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REVIEW

Interfacial mechano-nanoarchitectonics for chemical, materials and biological processes

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The recent surge of interest in mechanochemistry and mechanobiology indicates a convergence of historical mechanical processes with contemporary nanoprocesses. This review explores the interfacial mechano-nanoarchitectonics in chemical, materials, and biological processes. Active research is being conducted on mechanochemistry, which is more closely related to controlling functional materials through regulating surface structure and property. Advances in probe microscopy have enabled mechanochemistry researchers to analyze various nanoscale phenomena in conjunction with the application of mechanical stimuli. As demonstrated by numerous examples in the field of mechanobiology, the mechanical properties of basic interfaces have the capacity to regulate sophisticated biological properties by coupling mechanical effects from surfaces. This assertion is applicable to a broad spectrum of targets, ranging from the regulation of cell differentiation to the comprehension of disease mechanisms. In addition, the air-water interface is an optimal location for the mechanical adjustment of nanostructures and molecular structures. The mechanical processes occurring at this interface are characterized by their high efficiency and are driven by delicate forces analogous to those observed in biological systems. The review concludes that interfaces are essential for combining mechanical manipulation as the most traditional method in materials processing with the cutting-edge methodology of nanoarchitectonics.

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1. Introduction: Nanoarchitectonics with mechano-process

Nanoarchitectonics is a broad concept involving the construction of functional molecules from nanounits, such as atoms and molecules. It incorporates various methods.[1] Functional materials are constructed by combining techniques such as atomic and molecular manipulation, chemical reactions, physical transformations of materials, self-assembly and self-organization, and the use of external forces and fields for orientation and organization. Engineering techniques such as microfabrication and nanofabrication are also employed, as well as biochemical processes. Unlike self-assembly, which often uses a single equilibrium process, nanoarchitectonics is better suited for creating hierarchical and asymmetric structures.[2] Because the concept is comprehensive and not particularly restrictive, nanoarchitectonics can be used with many materials and for a wide range of applications. Since all materials are essentially composed of atoms and molecules, nanoarchitectonics, which builds materials from these components, can be said to apply to all materials. If the ultimate theory of physics is the theory of everything,[3] then nanoarchitectonics could be considered a method for everything in materials science.[4] Traditional materials science has been eclectic and has not always succeeded in creating desired substances. By infusing nanotechnology into this field and establishing the concept of nanoarchitectonics—building materials from atoms and molecules—it is expected that all substances can be rationally constructed. Nanoarchitectonics is not entirely independent from materials chemistry; rather, it can be seen as the means to fulfill the dreams of materials chemistry.

Humanity faces many challenges, including energy depletion, environmental pollution, and biomedical issues. We are developing science and technology solution to these problems. While some contributions have been made even in cyberspace with information technology and artificial intelligence, the key to solving these problems lies in developing existing functional materials that can address them. Considerable effort has been devoted to developing materials that generate and store energy,[5] materials that can detect and remove environmental pollutants,[6] and materials that can treat various medical problems.[7] One could argue that the history of human development is linked to the progress of materials science.

In the 20th century, various fields of material chemistry developed and became systematized. While it is not the purpose of this review to address each individual development in material chemistry, such progress and systematization can be understood from objective facts. This is evidenced by the fact that several research fields have emerged by being named and having their objectives clearly defined. This enabled the rational

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creation of materials. These efforts continue today, with inorganic[8] and organic[9] substances, hybrids and composites,[10] polymers,[11] supramolecules,[12] and bio-related materials[13] being created as functional materials. It was recognized during this time that controlling the nanostructure of materials is important for improving their functionality. This trend was spurred by the development of nanotechnology. Nanotechnology enables observation[14] and manipulation[15] of structures and phenomena at the atomic and molecular levels. Nanotechnology has also made it possible to analyse and understand phenomena in nano-sized reagions.[16]

The next step is to combine materials chemistry, which can synthesize materials, with nanotechnology, which can analyse nanostructures. This combination will lead to the development of functional materials that take advantage of nanostructures. Similar efforts are being made in fields such as supramolecular chemistry to create functional materials tailored to the characteristics of nanostructures.[17] However, much of the research is being carried out independently in each field. This differs from the situation during the emergence of nanotechnology, when significant unified advances were made in nanoscience and technology. A comprehensive, holistic concept for assembling functional materials using nano-knowledge is needed. Nanoarchitectonics fulfils this role.[18] It is a methodology for constructing functional materials using atoms, molecules, and nanomaterials.[19] As Richard Feynman pioneered nanotechnology in the mid-20th century,[20] Masakazu Aono proposed nanoarchitectonics during the transition from the 20th to the 21st century.[21] Nanoarchitectonics is considered a post-nanotechnology concept.[22]

One particularly interesting area among recent advances in science and technology is the application of mechanical processes that were widely used in the early days of humankind to cutting-edge science. Mechanochemistry[23] and mechanobiology[24] are at the forefront of this field. Many mechanical processes involve macroscopic displacements. However, cutting-edge science focuses on controlling even more delicate structures and phenomena at the micro- and nanoscales. Nanoarchitectonics techniques also involve mechanical processes, such as the orientation and organization of materials induced by external forces or fields and various fabrication techniques. Advances in materials science have led to materials that respond to light with specific energy or whose properties change based on redox processes. Nevertheless, more basic mechanical stimuli also offer significant advantages. This becomes clear when considering the stimulus response systems of organic substances. Photoisomerization applies only to substances possessing a chromophore group that absorbs specific light. Substances responding to electronic stimuli do so via their participating redox-active functional groups. In contrast, some degree of mechanical deformation can occur in all substances, albeit to varying extents. Materials that respond to optical or electrical stimuli are generally limited to those with receptor sites for those stimuli. In contrast, almost all materials can respond to mechanical stimuli. In terms of universality and applicability, controlling material structure and function through mechanical processes has an advantage.

