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Prediction and Experimental Verification of Electrolyte Solvation Structure from an OMol25-Trained Interatomic Potential

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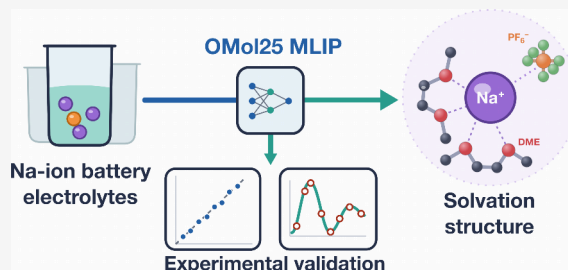


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ABSTRACT: A molecular-level understanding of electrolyte solvation structure and ion–ion correlations is critical to developing next-generation battery chemistries. Atomistic simulation capabilities with sufficient accuracy, speed, and transferability to deliver reliable structural insights while avoiding arduous system-specific reparameterization are thus highly desirable. Machine learning interatomic potentials (MLIPs) trained on large, chemically diverse data sets are revolutionizing computational chemistry, enabling molecular dynamics simulations of battery electrolytes with near-DFT accuracy over 10,000× faster than DFT. While previous MLIP training data sets with suitable elemental coverage for electrolytes have been based on inorganic materials, the Open Molecules 2025 (OMol25) data set provides large-scale molecular DFT MLIP training data with broad elemental coverage and specifically samples tens of millions of electrolyte configurations. Here, we integrate computational modeling with experimental validation to systematically assess the ability of large-scale MLIPs pretrained on materials data or on OMol25 to accurately resolve nanoscale structural organization and ion-solvation characteristics in Na-ion battery electrolytes across diverse physicochemical conditions and compositional regimes. We find that the OMol25-trained Universal Model of Atoms (UMA-OMol) predicts experimentally measured densities and X-ray structure factors in substantially better agreement compared to state-of-the-art models trained only on inorganic materials data. Using UMA-OMol, we further analyze systematic trends in solvation structure as a function of cation identity, anion chemistry, salt concentration, and solvent topology. We observe that increasing system temperature amplifies the heterogeneity within the solvation environment, perturbing cation–solvent interactions and promoting the formation of contact ion pairs (CIPs). Moreover, subtle variations in the solvent topology of glyme-based electrolytes cause pronounced changes in ion correlations and solvation structure. The experimental agreement and microscopic insights shown here position OMol25-trained MLIPs as a practical route to predictive, high-throughput electrolyte simulations beyond the limits of classical force fields and direct DFT molecular dynamics, serving as a powerful tool for accelerating the design of next-generation Na-ion battery electrolytes and beyond.



Accurately capturing microscopic solvation structure is crucial to predicting macroscopic properties in complex liquid mixtures like battery electrolytes. Predictive atomistic simulations of electrolyte solvation structure could dramatically accelerate electrolyte discovery and additive engineering via high-throughput *in-silico* screening or by elucidating mechanistic insight that enables rational chemical design.^{1–4}

Over the past several decades, the pursuit of chemically accurate molecular dynamics (MD) for complex, multi-component systems has been constrained by a fundamental tradeoff between accuracy and scale.⁵ *Ab initio* MD captures the delicate interplay of covalent and noncovalent interactions with quantum-mechanical rigor, but its prohibitive computational cost limits accessible length and time scales to regimes far from those relevant to mesoscale structure and transport.⁶ Classical and polarizable force fields, while computationally efficient, demand labor-intensive parameterization and are often anchored to a narrow domain of validity, relying on empirical adjustments that cannot fully capture the many-body

effects, electronic polarization, and subtle free-energy landscapes of chemically heterogeneous environments.⁷

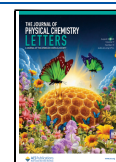
Recent advances in machine learning interatomic potentials (MLIPs) trained on density functional theory (DFT) data offer a compelling path forward: near-*ab initio* accuracy at a fraction of the computational cost, with the flexibility to study both covalent and noncovalent structure in response to the local chemical environment.^{8,9} Yet, as with any ML model, their fidelity is ultimately bounded by the size, chemical diversity, and consistency of the training data. While MLIPs trained on small, system-specific data sets can yield novel insight into

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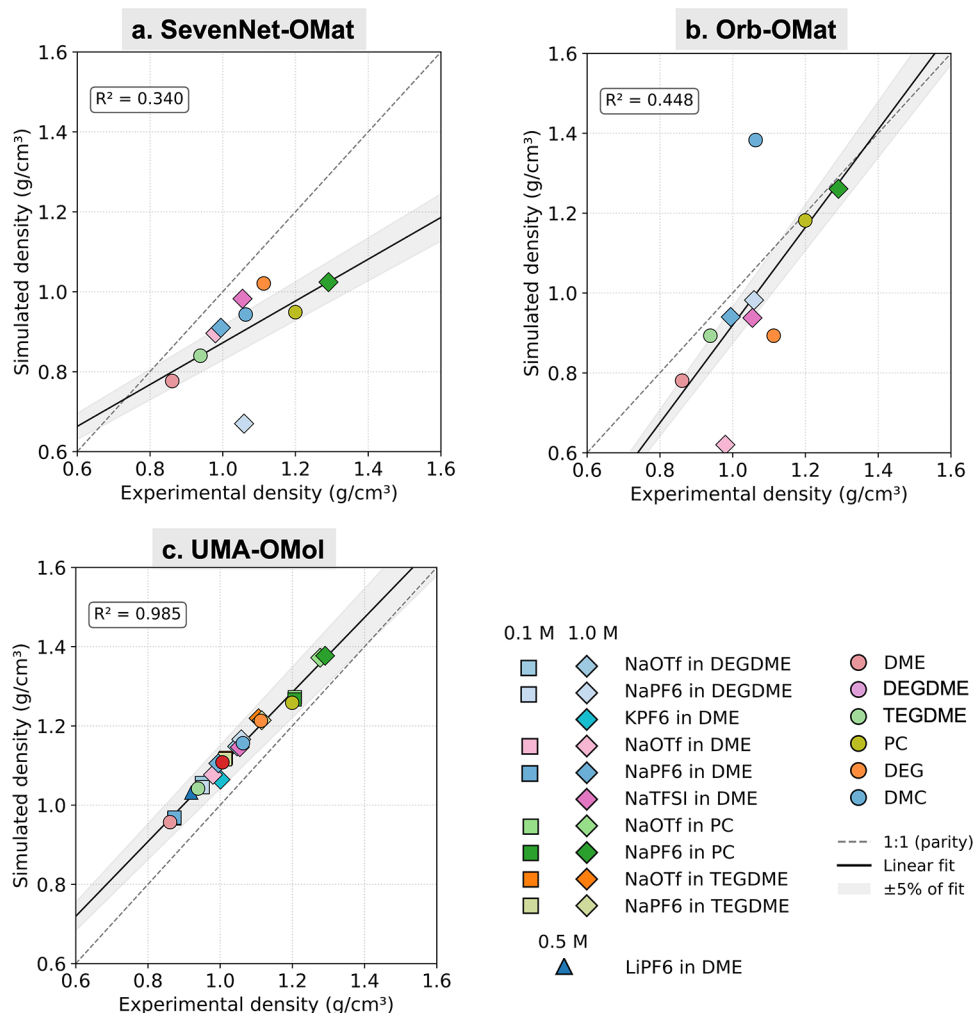


