Materials Science of Supported Lipid Membranes

Atul N. Parikh and Jay T. Groves, Guest Editors

Abstract

Supported membranes represent an elegant route to designing well-defined fluid interfaces which mimic many physical–chemical properties of biological membranes. Recent years have witnessed rapid growth in the applications of physical and materials science approaches in understanding and controlling lipid membranes. Applying these approaches is enabling the determination of their structure–dynamics–function relations and allowing the design of membrane-mimetic devices. The collection of articles presented in this issue of *MRS Bulletin* illustrates the breadth of activity in this growing partnership between materials science and biophysics. Together, these articles highlight some of the key challenges of cellular membranes and exemplify their utility in fundamental biophysical studies and technological applications. The topics covered also confirm the importance of lipid membranes as an exciting example of soft condensed matter. We hope that this issue will serve readers by highlighting the intellectual scope and emerging opportunities in this highly interdisciplinary area of materials research.

Keywords: *biological*, *biomimetic*, *cellular.*

Lipid bilayer membranes are the universal material of choice for defining and controlling cellular organization in living systems. They are a major constituent of the biological membranes that serve as the outer boundary of cells and organelles.[1] As such, they serve as a means to compartmentalize, juxtapose, regulate, and generally mediate biomolecular interactions. The astounding complexity of life is intimately associated with the diverse physical–chemical properties of these membranes,[2] which include two-dimensional fluidity, material elasticity, thermal fluctuations, chemical diversity, and rich phase behavior.[3] In this regard, developing a materials-science-based understanding of lipid membranes is an exciting endeavor.

Traditional approaches to materials synthesis have largely relied on uniform, equilibrated phases leading to static "condensed matter" structures, for example, monolithic single crystals. Over the past several decades, these approaches have led to the development of a wide range of technologically useful materials including semiconducting, ferroelectric, nonlinear optical, superconducting, and piezoelectric materials. Important as they are, the range of functions in these materials is constrained by their static structure. In contrast, lipid bilayer membranes exploit their chemical heterogeneity, phase behavior, and dynamics to produce an impressive set of time-dependent functions for the biological membrane. Examples include stimuli-induced protein clustering, lipid reorganization, and topographical changes that regulate broad classes of biological functions of membranes[4,5] including signaling, molecular recognition, and transport. This spatio-temporal mode of lipid organization, or dynamic self-assembly, in biological membranes[6] exemplifies a major shift in emphasis from thermodynamic to kinetic regimes. Here, equilibrium structures (global free-energy minima) are replaced by higher-order organizational states representing facile transitions between various local metastable free-energy minima of different structures. Examples of synthetic materials deliberately fabricated using such a dynamic self-assembly approach are rare, reflecting the current lack of a fundamental understanding of such processes. We assert that the creation of new materials capable of performing advanced technological functions, for example, complex and cooperative processes for memory, self-replication, and self-repair, will ultimately rely upon understanding and controlling dynamic self-assembly in both natural and synthetic systems. We emphasize that understanding the interrelations between structure, assembly, dynamics, and function in lipid membranes contributes toward these broad objectives of contemporary materials science.

Furthermore, there are many direct and practical benefits of learning to devise synthetic materials that mimic the key properties of cellular membranes. Such materials may provide new classes of biosensors, diagnostic tools, biocompatible materials, and high-throughput characterization platforms for rapid and early detection of interactions between cells and their environment. Examples include sensors and assays for detecting toxins and pathogens (e.g., in national security applications), air and water quality monitoring, and drug screening platforms. They also promise tools for life science research, for example, in developing a molecular-level understanding of cell surface mechanisms during cellular homeostasis and diseases.

Within this framework, one approach that is proving powerful involves interfacing lipid bilayer membranes to solid substrates via self-assembly and directed self-assembly methods. These membrane-mimicking architectures are collectively referred to as supported membranes[7] (Figure 1).

Supported membranes are typically formed at the solid–liquid interface when vesicular microphases of lipids spontaneously rupture and spread on hydrophilic surfaces (Figures 1a and 1b). Alternative strategies employ Langmuir–Blodgett troughs to successively transfer two lipidic monolayers from the air–water interface onto planar surfaces[3,8] (Figure 1c). Recent studies reveal that single lipid bilayers can also form when dried lipids stamped onto a hydrophilic surface experience hydration.[9] In all of these cases, single lipid bilayers are separated from the substrate surface through an intervening layer of hydration water (dark-blue layer in Figure 1d, variously estimated at 6–15 Å in thickness at bilayer–silica interfaces) and exhibit two-dimensional contiguity and fluidity resembling that of lipid membranes of vesicles and living cells.[10] Each of the two lipid layers in the lipid bilayer is called a "leaflet." In cells, the outer leaflet is in contact with the extracellular matrix, and the inner leaflet faces the cell cytoplasm. The precursor phases in these