On the other hand, optical and electrochemical stimuli have their own advantages. These stimuli can be transmitted remotely via units such as photons or electrons. In contrast, with the exception of ultrasound-based methods, mechanical processes require direct contact transmission of stimuli. Given this characteristic, controlling interfacial contact and phenomena is crucial for mechano-processes. Chemical reactions and structure formation mediated by interfacial phenomena are key to mechanochemistry. Furthermore, many mechanobiological studies discuss controlling bioprocesses by transmitting mechanical stimuli from interfaces. Taking a comprehensive approach to controlling nanostructures and regulating their functions, including both chemistry and biology, leads to the concept of mechano-nanoarchitectonics. Interfacial phenomena are also important here. Mechanical processes are fundamentally driven by macroscopic displacements. The desired functional and structural control occurs at the nanoscale or mesoscale. Interfaces connect these two scales. This review particularly emphasizes the importance of interfaces in mechanochemical processes. However, I would like to briefly comment on this point. The emphasis on surfaces stems mainly from the fact that they provide a convenient method for applying force when solid-state interactions are involved. Nevertheless, other systems exist. For instance, numerous examples of mechanochemical reactions conducted under hydrostatic pressure are available. It can be argued that intermolecular interactions can also be approached mechanochemically. While intermolecular interactions are typically considered from the perspective of interaction energy, the fundamental driving force is actually the energy gradient—the force that drives these interactions—which is inherently mechanochemical. From this viewpoint, this review can be positioned as highlighting the aspects where interfaces significantly contribute to mechanochemical processes.

From this perspective, this review discusses interfacial mechano-nanoarchitectonics in chemical, materials and biological processes (Figure 1). First, this review article will introduce examples of chemical and biological processes involving mechanical processes at interfaces. Through these examples, the role of interfaces in mechanical processes will be re-examined. The next section will discuss methodologies for rationally coupling macroscopic mechano-processes with nano-phenomena using liquid interfaces, mainly air-water interface. The review concludes that interfaces are essential for combining mechanical manipulation, the most traditional method in materials processing, with the cutting-edge

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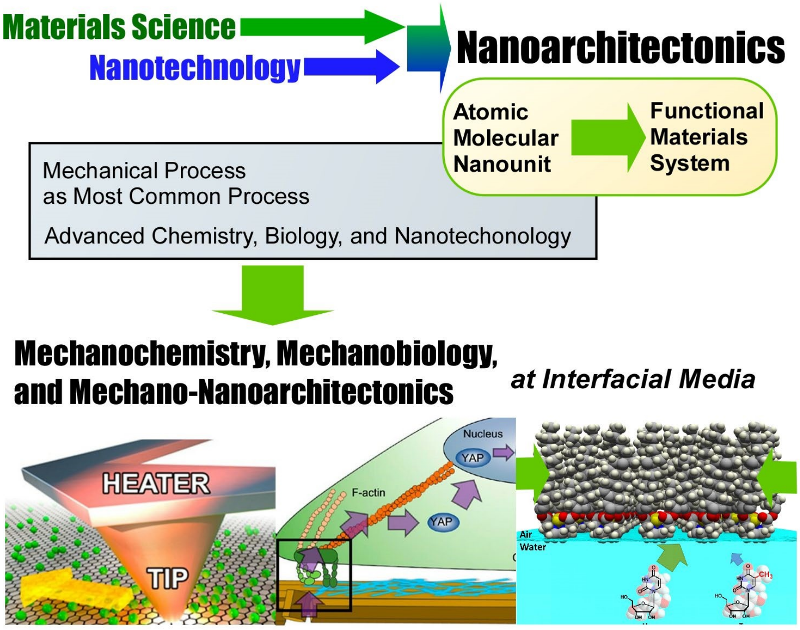

Figure 1. Outline of nanoarchitectonics concept (top) and targets of this review article (bottom). Reprinted with permission from Ref. 26 Copyright 2017 American Chemical Society. Reproduced under terms of the CC-BY license from Ref. 31, 2020 American Chemical Society.

methodology of nanoarchitectonics. Nanoarchitectonics is a growing concept. Rather than providing a sharp definition, it is a useful concept for enhancing the field's comprehensiveness. Therefore, comprehensively considering many examples as nanoarchitectonics can sometimes reveal a unified concept or significance. This means that by considering things that cannot be overtly labelled as nanoarchitectonics from the perspective of assembling matter, important aspects become apparent. This review aims to provide such an opportunity.

2. Interfacial mechano-process in chemical and materials science

Several examples illustrate the importance of examining and interpreting surface phenomena to understand mechanochemistry. This section collects and illustrates some recent examples of such studies.

The rate of mechanochemical reactions typically increases exponentially with applied stress. Therefore, the orientation of functional groups relative to the surface is an important factor. Tysoe and his co-workers analysed the effect of normal stress on adsorbates oriented perpendicular to the reacting surface.[25] They showed that a critical stress is required to initiate mechanochemical reactions. Actually, they investigated this using density functional theory calculations to simulate the effect of compressing a homologous series of alkylthiolate species on a hydrogen-terminated copper surface. A critical stress is necessary to decompose thiolates with perpendicular C–CH_3 bonds. In contrast, no critical stress is required for thiolates with nearly horizontal C–CH_3 bonds. These predictions were verified by measuring the mechanochemical reaction rates of these alkylthiolates on a Cu(100) substrate using an atomic force microscope (AFM). For ethyl thiolate, where the alkyl group ends are nearly parallel to the surface, no hysteresis was observed in the approach/retraction curves. This confirms that mechanochemical activity does not exhibit a critical stress. However, propyl thiolate exhibits critical behaviour, and the critical process is accompanied by a structural change due to normal stress-induced rotation around the C–C bond. These results suggest that critical phenomena can be caused by either a structural change or a transformation. These results provide an example that can be used to elucidate the details of mechanochemical reaction mechanisms at surfaces. This methodology may also provide insight into the origin of critical phenomena in general stress-induced processes. A more fundamental understanding of these effects could lead to the design of ultra-mechanically stable surface films.

Understanding mechanochemistry and surface engineering requires driving and measuring nanoscale chemical reactions at surfaces. To advance this goal, Felts and his co-workers developed an AFM technique that can measure nanoscale surface reactions triggered by multiple external stimuli (Figure 2).[26] Using this technique, they measured the local reduction of graphene oxide as a function of temperature and mechanical force at the contact point. By independently tuning the force and temperature of a heated atomic force microscope probe, they measured the kinetics and thermodynamics of oxygen group desorption from graphene oxide in air. They used a two-step routine involving driving scans that cleave functional groups at elevated temperatures and/or high forces, followed by measurement scans at room temperature and low forces. This enabled in situ characterization of the changing surface composition and a quantitative understanding of the locally

