Exploring Iridium-Doped ZrO_2 Structures Using an Interatomic Potential-Based Workflow

Mathias Stokkebye Nissen[1], Heine Anton Hansen[1]

[1]DTU Energy Atomic Scale Materials Modelling (ASM), Anker Engelunds Vej, 301, 110, 2800 Kgs. Lyngby, Denmark
matnis@dtu.dk

In a Nutshell

To discover new catalysts for the oxygen evolution reaction (OER), we have made atomistic simulations of Iridium (Ir) doped ZrO_2. This we have undertaken via an iterative workflow based around the interatomic potential MACE [1-2] in combination with density functional theory (DFT).

Motivation

The transition to renewable energy requires efficient energy storage and conversion. Hydrogen, production via water electrolysis in polymer electrolyte membrane (PEM) cells, offers a high efficiency storage solution and an ability to operate in a load-following mode [3-4]. However, its reliance on Iridium, which is scarce and expensive, for OER catalysts limits scalability. To address this, we have investigated Ir-doings into ZrO_2 examples of which are shown in Figure 1, which is considered a non-critical raw material by the EU [5].

Method

An overview of the applied workflow is observed in Figure 2. The workflow is based on iteratively training an ensemble of MACE interatomic potentials, relaxing the atomic configurations of interest, evaluating these structures via DFT and retraining an ensemble with the new DFT calculations included as training data. This approach resembles methods deployed in previous studies [6-7] where the interatomic potentials were based on the PaiNN [8] and NequIP [9] architectures.

Conclusions

For relaxing the monoclinic ZrO_2 surfaces (observed in Figure 1), we estimate a reduction of two-thirds in the required number of single-point DFT calculations as compared to relaxing the atomic structures solely using DFT. Additionally, although the employed method ensures structures with low forces are obtained within just a few iterations, the reduction of force magnitudes stagnates after five iterations. The process of achieving structures with forces below the threshold of 0.05 eV/Å (considered as relaxed) is thus slower. This is shown in Figure 3.

Acknowledgements

The project is supported by the Clean Hydrogen Partnership and its members Hydrogen Europe and Hydrogen Europe Research under Grant Agreement No 1012101479.

References

[1] Ilyes Batatia, David Peter, Kovacs, Gregor N. C. Simm, Christoph Ortner, Gabor Csanyi, (2022) https://openreview.net/forum?id=YPpSngE-ZU

[2] Ilyes Batatia, Simon Batzner, Dávid Péter Kovács, Albert Musaelian, Gregor N. C. Simm, Ralf Drautz, Christoph Ortner, Boris Kozinsky, Gábor Csányi, arXiv, arXiv:2205.06643 (2022), https://doi.org/10.48550/arXiv.2205.06643

[3] Katherine E. Ayers, Everett B. Anderson, Christopher Capuano, Blake Carter, Luke Dalton, Greg Hanlon, Judith Manco and Michael Niedzwiecki, ECS Transactions (2010) page 3, https://dx.doi.org/10.1149/1.3484496

[4] Y. Lin, Y. Dong, X. Wang, L. Chen, Adv. Mater. 35 (2023), page 2210565. https://doi.org/10.1002/adma.202210565

[5] EU: Critical raw materials. https://single-market-economy.ec.europa.eu/sectors/raw-materials/areas-specific-interest/critical-raw-materials_en

[6] Ai, C., Han, S., Yang, X., Vegge, T., & Hansen, H. A, American Chemical Society 16 (2024) page 12563; https://doi.org/10.1021/acsami.3c18734

[7] Yang, X., Petersen, M.H., Sechi, R., Hansen, W.S., Norwood, S.W., Krishnan, Y., Vincent, S., Busk, J., Cornet, F.R.J., Winther, O. and Lastra, J.M.Gy, ChemRxiv (2024), https://doi.org/10.26434/chemrxiv-2024-p5t3l

[8] Kristof T. Schütt, Oliver T. Unke, Michael Gastegger, arXiv, arXiv:2102.03150, https://doi.org/10.48550/arXiv.2102.03150

