI] on graphite at -2.0 V. (a) Lateral force vs. normal load curves measured after varying polarization times, i.e., 20, 40, and 60 min, showing the transition from single-layer to multilayer regimes. (b–d) The corresponding normal force vs. distance curves at 20, 40, and 60 min, respectively, revealing the progressive reorganization of interfacial layering with time. Reproduced with permission from Ref. [24], © American Chemical Society 2014.

5.3 Influence of the temperature on the friction of confined ILs

Temperature strongly influences the interfacial structure of ILs. In the experimentally studied range of 298–353 K, ILs can maintain well-defined interfacial layering, whereas increasing temperature generally broadens density profiles, weakens ion–ion correlations, and promotes more disordered orientations. This structural smearing weakens the stability of boundary layers [113]. Simulation studies on the graphene–[EMIM][BF₄] interface, performed over approximately 300–450 K, similarly showed that increasing temperature smears the layered ion distribution, reduces overscreening, and weakens ion–ion correlations [114].

Temperature modulates confined ILs primarily through viscosity reduction and dynamic rearrangements of the shear plane. The confined interface formed by graphene modifies the adjacent IL, reducing its viscosity, especially at low temperatures. This is attributed to noncovalent interactions such as π – π stacking and hydrogen bonding [115]. Manuel Martínez-Rubio et al. [116] demonstrated that near graphene sheets, the local structure and hydrogen bond network of the confined IL are disrupted, leading to a reduced viscosity, especially at low temperatures. However, this viscosity reduction is dependent on the concentration of graphene. At higher graphene concentrations, graphene–graphene and graphene–IL interactions dominate, leading to enhanced non-Newtonian behavior, an increased viscosity, and the emergence of yield stress. Therefore, the effect of graphene on the viscosity of the IL is nonmonotonic: it decreases the viscosity of ILs at low concentrations by disrupting the IL network, but it increases the viscosity of ILs at high concentrations due to the formation of a graphene network that impedes flow. Manuel Martínez-Rubio et al. [116] also found that the viscosity of both neat IL and graphene–IL dispersions decreases with increasing temperature, whereas the addition of graphene suppresses the thermal sensitivity of the dispersion, indicating enhanced structural stability over the investigated temperature range. Similarly, MD simulations by Ghoufi et al. [117] showed that increasing the temperature from 300 to 400 K enhances the diffusion of confined ILs, indicating faster ion mobility at elevated temperature. These results suggest that a higher temperature can facilitate slip by lowering viscosity and accelerating ion mobility but may also destabilize boundary layers [118–120]. Notably, for graphene-confined ILs, systematic nanoscale friction across temperature ranges is still lacking, representing a critical gap for high-temperature MEMS/NEMS applications.

In summary, the nanoscale friction of ILs confined between graphene surfaces is critically governed by three external factors,

i.e., humidity, electric field, and temperature, as schematically illustrated in Fig. 16. Each factor modulates friction through distinct interfacial mechanisms. Humidity typically increases friction because adsorbed water molecules compete with IL ions for adsorption on the graphene surface, disrupting interfacial layering and long-range order. The extent of this effect considerably depends on the hydrophobicity of the IL. High temperatures generally reduce the friction of graphene-confined ILs by lowering the viscosity of the confined ILs, but they simultaneously destabilize the well-defined boundary layers essential for effective lubrication, leading to a nonmonotonic response. In contrast, an applied electric field offers the most direct and reversible control, enabling the dynamic tuning of interfacial ion composition and orientation. This can induce dramatic transitions, even from high friction to superlubricity, by optimizing the structure and shear plane of the confined layers. Therefore, while humidity and temperature are environmental variables that must be managed to maintain low friction, the electric field is a powerful, active tool for obtaining precise and real-time control over friction in graphene–IL-based nanodevices.

6 Conclusions and outlook

The published literature indicates that nanofriction at graphene–IL interfaces is governed by interfacial ion structuring, which is regulated by the graphene surface properties, molecular architecture of ILs, and external stimuli. Building on Sections 3–5, this section synthesizes (i) established structure–property relationships, (ii) key challenges, and (iii) a roadmap toward programmable graphene–IL lubrication systems.