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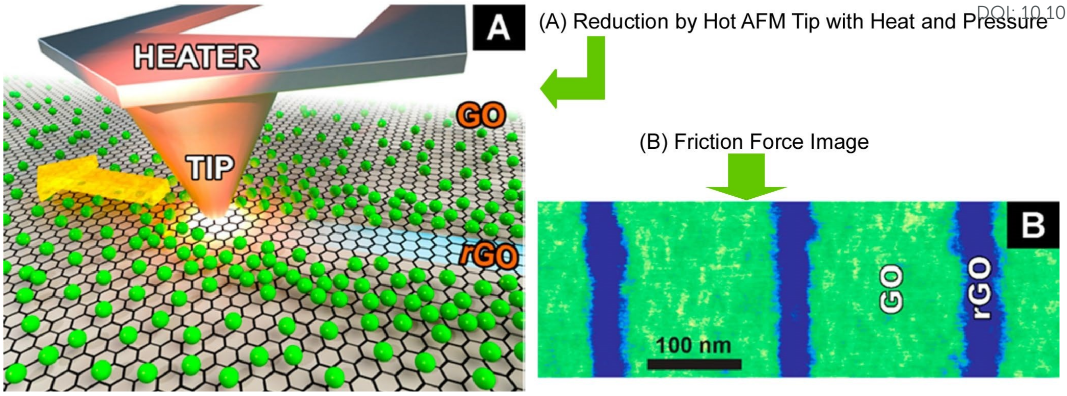

Figure 2. AFM technique for evaluation of nanoscale surface reactions triggered by multiple external stimuli: (A) reduction of graphene oxide by hot AFM tip with heat and pressure; (B) the resulting friction force image. Reprinted with permission from Ref. 26 Copyright 2017 American Chemical Society.

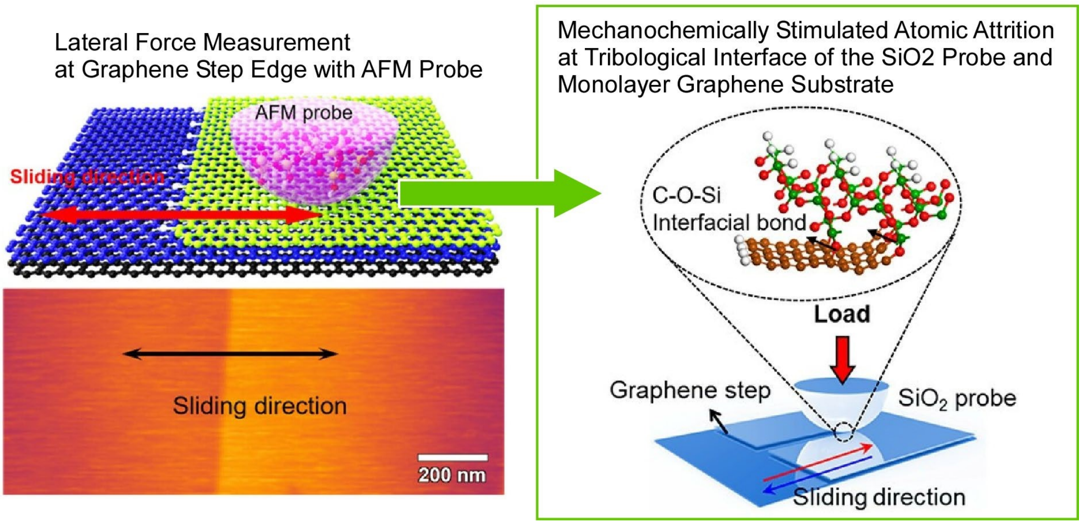

Figure 3. Wear sensitivity at the atomic step edge of a graphene monolayer: lateral force measurement at graphene step edge with AFM probe (left) and mechanically stimulated atomic attrition at tribological Interface of the SiO_2 Probe and monolayer graphene substrate (right). Reprinted with permission from Ref. 27 Copyright 2024 American Chemical Society.

driven chemical reactions at surfaces. They monitored the resulting changes in relative friction and found that, for example, oxygenated graphene exhibited friction levels 2–9 times higher than pristine graphene. This technique is useful for understanding the nanoscale surface chemistry of two-dimensional (2D) materials that respond to various external stimuli, including mechanochemistry. By varying the tip material, the functional groups to be cleaved, the environmental composition, and the substrate material, one can obtain the chemical kinetics of virtually any frictional interface. Appropriately passivating the tip material makes it possible to observe reactions on the tip without significantly altering the reaction pathway. This makes the technique ideal for studying the behaviour of real mechanochemical systems in an environment that closely resembles their operational environment.

Due to their unique mechanical and electronic properties, graphene-based materials have been widely used as wear-resistant protective coatings for micro- and nanodevices and as sensor channel materials. However, severe wear resulting from strong chemical interactions and mechanochemical reactions at the interface poses a significant challenge for moving parts. A detailed understanding of this phenomenon is necessary. Xiao, Chen, and their co-workers demonstrated that wear sensitivity at the atomic step edge of a graphene monolayer is dominated by the mechanochemistry of the frictional interface (Figure 3).[27] When chemically inert diamond is used, only mechanical damage, such as surface fracture and folding, occurs. In contrast, mechanochemical reactions activated through chemically active SiO_2 microspheres result in atomic wear. In the atomic wear mechanism induced by mechanochemical stimulation at the interface between an SiO_2 probe and a monolayer of graphene, thermally activated C–O–Si bond formation, promoted by mechanical stress during contact, is thought to lower the energy barrier for C–C bond scission. This study shows that shear-induced atomic wear of graphene step edges varies

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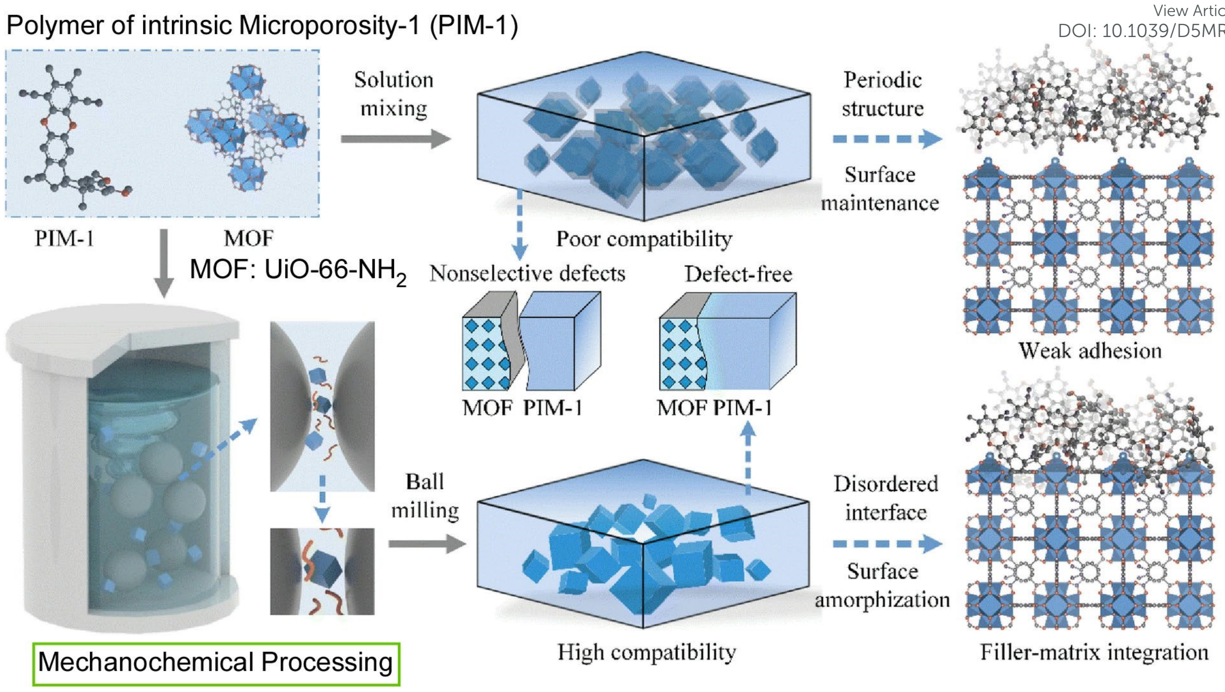