Figure 1. Pretrained MLIP predicted liquid density comparisons for different organic and sodium electrolyte systems. Experimental densities are compared with simulated densities from two inorganic materials-trained models [(a) SevenNet-OMat, (b) Orb-OMat] and one OMol25-trained model [(c) UMA-OMol] across battery electrolytes that vary in salt type, salt concentration, and solvent type. Here, OTf, TFSI, and PF₆ correspond to triflate, bis(trifluoromethanesulfonyl)imide, and hexafluorophosphate anions respectively. DME, DEGDME, PC, TEGDME, DEG, and DMC correspond to dimethoxyethane, diethylene glycol dimethyl ether (diglyme), propylene carbonate, tetraethylene glycol dimethyl ether, diethylene glycol, and dimethyl carbonate, respectively. The dashed line represents the ideal parity relation between simulation and experiment. The solid line denotes the least-squares linear regression fit to the simulated versus experimental densities. The shaded region indicates a $\pm 5\%$ envelope around the fitted regression line, highlighting the acceptable deviation window relative to the model trend.

individual systems of interest,^{10–12} the time and resources required to generate DFT training data and train models is often onerous, and the resulting trained models are not transferable. In contrast, MLIPs pretrained on very large, diverse data sets seek to provide sufficiently accurate predictions across chemical space to obviate the need for system-specific data generation and model training.^{13,14} However, widely used large-scale molecular DFT data sets like SPICE/2^{15,16} and AIMNet2¹⁷ have little to no coverage of critical metals for battery electrolytes (e.g., Li, Na, Zn) while electrolyte-specific works like BAMBOO¹⁸ and Wang et al.¹⁹ focus almost exclusively on Li-ion systems. Thus, the only large-scale pretrained MLIPs with sufficient elemental coverage for diverse liquid electrolytes have paradoxically been those trained on crystalline material planewave DFT training data from the Materials Project²⁰ (MPtrj),¹⁴ Alexandria²¹ and/or Open Materials 2024 (OMat24).^{22,23} However, the recently released Open Molecules 2025 (OMol25) now provides a large-scale, high-accuracy molecular DFT data set for training

MLIPs with extensive elemental coverage and directly targets electrolytes as one of its primary areas of focus.²⁴

In this study, we evaluate the predictive capability of large-scale, pretrained MLIPs for describing the macroscopic ensemble-averaged properties and the microscopic interatomic interaction patterns that govern the structure and dynamics of next-generation Na-ion battery electrolytes. While Li-ion electrolytes have dominated consumer electronics for decades, Na-ion batteries present major advantages in cost and material availability, motivating computational discovery.^{25,26} We investigate pretrained MLIP performance for simulating NaPF₆ (sodium hexafluorophosphate), NaOTf (sodium triflate), or NaTFSI (sodium bis(trifluoromethanesulfonyl)imide) in ether and carbonate solvent matrices and under different system conditions such as temperature or concentration. Further, only large-scale MLIPs pretrained on either inorganic materials DFT data or on a sufficiently diverse molecular data set like OMol25 are viable for simulating such systems. Therefore, Orb-OMat and SevenNet-OMat were