[9] Batzner, Simon, Albert Musaelian, Lixin Sun, Mario Geiger, Jonathan P. Mailoa, Mordechai Kornbluth, Nicola Molinari, Tess E. Smidt, and Boris Kozinsky, Nature communications 13, no. 1 (2022): 2453, https://doi.org/10.1038/s41467-022-29939-5

Figures

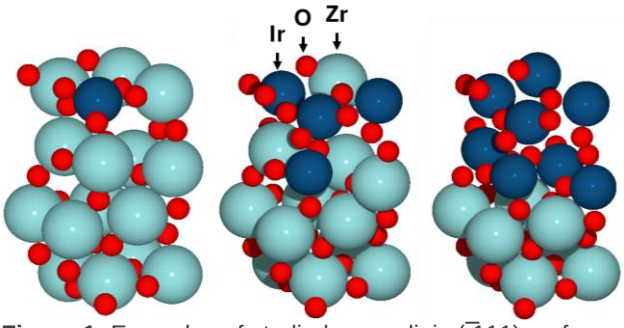

Figure 1. Examples of studied monoclinic ($\bar{1}11$) surfaces, with different amounts of Ir-dopant in overlayer.

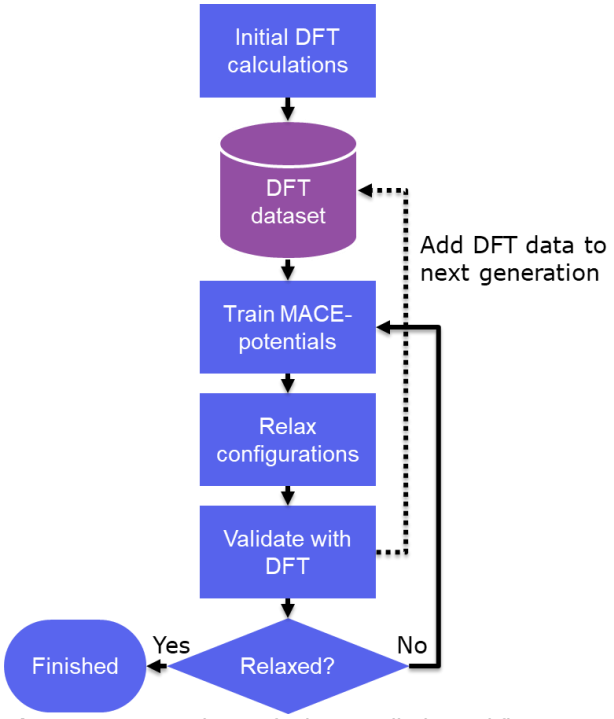

Figure 2. Overview of the applied workflow - see explanation in the text.

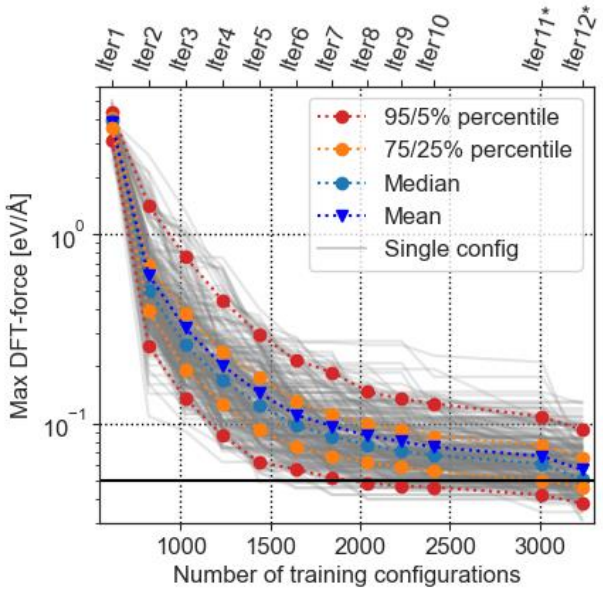

Figure 3. Plot of the maximum force, obtained via DFT, across 256 atomic configurations for each iteration of the employed workflow for structure optimization.