Figure 4. Preparation of interface-integrated mixed matrix membranes based on metal-organic frameworks (MOFs) and microporous polymers, achieving efficient gas separation: conventional solution mixing (top) and mechanochemical processing (bottom). Reprinted with permission from Ref. 28 Copyright 2025 Royal Society of Chemistry.

significantly with the chemical activity of the contact surfaces. This suggests that improving the wear resistance of graphene-based materials may require reducing mechanochemical interactions at the friction interface. Careful consideration of the surface chemistry of tribological materials and control of surface mechanochemistry will be crucial when using layered graphene materials in nanoscale devices.

Controlling mechanochemistry at interfaces between materials improves their performance, not just at nanoscopic surfaces, as observed with probe microscopes. For example, Qiao, Li, and their co-workers used mechanochemistry to create interface-integrated mixed matrix membranes based on metal-organic frameworks (MOFs) and microporous polymers, achieving efficient gas separation (Figure 4).[28] Mixed matrix membranes overcome the limitations of polymer and nanoporous membranes and are expected to enable energy-efficient material separation. Characterization and molecular dynamics simulations revealed that ball milling significantly improved the dispersion of the filler, interfacial compatibility, separation performance, and aging resistance of the mixed matrix membranes. MOF fillers dispersed well in the mixed matrix membrane, and polymer chains firmly attached to the filler surface through simple ball milling. Molecular dynamics simulations were used to investigate the effect of coordination bond cleavage on filler-matrix interactions. The simulations demonstrated that mechanochemical treatment disrupts the crystalline periodicity of the filler surface, forming a disordered interface and promoting the intercalation of polymers within the filler and the integration of the filler and the matrix. The prepared mixed matrix membranes exhibited significantly improved filler dispersion and interfacial compatibility. This significantly improved the separation performance of binary and ternary gas mixtures, surpassing that of state-of-the-art membranes. For instance, they demonstrated high CO_2/N_2 selectivity for gas separation. Taking interfacial processes into account, mechanochemical processing may be an alternative approach to achieving desirable properties and maximizing the performance of hybrid membranes.

Although there is a wealth of research on mechanochemistry, this section just presents examples that emphasize interfacial phenomena. Advances in probe microscopy technology have made it possible to analyse various nano-level phenomena in conjunction with the application of mechanical stimuli. These technological advances link mechanochemistry, interfaces, and nanotechnology. Even without nano-specific analysis, it is evident that interfacial phenomena are essential to mechanochemistry and its resulting functions. Many physical properties are determined by structures such as particle boundaries, and mechanochemical control of surface structure and state is reflected in material functions. The examples provided are just a few and are not necessarily representative. However, they demonstrate the importance of interfaces in mechanochemistry. Controlling interfaces is essential for adjusting and modifying nanostructures and their resulting physical properties.

3. Interfacial mechano-process in biology

Interfacial phenomena are important in mechanobiology as well as mechanochemistry. Mechanobiology controls biological

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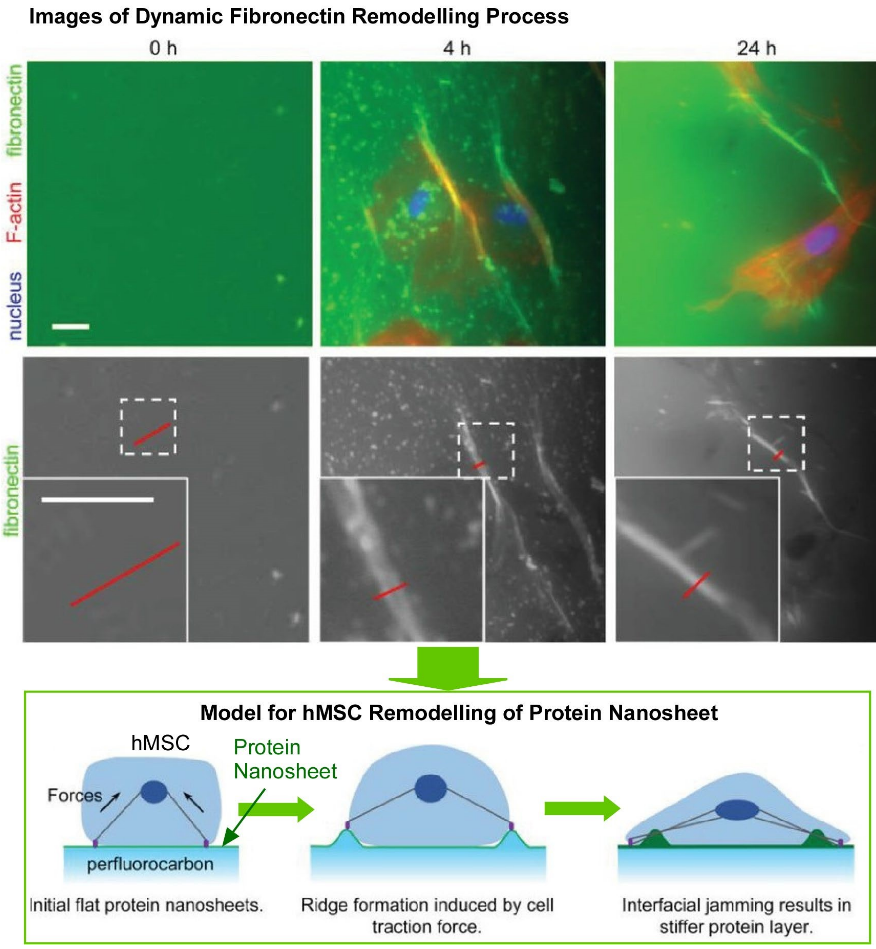

Figure 5. Behaviours of human mesenchymal stem cells (hMSCs) at liquid-liquid interface between a protein-containing aqueous phase and a perfluorocarbon: images of dynamic fibronectin remodelling process (top) and model for hMSC remodelling of protein nanosheet (bottom). Reprinted with permission from Ref. 29 Copyright 2020 Wiley-VCH.

phenomena through mechanical perturbations. What role does the interface play? One could argue that roles of interfaces in mechanobiology are more important than those for mechanochemistry. Within cells, substances and signals are transmitted efficiently, and stimuli on the cell surface are transmitted to the cell's interior. In other words, interfacial phenomena can trigger advanced biological processes within cells. This section provides examples of how mechanical phenomena at interfaces control biological functions.