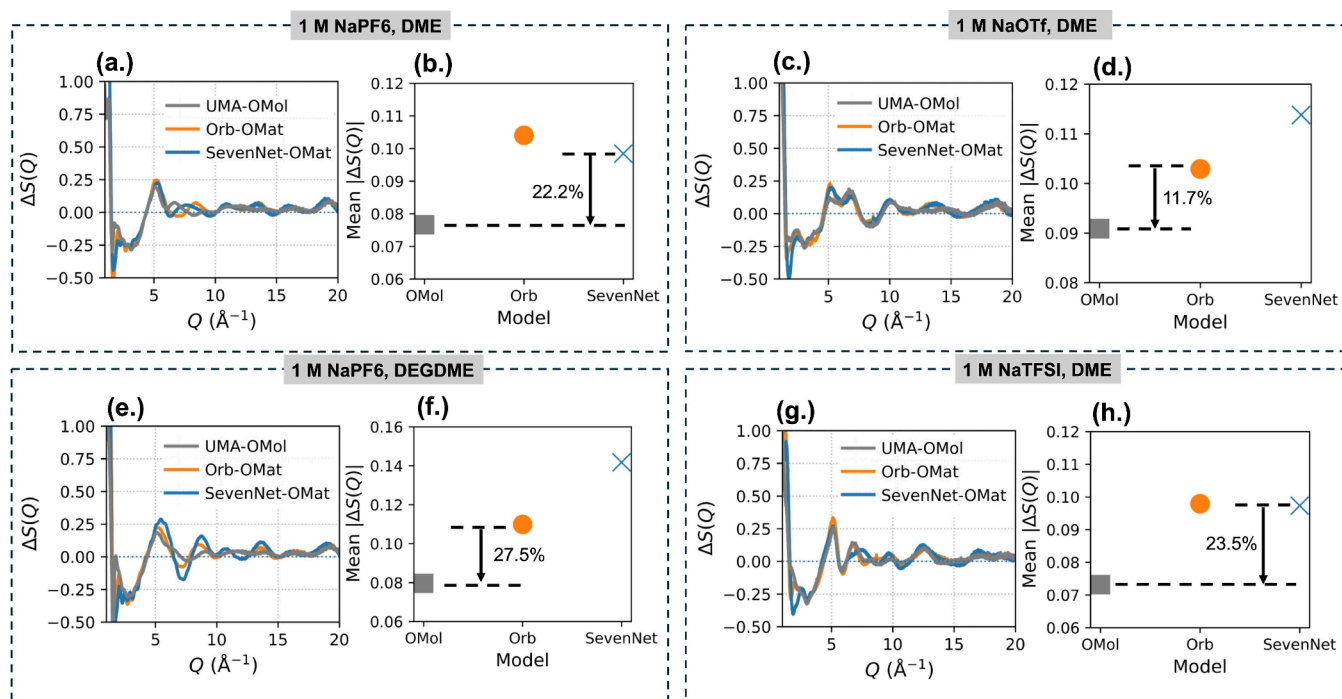


Figure 2. Residual X-ray structure factors and mean deviations for 1 M sodium salt glyme electrolytes. Panels (a), (c), (e), and (g) show the difference between simulated and experimental structure factors, i.e., the residual $\Delta S(Q) = S_{\text{sim}}(Q) - S_{\text{exp}}(Q)$ for NaPF₆ in dimethoxyethane (DME), NaOTf in DME, NaPF₆ in diglyme (DEGDME), and NaTFSI in DME, respectively, comparing UMA-OMol (gray), Orb-OMat (orange), and SevenNet-OMat (blue) over $Q = 1\text{--}20 \text{ \AA}^{-1}$. Panels (b), (d), (f), and (h) present the corresponding mean absolute deviations ($|\Delta S(Q)|$), showing that UMA-OMol systematically yields the smallest residuals, with Orb-OMat often intermediate and SevenNet-OMat often largest.

chosen as large, robust pretrained MLIPs whose training data cover the elements required for Na-ion electrolytes (Li, Na, K, P, S, F, etc.), which have been claimed to transfer beyond their original domain and can be benchmarked alongside UMA-OMol against the same experimental observables.²⁴

We benchmark three large-scale, pretrained MLIPs, two trained on the inorganic materials OMat24 data set—Orb-OMat²⁷ and SevenNet-OMat²⁸—and one trained on OMol25—UMA-OMol^{29,30}—against experimental Na-ion electrolyte density and X-ray structure factor measurements, establishing that UMA-OMol most accurately reproduces these macroscopic properties. We then examine the performance of UMA-OMol across varying battery operating temperatures, where Na-ion systems are compared to well-understood Li-ion systems as a baseline, followed by a systematic study of anion identity to capture the correct hierarchy of anion–cation interaction strengths (via coordination number) and their influence on solvation structure. We further investigate the concentration dependence of NaPF₆ electrolytes, quantifying the shift from free-ion populations to contact and solvent-separated ion pairs and linking these structural changes to transport-relevant trends observed in experiment. This is followed by an investigation of solvent effects³¹ on ion pairing and aggregation across six different solvent candidates for Na-ion batteries, and subsequent analysis of atomic-scale structuring at the graphite interface.

Our results show that the OMol25-trained MLIP UMA-OMol is an efficient surrogate for first-principles simulations of battery electrolytes and offers broad promise for predictive modeling of complex electrochemical systems, providing chemically transferable, *ab initio*-level predictions of nanoscale electrolyte behavior at orders of magnitude reduced computational cost versus DFT-based molecular dynamics. Further,

UMA-OMol shows performance superior to models trained on inorganic material DFT data sets that did not include liquid-phase, electrolyte-specific structural sampling. Overall, we view OMol25-trained models as powerful first-line tools for modeling battery electrolyte systems, to be complemented as needed by targeted electronic structure calculations and experimental data as model architectures continue to improve.

a. Inorganic Materials vs OMol25 Pretrained MLIPs. Among the macroscopic thermodynamic observables of an electrolyte, bulk density is particularly sensitive to systematic errors in the absolute energy scale of the potential; together with the experimental X-ray structure factor $S(Q)$, which probes local and intermediate-range pair correlations, density and $S(Q)$ provide complementary, nonredundant validation targets.^{32,33}

Accurately capturing experimental densities is essential for meaningful comparison and reliable prediction of material behavior under realistic conditions. Accordingly, we measured the densities of pure solvents and Na-ion battery electrolytes across different salt types, concentrations, and solvent environments, as well as one Li-ion and one K-ion system, and compared them with densities predicted by NPT molecular dynamics driven by UMA-OMol, Orb-OMat, and SevenNet-OMat in Figure 1, and the respective numerical values are provided in Table S1. Each system was simulated for 0.5 ns, with the final 0.2 ns used for analysis. Since each system was pre-equilibrated under a well-validated OPLS-AA force field^{34–37} (Figure S1) before propagation under the MLIP, all reported observables reach a stationary regime within the first 100 ps of the MLIP trajectory (Figure S2). Simulations performed using the SevenNet-OMat and Orb-OMat large-scale MLIPs trained on inorganic materials data sets (MPtrj,¹⁴ Alexandria,²¹ and OMat24²²) exhibit substantial deviations from experimental densities (see Figure 1a and b). For

SevenNet-OMat, the simulated densities show systematic underestimation and significant scatter ($R^2 \approx 0.34$), while Orb-OMat exhibits only slightly better agreement with experiment ($R^2 \approx 0.45$). Further, for both models we observed several MD simulations become unstable, causing an unphysical breaking of the simulation box and precluding the trajectory from ever reaching equilibrium. We hypothesize that these deficiencies stem from the absence of liquid-phase configurations and solvation environments in the inorganic training data sets, limiting the model's ability to capture subtle intermolecular and ion–solvent correlations.

In contrast, the parity plot between experimental and UMA-OMol predicted densities shows excellent agreement across linear and cyclic ethers, carbonates, and their 0.1 and 1.0 M sodium salt solutions (NaPF₆, NaOTf, NaTFSI, etc.), yielding an overall coefficient of determination $R^2 = 0.98$. We do observe a systematic shift in the UMA-OMol parity toward slightly higher values across all electrolyte systems. On average, the simulated densities are about 8.5% higher than experiment; we speculate that this overestimation is due to VV10 overbinding and the absence of long-range interactions, though a deeper evaluation is beyond the scope of this work.^{38,39,40,41}

We observed a similar behavior for density variations with temperature, as shown in Figure S3, which compares experimental and simulated densities for 0.5 M NaPF₆ and LiPF₆ electrolytes in DME. Despite the systematic shift, both systems exhibit the expected linear thermal expansion, with density decreasing as temperature increases, and our simulations reproduce this trend with a high fidelity. The simulated densities are about 10% higher than experiment for both electrolytes at different temperatures, reflecting a modest overestimation of solvent packing and ion-associated partial molar volumes. The narrow confidence band around the parity line further demonstrates consistent transferability across chemically distinct electrolytes, highlighting that UMA-OMol accurately predicts both pure solvent systems and overall neutral systems containing dissolved ions.

