

Functionalized Liquid–Liquid Interfaces

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2007 J. Phys.: Condens. Matter 19 370301

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PREFACE

Functionalized Liquid–Liquid Interfaces

Most natural processes take place at interfaces. For this reason, surface science has been a focal point of modern research. At solid–liquid interfaces one can induce various species to adsorb or react, and thus may study interactions between the substrate and adsorbates, kinetic processes, optical properties, etc. Liquid–liquid interfaces, formed by immiscible liquids such as water and oil, have a number of distinctive features. Both sides of the interface are amenable to detailed physical and chemical analysis. By chemical or electrochemical means, metal or semiconductor nanoparticles can be formed or localised at the interface. Surfactants can be used to tailor surface properties, and also to place organic molecular or supermolecular constructions at the boundary between the liquids. Electric fields can be used to drive ions from one fluid to another, or even change the shape of the interface itself. In many cases, both liquids are optically transparent, making functionalized liquid–liquid interfaces promising for various optical applications based on the transmission or reflection of light. An advantage common to most of these systems is self-assembly; because a liquid–liquid interface is not mechanically constrained like a solid–liquid interface, it can easily access its most stable state, even after it has been driven far from equilibrium.

This special issue focuses on four modes of liquid-liquid interfacial functionalization: the controlled adsorption of molecules or nanoparticles, the formation of adlayers or films, electrowetting, and ion transfer or interface-localized reactions. Interfacial adsorption can be driven electrically, chemically, or mechanically. The liquid–liquid interface can be used to study how anisotropic particles orient at a surface under the influence of a field, how surfactants interact with other adsorbates, and how nanoparticles aggregate; the transparency of the interface also makes the chirality of organic adsorbates amenable to optical study. Film formation goes a step beyond adsorption; some surfactants form monolayers or multilayers at the interface. A polymer microfilm or a polymer–particle matrix can be synthesized at the liquid–liquid boundary. Such films exhibit unique adsorption and ion-intercalation properties of their own.

Electrowetting refers broadly to the phenomenon in which an applied voltage modulates the shape of a liquid–liquid interface, essentially by altering the surface tension. Electric fields can be used to induce droplets on solid substrates to change shape, or to affect the structure of liquid–liquid emulsions. Various chemical reactions can be performed at the liquid–liquid boundary. Liquid–liquid microelectrodes allow detailed study of ion-transfer kinetics at the interface. Photochemical processes can also be used to control the conformations of molecules adsorbed at the interface.

But how much precise control do we actually have on the state of the interfacial region? Several contributions to this issue address a system which has been studied for decades in electrochemistry, but remains essentially unfamiliar to physicists. This is the interface between two immiscible electrolytic solutions (ITIES), a progressing interdisciplinary field in which condensed-matter physics and physical chemistry meet molecular electrochemistry.

Why is it so exciting? The reason is simple. The ITIES is chargeable: when positioned between two electrodes it can be polarized, and back- to-back electrical double layers form on both sides of the liquid–liquid interface. Importantly, the term immiscible refers not only to

oil and water but also to the electrolytes. Inorganic electrolytes, such as alkali halides, tend to stay in water, whereas organic electrolytes, such as tetrabutylammonium tetrphenylborate, stay in oil. This behaviour arises because energies of the order of 0.2-0.3 eV are needed to drive ions across the interface. As long as these free energies of transfer are not exceeded by the external potential bias, the ITIES works as an 'electrode'; there is no traffic of ions across it. Thus the interface can sustain fields of the order of 10⁶ V/cm, which are localized in a nanoscopic layer near the interface. This gives many new options for building various kinds of electrically tunable self assembled molecular devices.

Through the years, ITIES have been considered by electrochemists as a popular biomimetic model system, or for studies of interfacial reaction kinetics; ITIES were also used in industrial phase-transfer catalysis. Recently, this system has opened up new options for nano-scale engineering of functional assemblies (for dense information storage, efficient energy conversion, light-harvesting, and miniaturized sensors), which justifies its presentation in this issue.

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