Exploiting artificial neural networks in simulation of complex ionic chemical environment

Veronika Jurásková[a], Kevin Rossi[b], Raphael Wischert[c], Laurent Garel[d], Michele Ceriotti[b], Clémence Corminbœuf[a]

[a] *Laboratory for Computational Molecular Design (LCMD), Ecole Polytechnique Fédérale de Lausanne, Lausanne, Switzerland*
[b] *Laboratory of Computational Science and Modeling (COSMO), Ecole Polytechnique Fédérale de Lausanne, Lausanne, Switzerland*
[c] *Eco-Efficient Products and Processes Laboratory, Solvay, RIC Shanghai, China*
[d] *Aroma Performance Laboratory, Solvay, RIC Lyon, France*
clemence.corminboeuf@epfl.ch

Ab initio molecular dynamics of condensed phase systems featuring charged species rely upon the accurate quantum description of highly complex chemical environment (e.g., various structural patterns, subtle non-covalent interactions and charge transfer processes) at converged statistical sampling. Simulations of chemical reactions in solution are even more challenging due to the larger complexity of the free energy landscapes. Such condensed phase phenomena are ideally suited to drive quantum chemistry out of its comfort zone and combine the power of enhanced sampling MD techniques with the latest sophisticated innovations from quantum machine learning. Atomic potentials based on artificial neural networks (ANN) have been increasingly used to simulate bulk materials, surface adsorption, gas phase reactions or aqueous solutions.[1] ANN are capable of describing high dimensional PES at the same accuracy as the reference quantum chemical method but at a computational cost comparable to classical force field simulations.[2]
In the present work, we exploit ANN potentials based on PBE and PBE0 to model complex catalytic process in condensed phase. Particular emphasis is placed on the accurate description of subtle ion-solvent interactions under various compositions and thermodynamic conditions. Our simulation protocol is generally applicable to model thermodynamic and structural properties of condensed phase systems and chemical reactions in a complex environment. The synergic application of state-of-the-art algorithms such as multiple timestepping[3], replica exchange sampling, farthest point and CUR feature selection[4] accelerates the sampling of the reference training set and affords stable simulations with relevant time scales.

References

1. Behler, J. *Angew. Chem. Int. Ed.*, **56** (2017), 12828 12840.
2. Behler, J., Parrinello, M. *Phys. Chem. Lett.*, **98** (2007), 146401.
3. Kapil, V., VandeVondele, J., Ceriotti, M. *J. Chem. Phys.*, **144**(2016), 054111.
4. Imbalzano, G., Anelli, A., Giofr, D., Klees, S., Behler, J., Ceriotti, M. *J. Chem. Phys.*, **148** (2018), 241730.