The pronounced improvement from inorganic material-trained models to UMA-OMol underscores the importance of explicitly incorporating liquid-state chemistries in the MLIP training data set and the higher fidelity density functional accessible in a molecular context.

We further assessed the molecular structuring predicted by UMA-OMol and other models against experimental X-ray structure factors $S(Q)$ for a series of 1 M sodium salt electrolytes (Figures S4 and S5). The data set comprises NaPF₆, NaOTf, and NaTFSI dissolved in dimethoxyethane (DME), diglyme (DEGDME), and tetraglyme (TEGDME), with each panel reporting one salt solvent combination. For reference, the corresponding optimized-OPLS-AA classical-force-field^{34–37} $S(Q)$ for the NaPF₆/DME, NaPF₆/diglyme, and NaOTf/DME systems are provided in Figure S1. To quantitatively measure model accuracy, we evaluated residual structure factors $\Delta S(Q) = S_{\text{sim}}(Q) - S_{\text{exp}}(Q)$ over $Q = 1\text{--}20 \text{ \AA}^{-1}$ (Figure 2). UMA-OMol captures not only the dominant features of $S(Q)$ but also the high- Q oscillations that arise from bond-stretching, bond-angle fluctuations, and torsional motions of the ether chains, demonstrating an accurate short-range potential energy surface and realistic molecular flexibility. This fidelity is reflected in the mean absolute deviation $\langle |\Delta S(Q)| \rangle$, which is smallest for UMA-OMol and systematically larger for Orb-OMat and SevenNet-OMat across all electrolytes. Only four of the six systems are shown in Figure 2

because Orb-OMat and SevenNet-OMat MD simulations of NaOTf in DEGDME and NaPF₆ in TEGDME were consistently unstable.

Although the residual $\Delta S(Q)$ for the inorganic-materials-trained MLIPs may appear roughly comparable to that of UMA-OMol on a single panel, $\langle |\Delta S(Q)| \rangle$ is systematically 10–30% larger, and several of those simulations were outright unstable; the surviving residuals are therefore conditional on a stable subset. Splitting $\langle |\Delta S(Q)| \rangle$ into low- Q ($<3 \text{ \AA}^{-1}$) and high- Q ($3\text{--}20 \text{ \AA}^{-1}$) bands further resolves this asymmetry (Figure S6). For UMA-OMol, the residual is concentrated in the low- Q band, consistent with the 6 Å cutoff and the ~8–10% density overestimation; the high- Q residual is small and approximately system-independent, confirming that local solvation structure is well reproduced.

The reduced residuals and well-resolved oscillatory features at high- Q values indicate that UMA-OMol treats bonded interactions and short-range repulsion more accurately, which in turn leads to enhanced robustness in long molecular dynamics trajectories with minimal bond distortion and few unphysical structural instabilities.

Density and X-ray structure factor results show that UMA-OMol reproduces the structural properties of battery electrolytes more robustly than models trained on inorganic material data. We now proceed to employ UMA-OMol to interrogate molecular structure across different physicochemical conditions and variations in anion type and solvent topology.

b. Anion Effects. We compare Na⁺ association with three common battery anions in glyme electrolytes using radial distribution functions, $g(r)$, and running coordination numbers, $n(r)$, as these interactions directly control electrolyte speciation in Na-ion batteries. Figure 3a shows Na⁺ correlations with TFSI[−], PF₆[−], and OTf[−], referenced to their central N, P, and S atoms, respectively, in 1 M DME electrolytes. The first solvation shell exhibits a clear hierarchy in ion pairing strength, with OTf[−] displaying a sharp and intense peak at $r \approx 3.4 \text{ \AA}$ ($g_{\text{max}} \approx 45$), PF₆[−] showing a broader and weaker maximum at a similar distance ($g_{\text{max}} \approx 11$), and TFSI[−] exhibiting only weak structuring ($g_{\text{max}} \approx 4$). Integration of the corresponding coordination numbers to 5 Å (Figure 3b) yields $n = 3.0 \pm 0.1$ for OTf[−], 1.1 ± 0.1 for PF₆[−], and 0.3 ± 0.05 for TFSI[−], indicating that Na⁺ forms substantially more frequent and experimentally observed stable ion paired configurations with OTf[−] than with the more weakly coordinating anions, as expected from previous work.⁴²

Atom-resolved binding analysis further supports this hierarchy. We computed Na–X radial distribution functions and coordination numbers for the dominant coordinating sites in each anion (X = O in TFSI[−], F in PF₆[−], and O in OTf[−]; Figure S7). Across electrolytes, the contact ion pairing signature is most pronounced for Na⁺ binding to O in OTf[−], which exhibits a very sharp first-shell peak centered near ~2.3 Å, far exceeding the corresponding first-shell intensities for PF₆[−] and TFSI[−], consistent with substantially stronger and more frequent direct Na⁺–anion interactions in NaOTf relative to the more weakly coordinating anions.