6.1 Established structure–property relationships

Several transferable relationships emerge across experiments and simulations: (1) Interfacial layering and ordering determine shear localization and frictional response: Ion layering and ordering at graphene interfaces determine whether shear localizes within the confined IL film or closer to the wall, which in turn controls stick–slip versus smooth sliding and the magnitude of nanofriction. (2) Graphene surface chemistry strengthens ion binding and reduces interfacial slip, increasing dissipation. For example, hydroxyl-/oxygen-rich graphene oxide walls strengthen specific interactions (including hydrogen bonding) and increase solid–liquid coupling, which increases nanofriction and decreases slip length, leading to higher dissipation under confinement. (3) Supporting substrates and graphene electronic/wetting states modulate interfacial ordering and slip. Even for single-layer graphene, the supporting substrate can modify the interfacial

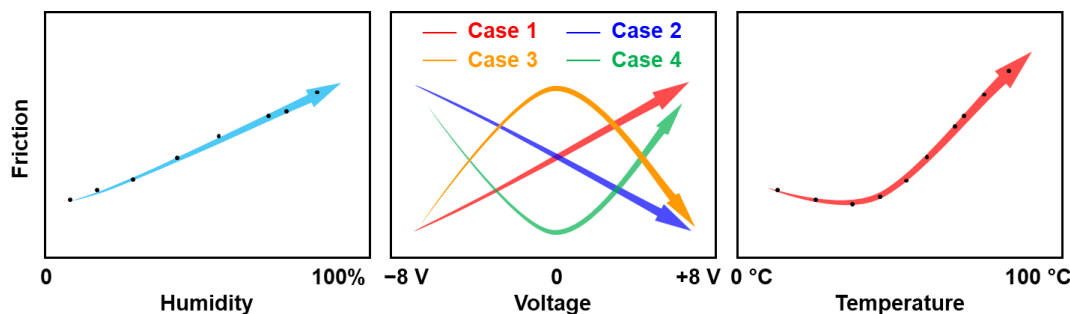


Fig. 16 Schematic summary of the influence of external stimuli on the friction of graphene-confined ILs. (a) Humidity: Friction increases with humidity as water molecules adsorb and disrupt the ordered ionic layers at the interface. (b) Electric field: Friction can be precisely and reversibly tuned using the applied potential; negative or positive biases enrich cations or anions at the interface, respectively, enabling transitions from high friction to superlubricity. (c) Temperature: Friction typically decreases with increasing temperature due to the reduced viscosity of the confined ILs, but it may increase at higher temperatures if interfacial layering is destabilized (nonmonotonic trend).

layering and slip of ILs, thereby changing dissipation at the interface, underscoring that graphene should be treated as a substrate-coupled interfacial state rather than a single universal surface. (4) Surface charge or applied potential reconfigures interfacial ion layers and shifts the effective shear plane, producing friction minima/maxima and switching. Charging or biasing graphene/graphite reorganizes the ion layers and shifts the effective shear plane, producing nonmonotonic friction–potential responses. Potential-controlled switching to ultralow friction has been demonstrated at IL–graphite interfaces, highlighting that friction can be reversibly tuned by electrically programmed interfacial states. (5) Ion architecture controls the balance between boundary-layer robustness and viscous dissipation. Longer alkyl chains and solvophobic tail aggregation can stabilize boundary ordering but can also increase viscous dissipation; therefore, the net friction outcome depends on the balance between structural robustness and mobility.

