Computational Studies of Supercooled Water with an *Ab-Initio* Deep Neural Network Model

Pablo G. Debenedetti[1], Pablo M. Piaggi[2], Thomas E. Gartner III[3] , Athanassios Z. Panagiotopoulos[1] and Roberto Car[2]

[1]Dept. of Chemical & Biological Engineering, Princeton University, Princeton, NJ 08544, USA

[2]Dept. of Chemistry, Princeton University, Princeton, NJ 08544, USA

[3]School of Chemical & Biomolecular Eng., Georgia Inst. of Technology, Atlanta, GA 30332, USA

pdebene@princeton.edu

Abstract

Using a combination of computational chemistry, machine learning and advanced sampling, it is possible to study, with *ab-initio* accuracy, problems involving time and length scales that until recently were only accessible using classical, empirical models [1]. This approach will be illustrated through three examples. First, the phase behavior of supercooled water is investigated via equation of state [1] and free energy calculations [2]. Both approaches yield evidence consistent with the existence of a metastable first-order phase transition between two different liquids, terminating at a liquid-liquid critical point. The second example involves calculations of ice homogeneous nucleation rates in supercooled water at ambient pressure, which yields predictions that are in good agreement with experiments [3]. Finally, melting curve calculations for several ice polymorphs allow the computational re-creation of the Mishima-Stanley experiment [4], which for many years provided the strongest evidence in support of the existence of a metastable critical point in supercooled water. The computational results suggest a different interpretation of experimental observations than that proposed by Mishima and Stanley [5].

REFERENCES

[1] Gartner III, T.E., Zhang, L., Piaggi, P.M., Car, R., Panagiotopoulos, A.Z., and Debenedetti, P.G., PNAS, 117 (2020) 26040.

[2] Gartner III, T.E., Panagiotopoulos, A.Z., Car, R., and Debenedetti, P.G., in preparation (2022).

[3] Piaggi, P.M., Weis, J., Panagiotopoulos, A.Z., Debenedetti, P.G., and Car, R., submitted to PNAS (2022).

[4] Mishima, O. and Stanley, H.E., Nature, 392 (1998) 164.

[5] Piaggi, P.M., Gartner III, T.E., Car, R., and Debenedetti, P.G., in preparation (2022).