Interestingly, UMA-OMol shows that solvent identity can significantly control these ion interactions. In particular, for the strongly coordinating OTf[−] anion, a substantially enhanced Na–OTf first shell peak and a steeper short-range growth in $n(r)$ in DME than in DEGDME (Figure 3c) demonstrates that the shorter glyme promotes stronger effective Na⁺–OTf association, whereas the longer glyme more effectively

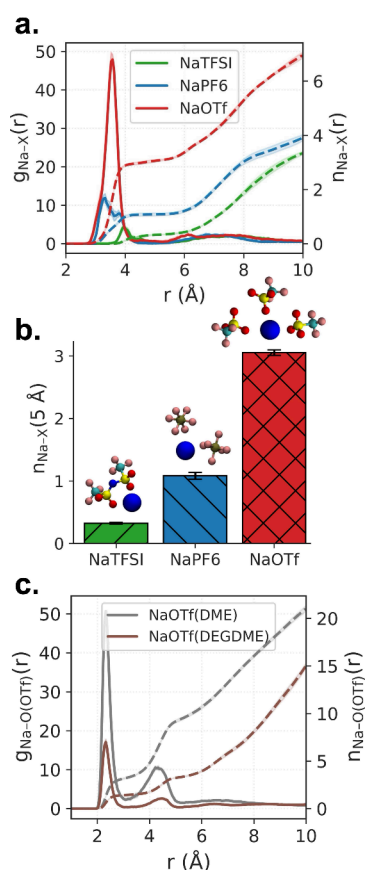


Figure 3. Anion-dependent Na^+ coordination in DME from UMA-OMol simulations. (a) Radial distribution functions $g_{\text{Na-X}}(r)$ (solid lines, left axis) and running coordination numbers (dashed lines, right axis) $n_{\text{Na-X}}(r)$ for bis(trifluoromethanesulfonyl)imide (TFSI^-), hexafluorophosphate (PF_6^-), and triflate (OTf^-), computed relative to N, P and S atoms of TFSI^- , PF_6^- and OTf^- respectively. (b) Average coordination numbers $n_{\text{Na-X}}(5 \text{ \AA})$ in primary cation coordination shell highlighting the hierarchy of ion-pairing strength across anions. (c) Radial distribution functions $g_{\text{Na-O(OTf)}}(r)$ and corresponding running coordination numbers $n_{\text{Na-O(OTf)}}(r)$ for 1 M NaOTf in DME and diglyme (DEGDME) media, demonstrating that solvent identity can modulate ion interaction strength.

competes for Na^+ coordination and suppresses direct anion binding.^{43–46} The observed solvent–anion coupling effect, where solvent topology regulates ion-pairing even for strongly binding anions such as OTf^- , may enable strategic modulation of Na^+ solvation structure and ion-association equilibria in Na -ion battery electrolytes.

c. Temperature Effects. Temperature profoundly reorganizes the solvation landscape in both Li^+ and Na^+ liquid electrolytes, but with element-specific signatures reflecting their distinct ionic radii and binding energetics.^{47,48} To probe UMA-OMol's capacity to capture temperature-dependent changes in solvation structure, we studied 0.5 M NaPF_6 and LiPF_6 in DME at 253, 300, and 353 K. At 253 K, both electrolytes exhibit highly ordered cation solvation, evidenced by sharp first-shell peaks in the $g_{\text{Na-O(DME)}}(r)$ and $g_{\text{Li-O(DME)}}(r)$ functions at $\sim 2.3 \text{ \AA}$ for Na^+ and $\sim 2.0 \text{ \AA}$ for Li^+ (Figures S8a and S9a), 100% free ions observed in fractional analysis (Figure 4a, Figure S10a), and the complete absence of a first-shell peak in $g_{\text{Na-P(PF}_6\text{)}}(r)$ and $g_{\text{Li-P(PF}_6\text{)}}(r)$ functions (Figure 4b, Figure S10b), which trace cation–anion proximity. These data indicate tightly bound first-shell solvent coordination due to reduced thermal fluctuations at low-temperature conditions, which together render CIP formation a rare-event process on the presently sampled time scales. We anticipate that extending the simulation time or employing enhanced sampling strategies would allow occasional $\text{Na}^+\text{--PF}_6^-$ contact configurations to emerge, thereby more fully resolving the underlying free energy landscape governing ion association.

As temperature rises, the ion–solvent first-shell peaks attenuate and broaden, and average coordination decreases, signaling enhanced solvent exchange and weakened cation–solvent binding (Figures S8 and S9). Concurrently, the Na-P radial distribution functions reveal an emerging first-shell contact peak near $3.0\text{--}3.3 \text{ \AA}$, with its intensity nearly doubling between 300 and 353 K (Figure 4b). The corresponding coordination integral $n_{\text{Na-P}}(r)$ confirms a monotonic rise (Figure 4c), and fraction analysis quantifies a significant decrease in the fraction of free ion (Figure 4a) and an increase in ion-paired species—from negligible at 253 K to ≈ 0.27 at 353 K (Figure S11).^{49,50} Similar trends were exhibited for LiPF_6 (Figure S10), with one notable difference—the Li-P radial distribution function shows a distinct double first-shell

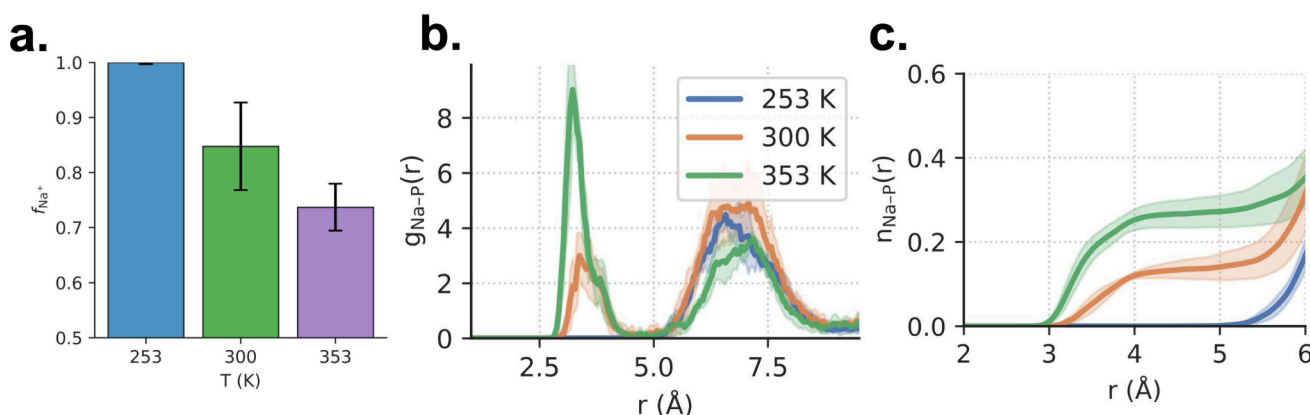


Figure 4. Temperature dependence of NaPF_6 (0.5 M) ion association in DME captured by UMA-OMol simulations. (a) Fraction of unpaired Na^+ ions at different temperatures. (b) Na-P (PF_6^-) radial distribution functions and (c) corresponding running coordination numbers at different temperatures. Error bars are obtained using the block-averaging methodology. The analysis reveals that all Na^+ ions are fully solvated at 253 K, while at higher temperatures the cation solvation shell weakens and allows for increasing ion pair formation.