6.2 Key challenges and open questions

Despite substantial progress, several challenges currently limit quantitative prediction and device-relevant translation: (1) Poorly defined and heterogeneous graphene surface properties. Real graphene-based surfaces can exhibit spatially heterogeneous functional groups, defects, and contamination, which can dominate ion adsorption and local structuring. Robust descriptors for graphene surface properties and standardized reporting are needed to enable cross-study comparability. (2) Water/impurity sensitivity and interfacial competition. Trace water and other impurities can restructure the interfacial ion layers and compete with ions for adsorption sites, leading to strong variability in layering, slip, and friction. Water in the ion layers of ILs on graphene has been directly discussed in recent studies, highlighting the need for controlled humidity/impurity protocols and reporting. (3) Time-dependent evolution and structural aging under shear or bias. Interfacial restructuring can evolve over minutes to hours even under a constant potential, producing frictional aging and regime transitions. Time-dependent reorganization of IL boundary layers on graphite has been reported, implying that steady state cannot be assumed a priori in electrically controlled nanofriction. (4) Limited coupling between structure measurements and friction under identical operating conditions. A mechanistic description requires simultaneous or tightly correlated measurements of the interfacial structure and frictional response under the same shear, load, and bias conditions. Integrative experiment–simulation frameworks remain essential but are not yet routine in most studies. (5) Gaps between simulation and experiment. MD provides molecular detail but remains constrained by force-field fidelity and limited time/length scales, whereas experiments often involve surface heterogeneity and slow aging processes. Bridging these gaps requires systematic benchmarking across multiple observables and multiscale strategies. (6) Translation to device-relevant conditions. Nanoscale insights must be connected to realistic contacts and operating environments. Graphene-enabled MEMS/NEMS and related nanodevice contexts motivate the need for scaling laws and robust performance envelopes.

6.3 Roadmap toward programmable graphene–IL lubrication systems

A practical roadmap toward programmable graphene–IL lubrication systems can be expressed as a compact inputs–interfacial states–outputs framework. The key controllable

inputs include the graphene state (e.g., graphite, graphene, GO, rGO, supporting substrate, defect density, and surface charge), the molecular architecture of ILs (cation/anion identity, chain length, symmetry, and charge distribution), and operating conditions such as humidity, applied potential, and temperature. These inputs do not determine friction directly; instead, they first define the relevant interfacial states, including layering periodicity, in-plane ordering, ion enrichment and orientation in the boundary layer, overscreening/crowding, and the position of the effective shear plane. The measurable outputs emerge from these interfacial states, including the friction coefficient (μ), interfacial friction coefficient (λ), slip length (l_s), stick–slip behavior, and frictional regime (e.g., multilayer lubrication, single-layer lubrication, or ultralow-friction/superlubricity state). Thus, the most useful design rule is not to optimize a single variable in isolation but to control friction through interfacial structuring.

Several actionable directions can already be identified. For low-friction operation, the most promising combinations are atomically smooth and weakly pinning graphene states, ILs capable of forming ordered yet weakly dissipative boundary layers, and operating windows that stabilize a favorable shear plane. For adaptive or switchable lubrication, electrically tunable systems are particularly attractive because the applied potential can reversibly select the composition and orientation of the first adsorbed layers and thereby switch between distinct frictional states. Future experiments should therefore prioritize in situ structure–friction measurements under identical operating conditions, especially for systems in which humidity, potential, and surface chemistry compete simultaneously. Standardized reporting of substrate class, surface chemistry, roughness/curvature, water content, load window, sliding velocity, and probe geometry will also be essential for extracting transferable design rules across studies.

A further opportunity lies in AI (artificial intelligence) and data-driven prediction, but here, the target variables and data requirements must be defined explicitly. Rather than predicting the friction coefficient alone, future models should aim to predict both direct outputs (μ , λ , l_s , frictional regime, etc.) and intermediate interfacial descriptors (number of layers, layer thickness, ion enrichment, ion orientation, and shear-plane location). This is realistic in view of recent tribology studies showing that AI models can successfully predict nanoscale friction from structured experimental datasets [121, 122]. For graphene–IL systems, the minimum data schema should include substrate class, oxygen/defect level, conductivity or wettability descriptors, roughness/curvature, supporting substrate, IL structural descriptors, humidity, potential, temperature, tip radius, load range, sliding velocity, and the corresponding AFM/SFA/MD outputs. Building such standardized datasets will be a prerequisite for reliable, transferable, and mechanism-aware prediction of graphene–IL nanofriction.

In summary, graphene-confined ILs provide a tunable platform where friction can be engineered by programming interfacial ion structures. The mechanism-oriented framework synthesized in this review connects controllable design variables to interfacial states and frictional outputs, enabling more reproducible interpretation and more targeted selection of graphene surface states, IL ions.