Interest in developing dynamically adaptive biomaterials to control cellular functions at interfaces through mechanobiology is growing. For example, Jia et al. controlled the differentiation of human mesenchymal stem cells (hMSCs) using a liquid-liquid interface between a protein-containing aqueous phase and a perfluorocarbon (Figure 5).[29] Protein monolayer nanosheets that mimic the extracellular matrix (ECM) were formed at the interface and functioned as adaptive materials that can be tuned to adapt dynamically to cellular traction forces. Ultrastructural changes from the protein monolayer to hierarchical fibers occurred due to interaction with hMSCs at the interface. In order to resist cellular traction forces, the protein nanosheets deformed, generating elastic forces and leading to the formation of protuberances. As the cells gradually spread, focal adhesion turnover occurred, accompanied by the formation and disassembly of focal adhesions. Subsequently, the protein aggregates compressed, resulting in the appearance of microwrinkles as the aggregates packed together. This triggered the spatial rearrangement of proteins, which propagated the stimuli within the cells and resulted in feedback for changes in the fate of stem cell. Elongated fibronectin fibers promote the formation of extended focal adhesion structures, enhance focal adhesion kinase activation, and promote neural differentiation of stem cells. These results help elucidate the feedback mechanisms linking ECM dynamics, biological signalling, and long-term stem cell fate. They pave the way for mechanobiology applications at liquid interfaces in tissue engineering and regenerative medicine.

Jia et al. reported an adaptive biomaterial based on a two-dimensional network of protein nanofibrils at a liquid-liquid interface by culturing hMSCs at a similar liquid-liquid interface between an aqueous phase and a perfluorocarbon (Figure 6).[30] This material promoted hMSC neural differentiation more effectively than flat protein nanosheets via focal adhesion kinase (FAK)-mediated signalling. The work also discovered that

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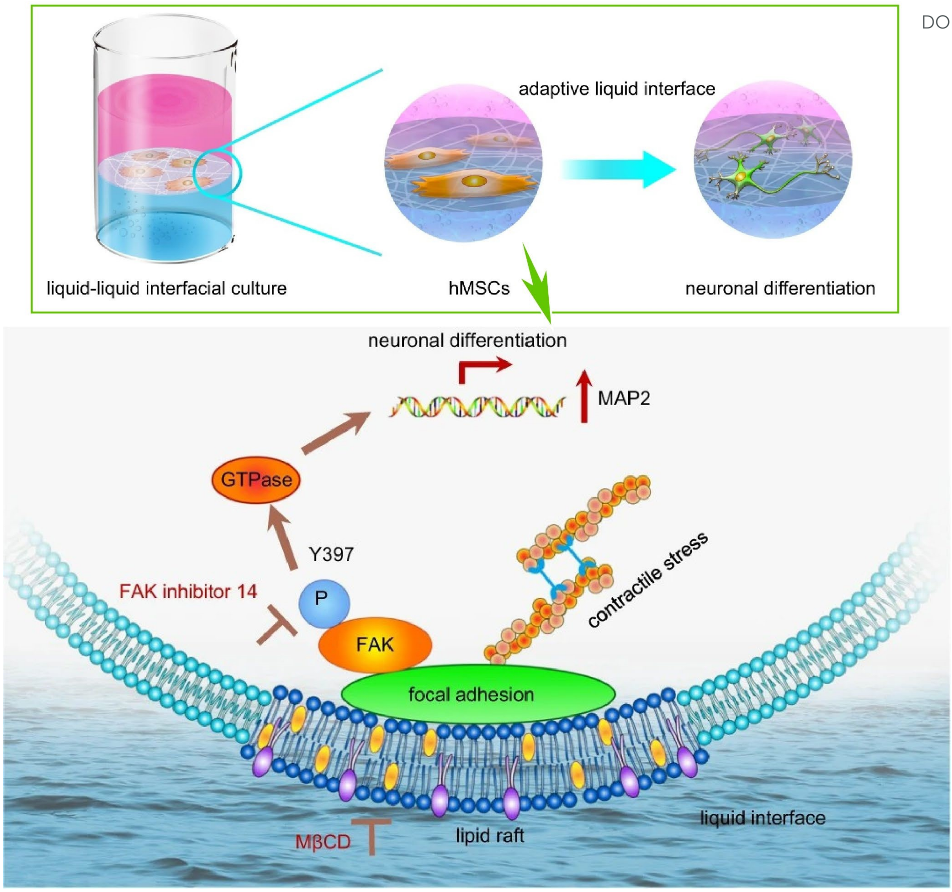

Figure 6. Neuronal differentiation of hMSCs at the interfacially assembled two-dimensional network of protein nanofibrils (top) and adaptive liquid interface inducing neural differentiation of hMSCs via lipid raft assembly and FAK phosphorylation (bottom). Reproduced under terms of the CC-BY license from Ref. 30, 2022 Springer-Nature.

lipid raft microdomains play a pivotal role in the initial adhesion of cells and their subsequent neural differentiation. Lipid rafts internalize and recruit cell adhesion molecules to various cell membranes. They also function as concentration platforms, inducing the integration of large signalling complexes. These functions enable cells to rapidly adapt to constantly changing microenvironments. In this context, FAK is one of the key mechanosensors at adaptive liquid interfaces. The spatial and temporal regulation of FAK phosphorylation is essential for hMSC neural differentiation. This discovery provides new insights into the fundamentals of dynamic cell-ECM interactions and a deeper understanding of the biophysics underlying mechano-transduction. Furthermore, incorporating bioactive proteins and responsive polymers has the potential to lead to further advancements. Liquid interfaces may enable the design of adaptable biomaterials with applications in regenerative medicine and tissue engineering that were previously unimaginable.