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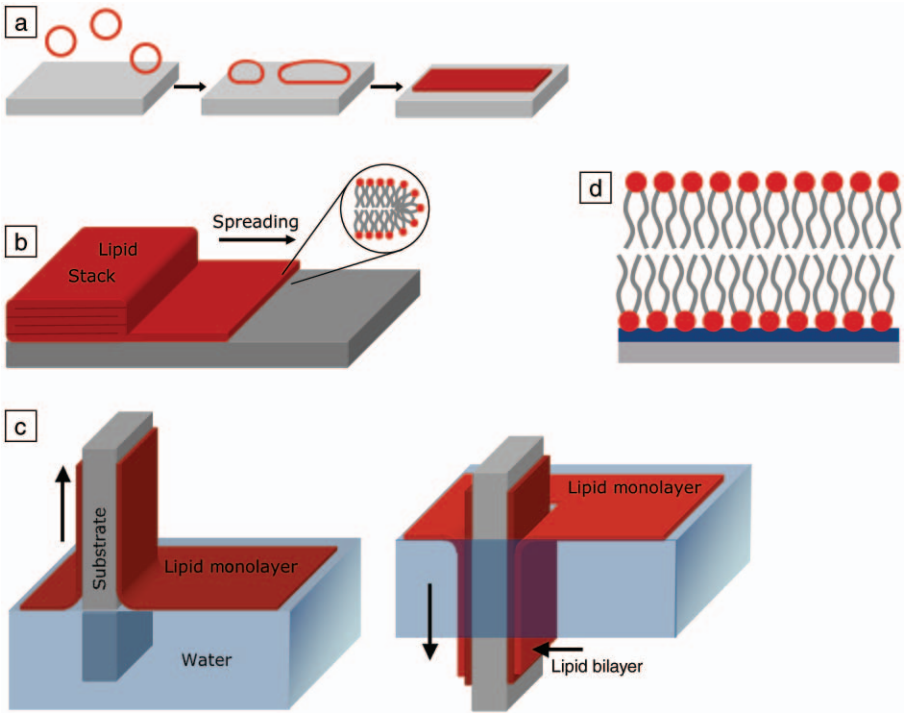

Figure 1. Schematic illustration of basic supported-membrane formation processes. (a) Vesicles adsorb, fuse, and rupture at solid surfaces; (b) lipid stacks spontaneously spread upon hydration; and (c) two successive transfers of Langmuir–Blodgett monolayers deposit pre-equilibrated lipid mixtures to form (d) single, supported bilayers on hydrophilic substrates. The dark-blue area is the intervening layer of hydration water between the substrate and the lipid.

approaches can consist of purified and synthetically tailored lipids and their complex mixtures in predetermined compositions, providing molecular-level control over their structure, chemical composition, phase state, and lateral fluidity. Moreover, because these constructs interface membrane architecture with solid substrates, they are amenable to a broad suite of surface analytical tools for molecular-scale characterization of their structure and dynamics and enable chip-based bioanalytical assays, sensors, and platforms.

We hope that this issue of *MRS Bulletin* serves to highlight the broad and rapidly evolving intellectual partnership between the materials sciences and membrane biophysics. This partnership underscores an interdisciplinary perspective in at least two ways.

First, it reveals how generating a scientific environment in which fundamental concepts obtained through detailed study of systems in one scientific area can be rapidly applied to generate understanding toward other, traditionally distinct, disciplines. For instance, it illustrates how the supported-membrane-based studies, aimed purportedly at understanding and mimicking biological membranes, provide an experimental test bed for questions in soft condensed matter, interfacial confinement, low-dimensional phases, chemistry in two dimensions, reaction-diffusion systems, and wetting.

Second, it demonstrates how the application of principles, tools, and methodologies developed extensively in one area of science can benefit the development of another, disparate discipline. We emphasize that the two-way traffic between fundamental studies of membranes and soft condensed matter research is rapidly elaborating. The articles presented here merely illustrate, rather than provide a comprehensive survey of, the breadth and the scope of this partnership.

Despite its popularity, the supported membrane configuration experiences many challenges in modeling physical—chemical properties of biological membranes because of the coupling of the bilayer to the rigid substrate surface. This interface suppresses thermally activated membrane fluctuations, limits the incorporation of membrane proteins, induces substrate-electrostatics-driven asymmetry, and hinders the translational mobilities and phase equilibration in bilayers. The article by Tanaka emphasizes these issues and offers a strategy to cushion the membrane–substrate interface via ultrathin polymer supports. He further demonstrates how the use of intercalated polymers may offer a materials route to modeling the generic role of the extracellular matrix and glycocalyx. The term glycocalyx refers to a variety of glycosylated proteins, glycolipids, and polymeric structures that often terminate cell surfaces.

An important structural feature of lipid membranes in biology is their ability to bend. There is a growing recognition that membrane curvature is an active regulator of membrane microphase separation and a concentrator of important biological functions. Sasaki and Stevens emphasize the issue, survey a physical-science-based understanding of the formation of these submembrane structural features, and highlight associated materials issues.