Appendix

A Abbreviations

See Table A1.

Table A1 Abbreviations

AFM	Atomic force microscopy
AI	Artificial intelligence
F_F	Friction force
F_N	Normal force
GO	Graphene oxide
Gr	Graphene
IL	Ionic liquid
l_s	Slip length
MD	Molecular dynamics
MEMS/NE	Micro- and nanoelectromechanical systems
MS	
OCP	Open circuit potential
SFA	Surface force apparatus
v	Slip velocity
η	Fluid viscosity
λ	Interfacial friction coefficient
μ	Nanoscale friction coefficient (introduced in the text as μ_N to distinguish it from μ_M)
μ_M	Macroscopic friction coefficient
τ	Tangential stress
[AOT] ⁻	Diocetyl sulfosuccinate
[BF ₄] ⁻	Tetrafluoroborate
[BMIM] ⁺	1-Butyl-3-methylimidazolium
[C2C1Im] ⁺	1-Ethyl-3-methylimidazolium
[CnMIM] ⁺	1-Alkyl-3-methylimidazolium
[EMIM] ⁺	1-Ethyl-3-methylimidazolium
[EtSO ₄] ⁻	Ethyl sulfate
[FAP] ⁻	Tris(pentafluoroethyl)trifluorophosphate
[HMIIm] ⁺	1-Hexyl-3-methylimidazolium
[N1114] ⁺	Butyltrimethylammonium
[N1123] ⁺	Ethyltrimethylpropylammonium
[NTf ₂] ⁻ / [TFSI] ⁻	Bis(trifluoromethylsulfonyl)imide
[OMIM] ⁺	1-Octyl-3-methylimidazolium
PAN	Propyl ammonium nitrate
[PF ₆] ⁻	Hexafluorophosphate

(Continued)

AFM	Atomic force microscopy
[P ₄₄₄₁] ⁺	Triethyl(butyl)phosphonium
[P ₆₆₆₁₄] ⁺	Trihexyl(tetradecyl)phosphonium
[pyr14] ⁺	1-Butyl-1-methylpyrrolidinium

B Explanation

(1) Commensurability: The geometric matching between the periodic atomic lattices of two contacting surfaces (e.g., the AFM tip and the ordered ion layer). High commensurability often leads to stronger interlocking and higher friction.

(2) Frictional dissipation: The conversion of mechanical energy (from sliding) into heat at the interface.

(3) Overscreening: A phenomenon in ILs where a surface charge is overcompensated by the first layer of counterions, leading to alternating layers of excess charge.

(4) Quantized friction: Quantized friction refers to a phenomenon in nanoscale tribology where the frictional force or torque exhibits discrete, step-like transitions as a function of normal load or confinement when two solid surfaces are separated by a molecularly thin film (e.g., ILs, simple lubricants). This behavior is often interpreted as a mechanical signature of the sequential expulsion of molecular layers or structural transitions within the highly confined film, arising from collective interlayer interactions and energy barriers rather than quantum mechanical effects.

(5) Shear plane: The shear plane is a physical model concept in fluid dynamics used to describe interfacial slip. When a fluid undergoes shear flow near a solid boundary, the no-slip condition may breakdown at the nanoscale. The shear plane represents the location where the relative velocity between the fluid and the solid wall is defined, and its position determines the effective “slip length”, a key parameter for calculating friction forces in thin films.

(6) Solvophobicity: Solvophobicity describes the thermodynamic tendency of a substance (e.g., an ion, molecule, or functional group) to minimize its contact with a specific solvent environment, such as an IL. This is the generalized concept of hydrophobicity applied to nonaqueous solvents. In ILs, solvophobic interactions, often driven by the segregation of nonpolar moieties from charged or polar networks, are crucial in driving self-assembly, nanostructuring, and the formation of distinct domains both in the bulk and at interfaces, significantly affecting lubrication properties.

(7) Stick-slip dynamics: A frictional instability characterized by periodic cycles of sticking (energy accumulation) and sudden slipping (energy release).

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Declaration of competing interest

The authors have no competing interests to declare that are

relevant to the content of this article. The author Braham Prakash is the Editorial Board Member of this journal.

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