How to Assess AIMD Papers: a Guide for the Computational Potential of Zero Charge

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Purpose of this Note

This short guide is written for colleagues in our research group who are primarily experimentalists. Its goal is to help you navigate and evaluate scientific papers that rely on **ab initio molecular dynamics (AIMD)** simulations, a method widely used in computational chemistry to model the atomic-scale behavior of systems in realistic (e.g., liquid, finite temperature) conditions. This guide places particular emphasis on how AIMD is used to evaluate the potential of zero charge (PZC) of metal/water interfaces.

Where to Look: The "Computational Details" Section

Most computational chemistry papers include a section called:

- **Computational Details**
- **Methods**
- **Theoretical Methods**

This section typically appears:

- After the Introduction section
- At the end of the main text (before the Acknowledgements)
- In the Supporting Information (SI)

Key Elements to Look For

To evaluate whether a PZC computed from an AIMD study is trustworthy and of high quality, check for the following information (you can directly Ctrl+F the keywords written in blue in the following subsections for a faster research in the publications):

1. Software and Level of Theory

- Type of calculation: **AIMD** (Ab Initio Molecular Dynamics) should be clearly stated. While other computational approaches such as classical Molecular Dynamics (cMD), also referred to as Force Field Molecular Dynamics (FFMD), or static Density Functional Theory (DFT) calculations can be used, they are not reliable for evaluating the PZC.

- Name of the **code** or **software package** used should be provided—for example, CP2K, VASP, or Quantum ESPRESSO. If the code is not explicitly mentioned, be aware that the following are classical MD codes (cMD/FFMD) and are not suitable for PZC evaluation: LAMMPS, GROMACS, AMBER, CHARMM, and MetalWalls. Note that cMD/FFMD codes are often used to *pre-equilibrate* the simulation cells. Make sure that the *production runs* are produced with and AIMD simulation.

- Exchange-correlation **functional**: The choice of exchange-correlation functional should be specified, as it significantly influences the accuracy of the results. GGA functionals, such as PBE, RPBE, PBEsol, and BLYP, are widely used due to their computational efficiency, but they typically underestimate the WF/PZC by approximately 0.2–0.4 V. Meta-GGA functionals, such as SCAN, seems to provide improved accuracy.

- **Dispersion** corrections, stated as DFT-D3, Grimme corrections, or van der Waals functionals (e.g. SCAN+rVV10) should be included. No dispersion corrections can lead to a stronger underestimation of the WF/PZC.

2. **Simulation Parameters**

- **Time step** for integration (typically 0.5–1.0 fs)

- **Simulation time** (at least several picoseconds, ideally more than 10 ps)

- **Thermostat**, e.g., Nosé-Hoover, CSVR, Langevin

3. **System Setup**

- Size of the simulation **cell**, e.g., 3 x 4, 6 x 6. A cell smaller than 6 x 6 seems to lead to an underestimation of the WF/PZC by about 0.2 – 0.6 V.

- Number of **atomic layers** (at least 4 layers)

- Length of the **water** region (should be > 20 Å, otherwise possible uncertainty on the PZC of about 0.3 V for the cSHE method)

- *Discuss symmetric vs. non-symmetric cells*

4. **Reference Potential Method**

- The method **WF** or **cSHE** is always clearly stated

- Any PZC obtained via the WF method is hardly reliable due to the non-equilibration of the surface potential of water at the water/vacuum interface

- If the cSHE method has been used, consider a possible uncertainty of 0.15 – 0.25 V

Final Advice

- A good AIMD paper clearly states its approximations and limitations.

- Be cautious with very short simulations or small system sizes.

- If anything is unclear, feel free to reach out—I'm happy to help interpret the computational part of any paper.