Developing hMSC-based therapeutics is crucial, yet a fundamental problem remains. That is the shortage of adult stem cells. Technology capable of maintaining pluripotency and stem cell-like phenotypes over long periods during large-scale in vitro expansion is necessary. Song et al. developed a differentiation-free hMSC expansion technique that uses the mechanical effects of solid surfaces.[31] In this method, hMSCs are controlled using highly aligned fullerene nanowhisker (FNW) nanopatterned scaffolds (Figure 7). The FNW surface arrays were fabricated using a simple Langmuir-Blodgett (LB) technique. Protein nanopatterns were adsorbed onto these arrays, and cell spreading occurred along the long axis of the FNWs. This resulted in more uniform biophysical signals and activation of the hMSC mechano-transduction process. These effects led to the uniform elongation and strong orientation of hMSCs. They also influenced histone acetylation and methylation patterns and induced a self-renewal pattern in the hMSC nuclei. Cells cultured on nanopatterned surfaces of highly aligned FNWs maintain long-term pluripotency and improved regenerative capacity through appropriate mechanical cell contractility and nuclear localization of Yes-related proteins. The LB method and its simplified variants are relatively simple and can be easily adopted in biomedical laboratories. This could facilitate the production of centimeter-scale nanotopography substrates for large-scale hMSC expansion in clinical settings

Understanding mechanobiological phenomena requires sensing the mechanical state of surfaces. However, there is a lack of documentation on molecular probes for measuring compressive forces at surfaces, which pose a challenge for mechanobiological research. Salaita and co-workers developed

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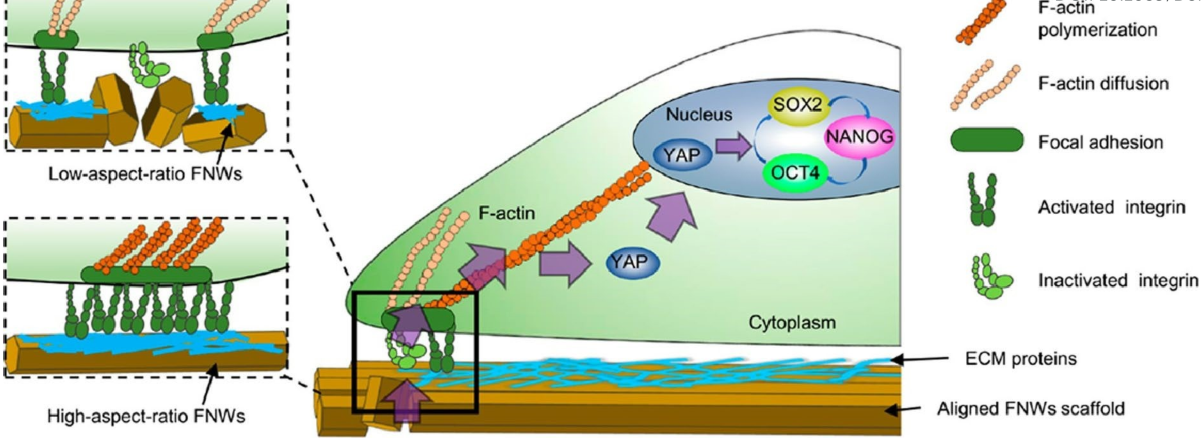

Figure 7. fullerene nanowhiskers (FNWs) nanopatterned scaffolds control the hMSC self-renewal process by mechanotransduction. Reproduced under terms of the CC-BY license from Ref. 31, 2020 American Chemical Society.

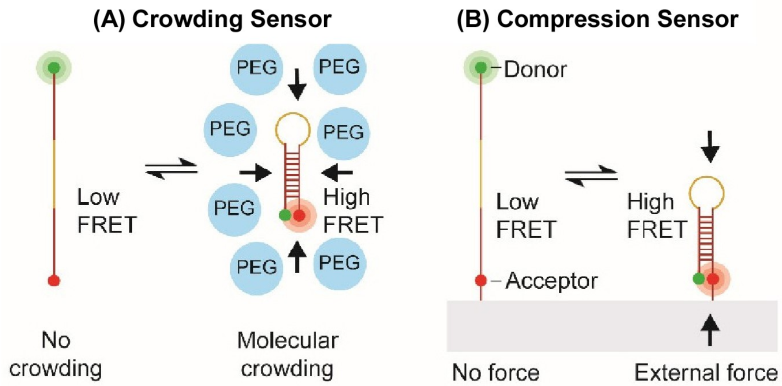

Figure 8. Molecular compression reporter using pseudostable hairpins of pseudostable DNA structure through Förster resonance energy transfer (FRET): (A) crowding sensor; (B) compression sensor. Reproduced under terms of the CC-BY license from Ref. 32, 2024 American Chemical Society.

a novel molecular compression reporter using pseudostable hairpins (Figure 8).[32] This probe is based on a pseudostable DNA structure that folds in response to external compressive forces. Using Förster resonance energy transfer (FRET), this work quantified hairpin folding as a function of temperature and molecular crowding. This method applied the principle of molecular crowding, in which the entropic degrees of freedom of a biomolecule or polymer are reduced by isotropic concentration, to an interfacial molecular compression sensor. This provides a powerful tool for mapping compressive forces in biological systems. Indeed, the analysis mapped the compressive forces generated by primary naive T cells. These forces are generated by cytoskeletal protrusions triggered by actin myosin activity. This study may provide important information for developing tools to study the mechanical forces exerted by cells. It has the potential to improve our understanding of not only T cell mechano-transduction, but also a wide range of biological processes.

Mechanical stimuli, such as surface vibration, are useful for understanding the mechanobiology of biological subjects, such as bacteria. Although mechanical stimuli have been used to control mechano-dependent behaviour in mammalian cells, a similar level of understanding is lacking in bacteria. Surface adhesion, an early stage in biofilm formation and surface biofouling, is mechanically dependent and therefore a potential target for mechano-control. Mendes and co-workers mechanically stimulated bacteria using nanometer-scale surface vibrations and investigated their effects on adhesion (Figure 9).[33] They found that nanoscale vibration stimulation altered the cell membrane potential and reduced surface adhesion consistently. Nanovibration stimulation with picoNewton intensity depolarized cells on surfaces, thereby inhibiting the adhesion of highly polarized cells. This disrupted the reversible adhesion dynamics of cells, suppressing surface adhesion, sessile transition, and biofilm formation. These findings suggest the potential benefits of using mechanobiology to control bacterial behaviour. They pave the way for controlling bacterial behaviour through nanometer-scale mechanical vibrations at interfaces.

