Electrowetting for advanced electrochemical technologies

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Wetting phenomena hold a pivotal role in a vast range of physicochemical processes occurring in both nature and artificial systems. Elucidation of the chemical physics underlying these processes paves the way towards their elaborate control and inspires the development of technologically advanced devices with a diverse range of applications spanning various micro-/nano-fluidics platforms and electrotribology to electrochemical energy conversion, storage, and capacitive deionization. Several strategies have been sought to control wetting through external stimuli. The oldest such approach can be traced back to Gibbs – Lippmann theory of electrocapillarity and relies on the phenomenon of electrowetting.[1,2] Despite the experimental evidence of electrowetting on conducting substrates by Frumkin, this fascinating phenomenon has been initially overlooked due to its apparent irreversibility on solid electrodes arising by their intrinsic surface reactivity. Herein, almost a century later than these seminal works, we discuss a series of novel insights into the overall phenomenon by employing highly oriented and low dimensional carbon allotropes. A pioneering strategy is introduced to control electrowetting on graphite by effectively suppressing the underlying parasitic, electrochemically induced surface processes.[3] In this way, the electrowetting response exhibits a fully symmetric and reversible wetting/dewetting behaviour in line with the classical electrowetting theory described by the Young – Lippmann equation. To describe the thermodynamics of the electrochemical interface, a multiscale quantum mechanics–classical molecular dynamics approach is presented that establishes a conceptual framework for the investigation of wetting phenomena on a single layer graphene model surface under the application of potential bias.[4] It is shown that the charge stored at the electrochemical double layer is not the only factor that governs the interfacial energetics. Competitive effects emerging by ion adsorption and hydrogen bond number have a strong impact on the interfacial surface tension. Extending our general strategy to a reactive interface, electrowetting has been studied under the occurrence of foundational electrochemical reactions related to electrochemical energy storage and conversion systems. By exploiting the unique layered structure of graphite with its tuneable interlayer distance that sets almost ideal conditions for the accommodation of ions into its structure, we demonstrate the significant effect of anion intercalation on the electrowetting response of graphitic surfaces in contact with concentrated aqueous and non-aqueous electrolytes as well as ionic liquids.[5] Furthermore, cathodically induced intercalation is proved to drive superhydrophilicity in other materials, such as high entropy thin alloy films.[6] Both phenomena are interpreted in terms of changes in the physical properties of the substrates impelled by the intercalation reactions. Our work provides fundamental insights into wetting phenomena under the application of an external electrical field, while at the same time stimulates novel research towards utilizing electrowetting to boost the performance of devices operating based on physicochemical processes at the conductor/electrolyte interface.

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References

1. A. N. Frumkin, A. V. Gorodetskaya, B. Kabanov and M. Nekrasov, *Phys. Z. Sowjetunion*, 1932, 255–284.
2. A. A. Papaderakis and R. A. W. Dryfe, *Current Opinion in Electrochemistry*, 2023, **38**, 101245. https://doi.org/10.1021/acs.jpcc.2c06517
3. A. A. Papaderakis, K. Polus, P. Kant, F. Box, B. Etcheverry, C. Byrne, M. Quinn, A. Walton, A. Juel and R. A. W. Dryfe, *J. Phys. Chem. C*, 2022, **126**, 21071–21083. https://doi.org/10.1021/acs.jpcc.2c06517
4. Z. Wei, J. D. Elliott, A. A. Papaderakis, R. A. W. Dryfe and P. Carbone, *J. Am. Chem. Soc.*, 2024, **146**, 760–772. https://doi.org/10.1021/jacs.3c10814
5. A. A. Papaderakis, A. Ejigu, J. Yang, A. Elgendy, B. Radha, A. Keerthi, A. Juel and R. A. W. Dryfe, *J. Am. Chem. Soc.*, 2023, **145**, 8007–8020. https://doi.org/10.1021/jacs.2c13630
6. A. A. Papaderakis, E. M. Paschalidou, L. Zendejas Medina, B. Gault, B. Sefer, D. Malmström, K. Tschulik, A. Juel, R. A. W. Dryfe, *Under submission*