Algorithms and codes for the simulation of explicit electrodes

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1. Proposal

1.1 Introduction and motivation

There has recently been huge interest from the molecular simulation community for studying electrochemical systems. This is due to the need for new energy production and storage devices (such as photo-electrochemical cells, batteries or supercapacitors) and to the development of advanced materials that require to be understood at the molecular scale. However, methodological difficulties still need to be overcome before one can model realistic devices in order to propose a correct interpretation of the mechanisms at play.

1.2 State of the art

The level of sophistication of the simulations depend on the nature of the studied devices. For example, supercapacitors are charged by the adsorption of the ions at the surface of porous carbon electrodes - no redox reactions are involved. Such systems are therefore widely studied using classical molecular dynamics with explicit electrodes with a constant applied potential between them. On the contrary, in photocatalysis devices, coupled proton electron transfer reactions occur. It is therefore necessary to use density functional theory-based molecular dynamics, in which thermodynamic integration approaches yield accurate free energies for the various reactions.

1.3 Objectives

Despite the success of these approaches, a lot of work remains to be done for increasing the range of electrochemistry problems that can be tackled using molecular simulations. The objective of this discussion meeting is to propose new algorithms and how they can be implemented in various simulation techniques. In particular, we will mainly discuss the two following points:

i) Is it possible to control a computational electrode in molecular dynamics simulations using fields rather than potentials?

ii) How can we include explicit electrodes in simulation approaches involving implicit solvents (Brownian dynamics, molecular density functional theory)?

The discussion meeting will gather a small number of participants, who are all involved in code development. We aim at proposing a larger-scale CECAM workshop on this topic when the discussions will be more mature.

1.4 **Participant List** (*italic: invited)*

P. Madden (Oxford)
M. Sprik (Cambridge)
G. Jeanmairet (Stuttgart)
R. Vuilleumier (PASTEUR - ENS/UPMC)
M. Jardat (PHENIX - UPMC)
M. Haefele (Maison de la Simulation - CNRS)
Z. Li (Maison de la Simulation - CEA)
T. Mendez Morales (Maison de la Simulation - CEA)
M. Burbano (Maison de la Simulation - CEA)
A. Asta (PHENIX - UPMC)
D. Borgis (PASTEUR - CNRS et Maison de la Simulation - CNRS)
M. Levesque (PASTEUR - CNRS et Maison de la Simulation - CNRS)
B. Rotenberg (PHENIX - CNRS)
M. Salanne (PHENIX - UPMC et Maison de la Simulation - CNRS)
D. Arrismendi Arrieta (Madrid - Maison de la Simulation)

2. Financial Support

We ask CECAM-MOSER 1760 euros to fund the following:
-Transportation for P. Madden: 250 euros
-Transportation for M. Sprik: 250 euros
-Transportation for G. Jeanmairet: 150 euros
-Hotel for P. Madden (1 night): 120 euros
-Hotel for M. Sprik (1 night): 120 euros
-Coffee breaks: 120 euros
-Dinner on the 20th for 16 participants: 750 euros

3. Tentative program

20/07

14h-14h40: P. Madden "History of Metalwalls" + discussion

15h-15h20: M. Haefele "Recent improvements in Metalwalls + roadmap" + discussion

15h40: Coffee break

16h-16h40: M. Sprik "Maxwell electrode" + discussion

21/07

9h-9h15: B. Rotenberg "ANR project: Listening to the Electrical Noise for Nanofluidic Sensing" + discussion

10h-10h30: M. Levesque "Molecular density functional theory" + discussion

11h: Coffee break

11h30: Z. Li "Marcus free energies for Fe^{3+}/Fe^{2+} couple in ionic liquids"

12h: T. Mendez-Morales "Modelling nanoporous graphene-based supercapacitors"

12h30: G. Jeanmairet "Extension of Marcus picture for electron transfer reactions with large solvation changes"