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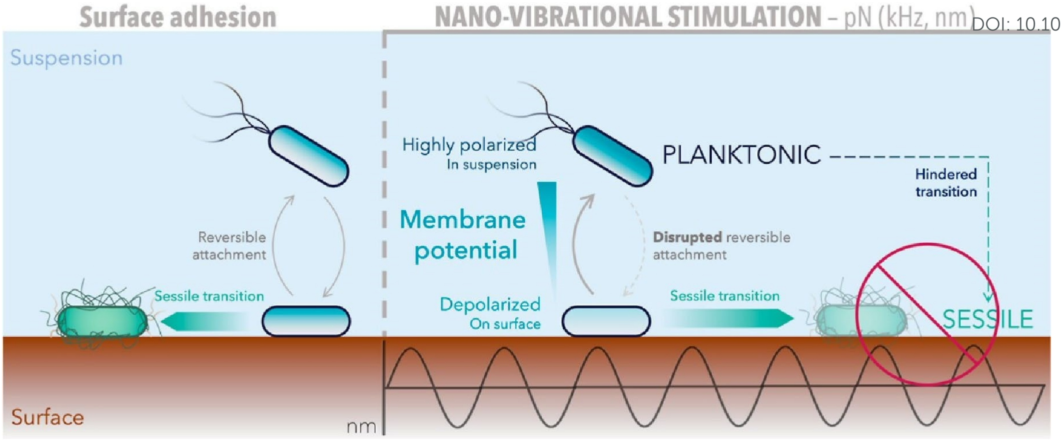

Figure 9. Effect of nanovibrational stimulation on E. coli surface adhesion and sessile transition: surface adhesion scheme (left) and effect of nanovibrational stimulation (right). Reproduced under terms of the CC-BY license from Ref. 33, 2024 American Chemical Society.

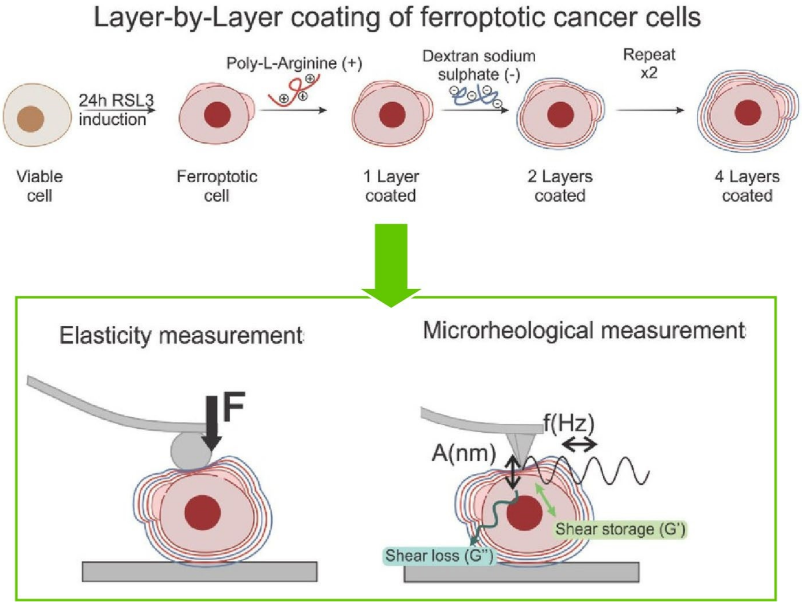

Figure 10. Nanocoating of a cell using layer-by-layer (LbL) assembly for mechanical modulation: LbL coating of ferroptotic cancer cell (top), elasticity measurement (bottom left), and microrheological measurement (bottom right). Reprinted with permission from Ref. 34 Copyright 2023 Wiley-VCH.

Removing dead cells is important for maintaining normal tissue homeostasis and regulating immune responses. Surface mechanobiological properties are key to this process. Young's modulus decreases in cancer cells undergoing ferroptosis (a type of programmed cell death dependent on iron). Van der Meeren, Skirtach, and their co-workers nanocoated cells using layer-by-layer (LbL) assembly to modulate Young's modulus and study its effects (Figure 10).[34] This study aimed to analyse how the mechanobiology of ferroptotic cancer cells affects the efficiency of efferocytosis. With LbL coating technology, ferroptotic cells' mechanical properties can be altered by increasing their Young's modulus and decreasing loss tangent. They demonstrated that an increase in Young's modulus primarily leads to increased efferocytosis (the process by which apoptotic cells are removed by phagocytic cells). An increase in Young's modulus, which depends on the number of LbL layers and is due to surface nanoarchitectonics, results in enhanced efferocytosis by primary macrophages. This study demonstrates the important role of dead cell mechanobiology in regulating macrophage efferocytosis and suggests the possibility of using dead cell surface coatings to co-deliver drugs, adjuvants, and other types of transporters.

To understand various biomedical problems, it is necessary to know how matrix stiffness gradients affect cellular behaviour and function at the cellular level. One example is the effect on mesenchymal stem cell (MSC) immunoregulation. For instance, in periodontitis, the strength of the stiffness gradient at the soft-hard tissue interface is significant. Zhang et al. investigated

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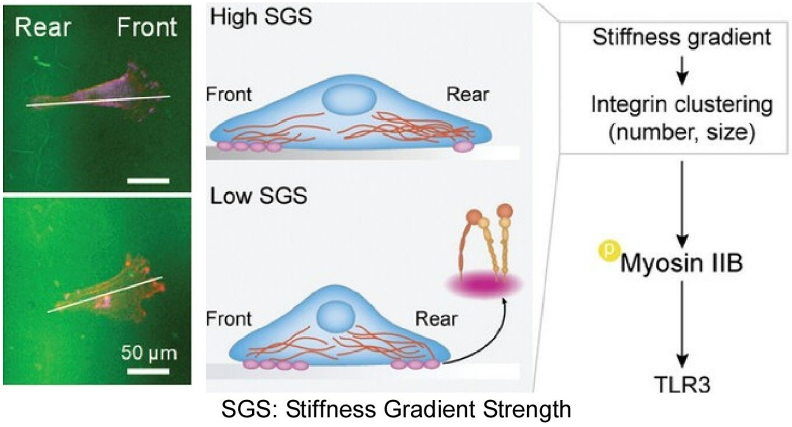

Figure 11. The features of the integrin β1 clusters at cellular front and rear(left and middle) and the schematic downstream pathway for regulation the immunophenotype of mesenchymal stem cell (MSC) (right) in the presence of the cellular-scale matrix stiffness gradient. Reprinted with permission from Ref. 35 Copyright 2024 Wiley-VCH.