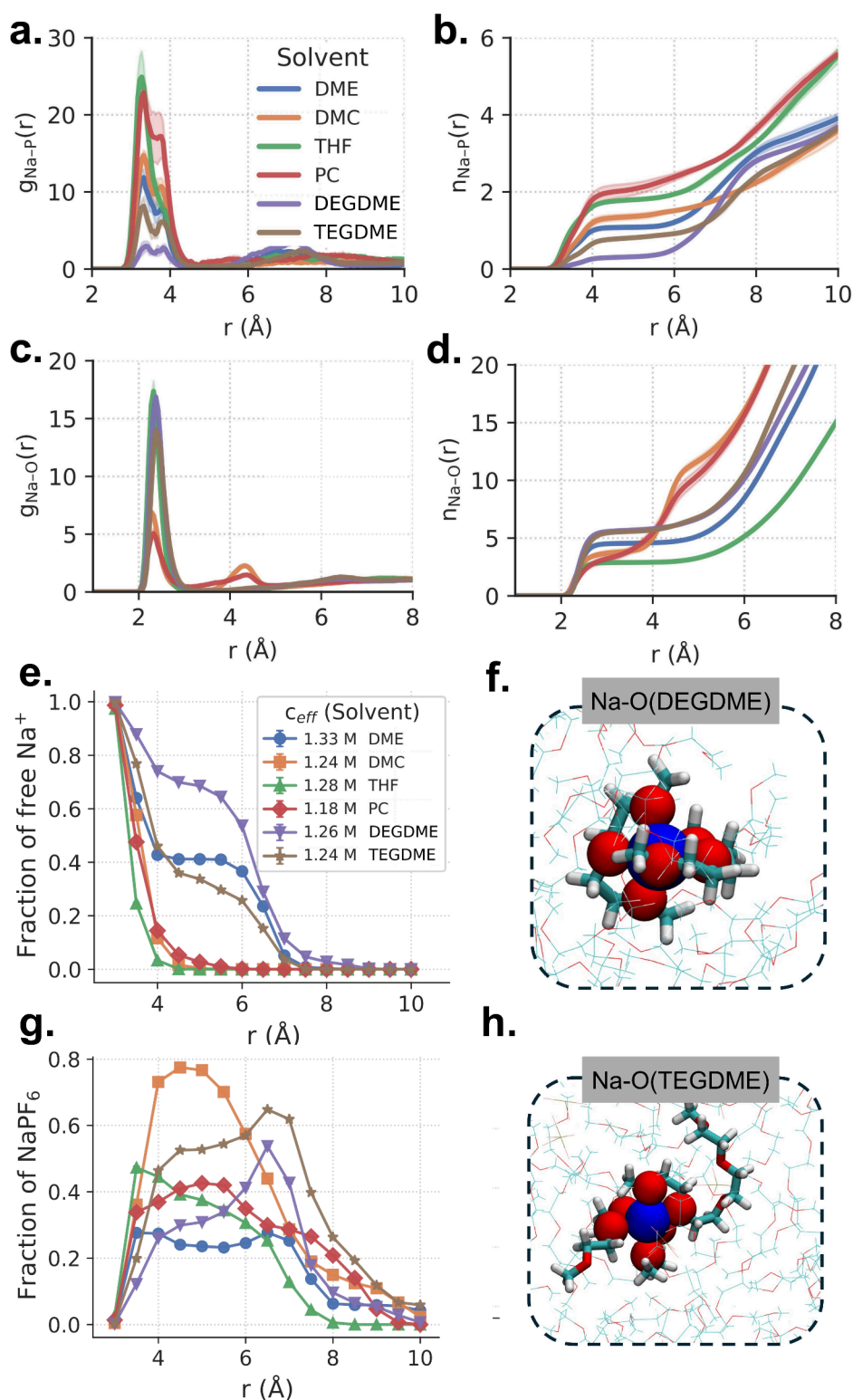


Figure 5. Solvent effects on NaPF_6 association in 1 M sodium electrolytes from UMA-OMol simulations: (a) Na-P radial distribution functions $g_{\text{Na-P}}(r)$ for dimethoxyethane (DME), dimethyl-carbonate (DMC), tetrahydrofuran (THF), propylene carbonate (PC), diglyme (DEGDME), and tetraethylene glycol dimethyl ether (TEGDME). (b) Corresponding Na-P running coordination numbers $n_{\text{Na-P}}(r)$. (c) Na-O radial distribution functions $g_{\text{Na-O}}(r)$ for the same solvent series. (d) Na-O running coordination numbers $n_{\text{Na-O}}(r)$. The variation in peak positions and coordination behavior highlights the strong solvent dependence of $\text{Na}^+ - \text{PF}_6^-$ association and Na^+ solvation structure across linear glymes, cyclic ethers, and carbonate solvents. Comparison of DEGDME and TEGDME solvation of Na^+ in 1 M NaPF_6 electrolytes: (e) Fraction of free Na^+ ions versus radial cutoff distance. (g) Fraction of Na^+ forming CIPs. (f,h) Representative snapshots showing compact chelation by DEGDME excluding PF_6^- versus open-chain TEGDME conformation enabling overcoordination and PF_6^- insertion into the first solvation shell.

peak, indicative of both monodentate and bidentate coordination motifs, as expected for Li^+ .⁵¹

A per-cation, time-resolved analysis of the Na^+ coordination environment (Figure S12) shows that individual Na^+ ions fluctuate on short time scales between solvated, contact, and solvent-separated configurations, but the population fractions remain stationary over the analysis window and are consistent with the ensemble average values.

