DEVELOPMENT OF MACHINE-LEARNED POTENTIALS FOR THE COARSE-GRAINED MODELLING OF MOLECULAR SYSTEMS

D.-P. Gerakinis[1,3], Ricci E.[1,2], Giannakopoulos G.[2], Karkaletsis V.[2], Theodorou D.N.[3], **N. Vergadou[1,*]**

[1]Institute of Nanoscience and Nanotechnology, National Centre for Scientific Research "Demokritos", Athens, Greece

[2]Institute of Informatics and Telecommunications, National Centre for Scientific Research "Demokritos", Athens, Greece

[3]School of Chemical Engineering, National Technical University of Athens, Greece

(*n.vergadou@inn.demokritos.gr)

ABSTRACT

Molecular simulation is a particularly efficient and reliable means for the elucidation of molecular mechanisms that underlie materials macroscopic behaviour, the design of new materials and the prediction of their properties. The wide length- and time scale phenomena that are present in complex chemical systems necessitate the development and implementation of hierarchical multiscale schemes. Coarse-Graining (CG) is, in many cases, at the core of multiscale methods. With the increasing advancements of Artificial Intelligence (AI) in materials science, and in particular of Machine Learning (ML) techniques, the possible synergies between these methods and molecular modelling and simulation are currently explored, towards the development of more generalizable and accurate multiscale simulation schemes[1]. ML techniques have been investigated in the recent years for the development of improved atomistic force fields based on quantum mechanical calculations[2]. However, the integration of ML methods into the development of CG force fields required for hierarchical multiscale modelling schemes for bulk fluids and complex chemical systems, , on the basis of atomistic simulations, is still very scarce[3].

In this work[4], neural network-based models capable of learning complex potential energy hypersurfaces and many-body interactions are utilized for the approximation of the CG potentials of mean force (PMFs), replacing predefined analytical functionals. Graph Convolutional Neural Networks architectures are adopted[5,6] to develop CG Machine Learned potentials, implementing and optimizing a scheme[7] that includes a force-matching procedure[8]. The obtained models are used to perform CG Molecular Dynamics simulations and the structural and thermodynamic properties of the CG systems are systematically compared with the underlying atomistic reference to quantify the effectiveness of the developed models. The effects of hyperparameters, loss function construction and GCNN architecture size were thoroughly investigated, providing a wealth of information that can serve as a general basis for the reliable use of ML-based CG approaches in the wide field of bulk soft matter systems.

KEYWORDS: Multiscale Modeling, Molecular Simulations, Coarse-graining, Machine Learning, Graph Convolutional Neural Networks

REFERENCES

[1] Peng GCY, Alber M, Buganza Tepole A, Cannon WR, De S, Dura-Bernal S, Garikipati K, Karniadakis G, Lytton WW, Perdikaris P, Petzold L, Kuhl E. (2021). *Arch. Comput. Methods Eng.*, 28(3), 1017–1037.

[2] Unke OT, Chmiela S, Sauceda HE, Gastegger M, Poltavsky I, Schütt KT, Tkatchenko A, Müller K-R. (2022). *Chem. Rev.*, 121(16), 10142–10186.

[3] Ricci E, Vergadou N. (2023). *J. Phys. Chem. B*, 127(11), 2302–2322.

[4] Gerakinis D-P, Ricci E, Giannakopoulos G, Karkaletsis V, Theodorou DN, Vergadou N. (2024). *Zenodo,* https://doi.org/10.5281/zenodo.10501038
[5] Schütt KT, Sauceda HE, Kindermans P-J, Tkatchenko A, Müller K-R. (2018). *J. Chem. Phys.*, 148(24), 241722.
[6] Batzner S, Musaelian A, Sun L, Geiger M, Mailoa JP, Kornbluth M, Molinari N, Smidt TE, Kozinsky B. (2022). *Nat. Commun.*, 13(1), 2453.
[7] Ricci E, Giannakopoulos G, Karkaletsis V, Theodorou DN, Vergadou N. (2022). Association for Computing Machinery (ACM) – International Conference Proceeding Series, 52, 1–6, https://doi.org/10.1145/3549737.3549793
[8] Noid WG, Chu JW, Ayton GS, Krishna V, Izvekov S, Voth GA, Das A, Andersen HC. (2008). *J. Chem. Phys.*, 128(24), 244114.