The perspective by Zhang and Granick illustrates the importance of advanced characterization methods in characterizing the lateral dynamics in supported membranes. A long-standing issue, one with many inconsistencies, relates to the issue of complex translational dynamics. Structural, chemical, and phase heterogeneities within membranes exist both statically and dynamically (such as in response to external stimuli, e.g., protein binding). Moreover, these heterogeneities often emerge independently in the two leaflets of the bilayer architecture. Complex dynamics attending such asymmetries requires advanced characterization tools capable of measuring molecular diffusivities at a range of length and time scales, also discriminating the two leaflets.

The article by Wu et al. is focused on the permeability characteristics of cellular membranes. Using supported membranes and lipid monolayers, the authors illustrate how synthetic molecules (e.g., triblock copolymers) can be used to repair damaged membranes. They demonstrate the ability of a class of triblock copolymers to selectively incorporate within damaged or void regions of the membranes. Their study further reveals how cells might rid the polymer from their membranes once the structural integrity of the membrane is restored.

Another long-standing issue in solid-supported lipid bilayers that continues to hamper their practical application in sensing and detection relates to their long-term stability. As Daniel et al. correctly point out, "they lack the resiliency to withstand air exposure and the thermal and mechanical stresses associated with device transport, storage, and continuous use over long periods of time." Their article summarizes several new strategies

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that mimic cellular cytoskeletons, thus enhancing bilayer stability.

The article by Fang et al. summarizes the current state of biomembrane technologies that enable the preservation and parallel display of functionally active membrane targets. This ability promises to deliver much-needed membrane-mimetic platforms for screening potential drug compounds against their membrane targets (e.g., G-protein coupled receptors, or GPCRs—a pervasive family of transmembrane proteins, targeted by 40–50% drugs, that transduce a host of extracellular ligand-binding signals into intracellular signals). The overwhelming role of membrane proteins as pharmacological targets put this issue at the center of this technology development.

The collection of review/perspective articles presented in this issue of *MRS Bulletin* represents a growing body of materials work targeted at understanding and controlling lipid membranes, largely in synthetic settings. These are complex systems that present many challenges—many of which are highlighted within these reviews. Complex as the issues are, there is plenty of room for optimism. Supported membranes have proved remarkably successful in a number of applications, including as a means of creating surrogate cell surfaces for interactions with other living cells. Most visually evocative among these implementations, perhaps, are the immunological synapses that have been induced to form between living T cells and supported membranes.[11,12]

Building on this methodology, the supported membrane configuration provides an avenue to introduce patterns of external stimuli—including inorganic nanostructures, surface patterns, signaling molecules, and oxidative stresses—to cells using their own receptors as liaisons (see Figure 2). These abilities offer promise toward understanding molecular interactions that characterize cellular homeostasis and disease. Several of these exciting applications are already paying dividends, and with greater development of the system from a controlled materials perspective, we can anticipate much more yet to come.

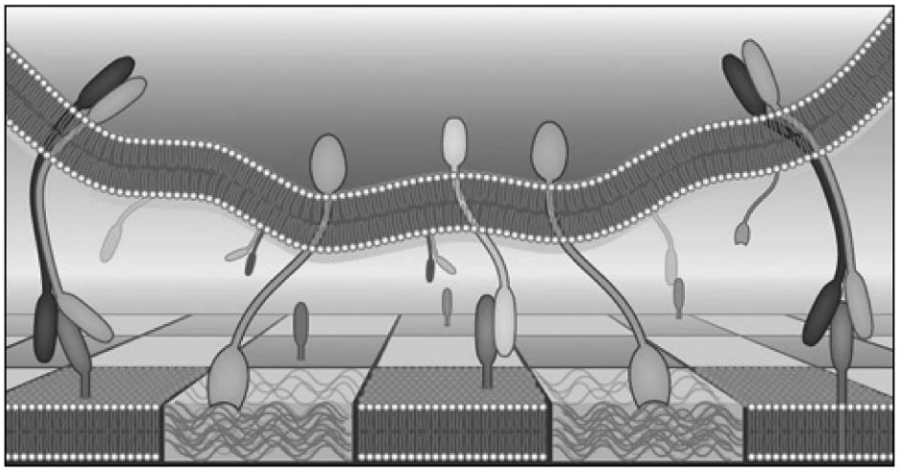

Figure 2. Schematic image of a living cell (at top) interacting with a supported-membrane-coated synthetic surface (at bottom). Natural protein signaling molecules may be incorporated into the synthetic membrane, providing specific functionalities with which the living cell may interact.

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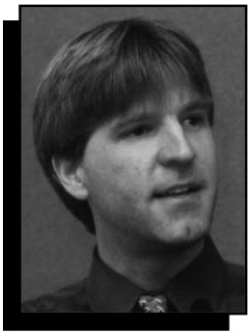

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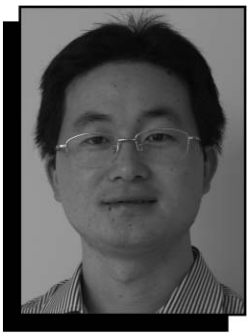

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