Overall, the thermal liberation of DME from the Na^+ and Li^+ solvation shells weakens the cation cage, enhances anion access, and shifts speciation toward contact and solvent-shared ion pairs, yielding an entropy-driven reduction in the population of free Na^+ or Li^+ carriers despite decreasing viscosity.⁵⁰ These results show that UMA-OMol successfully captures the interplay between temperature-dependent enthalpic stabilization and entropic gain.

d. Concentration Effects. We next examined the correlations between electrolyte concentration and the propensity of ions to remain free or to form contact ion pairs, solvent separated pairs, and larger aggregates, each of which directly influences the effective transport of charge carriers especially under an applied potential.⁵² Figure S13 summarizes the effect of NaPF_6 concentration in DME on the population of free Na^+ ions—defined here as cations without any PF_6^- in their primary coordination shell—and on the fraction of Na^+ coordinated to exactly one PF_6^- anion, between radial distances 3 and 10 Å. We observe an increased probability of Na^+ – PF_6^- CIPs followed by an expected decrease in the fraction of free Na^+ ions with increasing electrolyte concentration. Further details are provided in the Supporting Information.

e. Solvent Effects. Effective charge transport is strongly governed by the interactions between the charge carriers and their surrounding solvent environment.^{53–55} Therefore, we next investigated ion and solvent correlations across different solvent systems to quantify how variations in local solvation directly influence charge transport behavior.⁵⁶ Radial distribution functions and running coordination numbers provided in Figure 5a–d, map how solvent identity modulates NaPF_6 association in different electrolyte solutions. In the Na–P radial distributions (Figure 5a), the first-shell peak height decreases in the order $\text{PC} \approx \text{THF} > \text{DMC} > \text{DME} > \text{TEGDME} > \text{DEGDME}$. Integration of $g(r)$ to 10 Å (Figure 5b) confirms that PC and THF stabilize the greatest number of contact/aggregate ion pairs ($n_{\text{Na-P}} \approx 5$ –6), whereas DME, DEGDME and TEGDME limit Na– PF_6 CIPs to lower values. The complementary Na–O (DME) curves (Figure 5c,d) show the inverse trend, where glymes furnish the densest oxygen solvation in the primary Na^+ coordination shell. Thus, solvents with high donor number ether oxygens (DME, DEGDME, or TEGDME) most effectively compete with PF_6^- for Na^+ coordination, yielding a larger population of “free” charge-carrying Na^+ ions (Figure 5e), consistent with experimental trends. Overall, major factors governing the solvent-dependent solvation hierarchy include (1) Lewis basicity of coordinating oxygen (Gutmann donor number), which sets intrinsic Na^+ –O affinity; (2) solvent topology and flexibility, which determine whether donor oxygens can simultaneously coordinate Na^+ (DEGDME forms a compact, near-octahedral chelate that excludes PF_6^- , while more flexible TEGDME leaves space for PF_6^- in the first shell); and (3) local dipole symmetry, which differentiates cyclic vs linear solvents and explains stronger CIP stabilization in PC and THF compared to DMC. The trends in Figure 5 arise from the interplay of these factors.^{57–62}

Interestingly, the running coordination number profiles show that Na–O coordination is nearly identical in DEGDME and TEGDME (Figure 5d), yet the extent of ion pairing differs substantially (Figure 5b). Figure 5e–g rationalize this behavior, demonstrating that while both solvents supply six ether oxygens to the primary solvation shell, differences in chain flexibility and local packing modulate the stabilization of contact and solvent shared ion pairs. Figure 5e shows that the fraction of unpaired Na^+ in DEGDME decays more slowly, remaining above 65% (of total Na^+) at $r = 5$ Å, whereas TEGDME falls below 40% by the same radial distance from the Na^+ centers. The complementary metric in Figure 5g reveals a larger population, $>20\%$ at $r = 5$ Å, of NaPF_6 CIPs in TEGDME relative to DEGDME. Snapshots in Figure 5f,h clarify the microscopic origin, where DEGDME wraps into a compact, nearly octahedral chelate that sterically excludes PF_6^- , favoring solvent separated or fully dissociated Na^+ . In contrast, the extended TEGDME chain adopts a flexible conformation that coordinates Na^+ while leaving void space that accommodates a PF_6^- (Figure 5f,h), thereby promoting CIP formation in the primary coordination shells. These conformational preferences demonstrate that subtle variations in solvent topology reshape the free-energy landscape of ion association, and that UMA-OMol captures the resulting redistribution among CIPs, SSIPs, and higher aggregates, with direct consequences for local solvation structure and charge transport.^{58,63,64}

f. Solid/Liquid Interfacial Structure. To assess the capability of UMA-OMol to capture electrolyte structuring at the electrode interface,^{65–68} we simulated a graphite/DME system containing 1 M NaPF_6 at 300 and 353 K. A representative snapshot of the interfacial configuration is shown in Figure S14. The number density profiles in Figure 6 reveal pronounced solvent and ion layering at the graphite interface, indicating a highly ordered interfacial region. At 300 K, DME oxygen atoms form a compact adsorption layer that preferentially coordinates interfacial Na^+ ions, while PF_6^- anions exhibit weaker interfacial accumulation, consistent with solvent-mediated

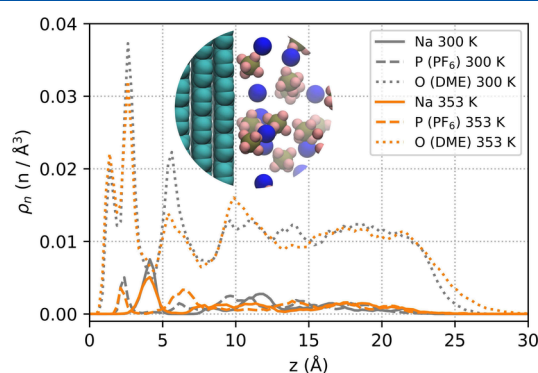


Figure 6. Number density profiles (ρ_u) of Na^+ , P (PF_6^-), and O (DME) atoms along the surface normal (z) at the graphite–electrolyte interface for NaPF_6 in DME at 300 K (gray) and 353 K (orange). At 300 K, pronounced solvent and ion layering is observed, with DME oxygens forming a compact interfacial layer that coordinates Na^+ near the electrode surface, while PF_6^- remains more weakly adsorbed. Elevated temperature reduces the amplitude of these oscillations, indicating thermal disruption of interfacial ordering and enhanced ion mobility. The inset shows a representative partial snapshot of the interfacial configuration highlighting solvent structuring adjacent to the graphite surface.

cation stabilization and limited anion contact with the electrode. The resulting oscillatory density profiles of oxygen, sodium, and phosphorus reflect a structured solvation environment in which Na^+ resides within the first solvent shell coordinated primarily by interfacial oxygens, as shown in Figure 6. This organization stabilizes Na^+ near the surface and constrains the initial electron transfer pathways that may initiate SEI formation.⁶⁹ Upon increasing the temperature to 353 K, the amplitude of these density oscillations decreases and the profiles broaden, signaling enhanced thermal motion, partial desolvation, and weakened ion–solvent correlations within the interfacial region. Such interfacial disorder is known to promote molecular exchange between the interface and bulk, accelerating ion transport while simultaneously altering the accessibility and reductive stability of Na^+ species at the electrode.^{70,71}

In this work, we benchmarked state-of-the-art large-scale MLIPs trained on inorganic materials DFT (Orb-OMat and SevenNet-OMat) and on the molecular DFT OMol25 data set (UMA-OMol) against experimental observables to assess their reliability in describing the structure and thermodynamics of Na-ion battery electrolytes. Starting from the macroscopic density across a diverse set of organic liquids and electrolytes, parity plots of simulated versus experimental densities show that UMA-OMol achieves near-unity correlation ($R^2 > 0.98$) with minimal systematic bias, while MLIPs trained on inorganic materials consistently underpredict experimental values and exhibit larger scatter. X-ray structure factors $S(Q)$ demonstrate that UMA-OMol captures all principal experimental features, validating its accuracy in representing medium-range ordering. Small deviations observed at low Q likely originate from long-range density correlations not captured by the MLIP given its 6 Å cutoff. Difference plots, $\Delta S(Q)$, show that UMA-OMol exhibits the smallest residuals and minimal oscillatory behavior compared to SevenNet-OMat and Orb-OMat, confirming its superior fidelity in reproducing experimental scattering observables. Together, these results highlight how the chemically diverse and high-quality OMol25 molecular DFT training data set increases MLIP prediction fidelity for intermolecular forces and improves thermodynamic consistency across liquid and electrolyte chemistries.

Subsequent investigations using UMA-OMol to simulate Na-ion electrolyte systems with molecular dynamics while varying anion identity, temperature, concentration, solvent, and of a solid–liquid interface revealed atomistic insights useful to battery electrolyte design. Across the anion chemistries considered, OTf^- displays substantially stronger association with Na^+ than PF_6^- or TFSI^- , with ion pairing strengths strongly modulated by solvent topology through its control over primary coordination shell structure and competitive solvent binding. Our results demonstrate that UMA-OMol captures ion-pair hierarchies and binding strengths. Temperature-resolved simulations uncover the dynamic evolution of solvation and ion association. As temperature increases, $\text{Na}^+ - \text{O}(\text{DME})$ coordination weakens while $\text{Na}^+ - \text{P}$ coordination strengthens, showing a transition from solvent-separated to contact or solvent-shared ion pairs. This structural reorganization parallels that observed in Li-based electrolytes but is more pronounced for Na due to its lower charge density and weaker solvation enthalpy. Finally, solvent topology emerges as a critical determinant of ionic association. Short-chain glymes (DME, diglyme) suppress NaPF_6 CIPs by stabilizing solvent-separated configurations,

whereas conformationally flexible and extended TEGDME enhances overcoordination and ion aggregation. This solvent-dependent modulation of ion-pairing directly rationalizes the experimentally observed relationship between free-ion population and ionic conductivity. Similarly, our simulations revealed that UMA-OMol reproduces strong, temperature-dependent structuring of electrolyte species at the graphite interface, with pronounced solvent and cation layering at room temperature that weakens at elevated temperature, driving migration of ions from the electrode–electrolyte interface to the bulk phase.

OMol25 and UMA have demonstrably advanced MLIP capabilities for liquid electrolyte simulations; however, there remain important areas for future improvement. *First*, while UMA-OMol substantially improves electrolyte MD stability compared to Orb-OMat and SevenNet-OMat, trajectories do still occasionally become unstable, even after equilibrating initial simulation boxes with a classical force field. Driving discovery with, e.g., high-throughput screening will thus likely require either MLIP architectures with improved potential energy surface smoothness⁷² or an automated MLIP-MD workflow infrastructure with robust on-the-fly error handling.⁷³ *Second*, electrolyte simulations in particular will benefit from MLIP architectures that go beyond short-range graph construction cutoffs—e.g., via explicitly physics-informed approaches^{74,75} or global attention-based schemes^{76,77}—to accurately describe long-range electrostatics while still effectively scaling to tens of thousands of atoms. UMA-OMol is trained on molecular cluster configurations computed in a Gaussian basis; the residual low- Q discrepancy in the simulated $S(Q)$ is consistent with this cluster-to-periodic extrapolation and with the absence of an explicit long-range Coulomb tail in the model. Architectures that capture long-range interactions offer a natural route to closing this gap. Importantly, model architectures that explicitly account for long-range interactions have only recently become available, such as MACE-POLAR, AllScAIP, and OrbMol-v2. Our results indicate that incorporating long-range interactions can further improve predictions relative to experimentally characterized structures (Figure S18). A systematic head-to-head comparison of these approaches using the present experimental benchmark set represents a natural next step and is the focus of ongoing work.^{78,79}

Third, simulating transport properties in liquid electrolytes, and even more so in polymer electrolytes,⁸⁰ necessitates reaching MD time scales that are out of reach for current large-scale pretrained MLIPs. While improving GPU hardware will help, model distillation also shows great promise,⁸¹ provided that a standardized and automated procedure can be established that marries pretrained model accuracy with at least an order of magnitude inference speedup. Transport observables such as self-diffusivities and ionic conductivities converge on substantially longer time scales than the structural observables; distilled, system-specialized variants of UMA-OMol designed for such time scales are an active line of development. *Fourth*, it remains unclear if range-separated hybrid DFT, as was used to construct OMol25, is sufficient to train MLIPs that predict experimental observables within chemical accuracy. It may be that electronic structure data beyond DFT accuracy will be required for model fine-tuning, although generating such data spanning the same chemical diversity as OMol25 will require massive computational resources. *Finally*, we see great potential in more closely

marrying high-quality experimental data to MLIP-based atomistic simulations. Establishing comprehensive experimental benchmarks with broad chemical diversity and varied observables that can be directly compared to atomistic simulations will quantify model accuracy as architectures and training data improve and facilitate hybrid approaches like digital twins⁸² and multimodal learning on both experimental and ab initio data.^{83,84}

■ ASSOCIATED CONTENT

■ Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.jpclett.6c01037>.

Methodology, system densities, structure factors, band-resolved mean absolute residuals, radial distribution functions and coordination number, time evolution of temperature, density, and total energy, concentration effect, system snapshot, fraction of free ions, and raw data for all experimental measurements (PDF)

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