how high and low stiffness gradient strength affect MSC immunoregulation in vitro (Figure 11).[35] They examined the microenvironment and immunophenotype of the stiffness gradient of MSCs at a typical soft-hard tissue interface in healthy individuals and patients with periodontitis. They found that patients with periodontitis exhibited lower stiffness gradient strength at the periodontal ligament-atherosclerotic junction compared with healthy individuals. This is primarily due to the lower mineral content in patients with periodontitis. These effects mechanically regulate the development of MSC immunophenotypes through cell polarization. The polarized localization of integrin β1 clusters, the activation of myosin IIB, and the remodelling of chromatin via nuclear mechano-transduction play a role in this process. Decreased cell polarization promotes an anti-inflammatory phenotype in response to low stiffness gradient strength at the cellular level. These results also have implications for the design of biomaterials for interfacial tissue engineering and regenerative medicine, in which mechanical factors regulate the development of MSC immune phenotypes via cell polarization.

This section presents several examples of mechanobiology driven by mechanical surface effects. The examples were chosen to demonstrate a variety of topics, ranging from fundamental phenomena to medical applications. They are not exhaustive or representative of the field. However, they demonstrate that the physicochemical and sometimes simple mechanical properties of interfaces can control advanced biological properties. This applies to a variety of targets, including controlling cell differentiation and disease mechanisms. This is due to the highly organized functional morphology of cells. Conversely, if such organized functional structures could be constructed using nanoarchitectonics, simple mechanical stimuli on surfaces could drive extremely complex functions. The above mechanobiology examples point the way toward achieving this.

4. Langmuir-Blodgett science as foundation of finer controls of mechano-nanoarchitectonics

The previous two sections presented examples of mechanical processes, such as mechanochemistry and mechanobiology, reflected in structure fabrication and function development. Controlling mechanical processes at interfaces has been shown to be key to these advances. Despite these commonalities, these systems are highly diverse, operating according to a wide variety of principles. Progress in research can easily be seen as specific. In this context, a significant scientific challenge is devising a comprehensive methodology that addresses all phenomena in a unified manner. This review focuses on the Langmuir-Blodgett (LB) technique[36] as a candidate for such a methodology, given its over 100-year history.[37] Specifically, this section argues that monolayers and ultrathin films at gas-liquid interfaces, primarily air-water interfaces, can serve as a foundation for the development of mechanochemistry, mechanobiology, and mechano-nanotechnology. It discusses the reasons for this, provides some promising research examples, and explores future prospects.

A thin film of amphiphiles spread at the air-water interface is called a Langmuir monolayer. This interface is ideal for forming various ultrathin film structures, including monolayers[38] and related thin films.[39] These thin films can be compressed or expanded laterally by macroscopic mechanical deformation. This allows control over nanostructures, such as the orientation and aggregation state of components within the thin film. In other words, a liquid interface is an ideal environment for coupling mechanical stimuli with nanostructural changes,[40] despite their significant size differences. Furthermore, the interface between heterogeneous media with widely differing dielectric constants is where materials with different solubility properties meet. It is also where molecular recognition[41] and the formation of supramolecular polymers[42] occur and where reactions[43] and the immobilization of biocomponents such as enzymes,[44] take place.

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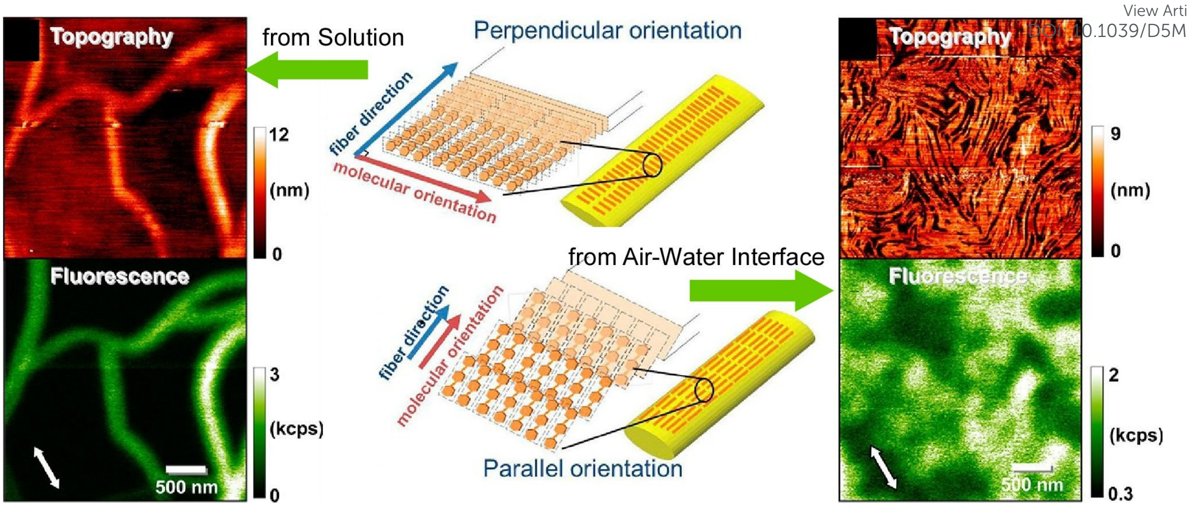

Figure 12. Aligned nanorods prepared formed by spreading oligo(p-phenylene vinylene) at the air-water interface (middle bottom and right images) and entangled nanofibers formed in its solution as gel fibers (middle top and left images) with near-field scanning optical microscopy observations. Reprinted with permission from Ref. 45 Copyright 2014 American Chemical Society.

This provides a rational platform for linking mechanical stimuli with nano-, molecular-, and bio-phenomena. Though the Langmuir-Blodgett (LB) method is a traditional technology with a history of over 100 years, it is also a field in which cutting-edge fields such as mechanochemistry, mechanobiology, and mechano-nanoarchitectonics can be developed. To explore its potential, the following section presents some examples.

The air-water interface allows for the nano-control of the internal structure of supramolecular assemblies. Sakakibara et al. reported that they formed aligned nanorods measuring 340 ± 120 nm in length and 34 ± 5 nm in width by spreading oligo(p-phenylene vinylene) at the air-water interface and mechanically compressing it (Figure 12).[45] These supramolecular assemblies differ significantly from the entangled nanofibers that form as gel fibers in solution. Near-field scanning optical microscopy observations revealed that changes in molecular orientation result in distinct excited-state characteristics upon localized photoexcitation in these two types of supramolecular assemblies. In entangled gel network fibers, which are typically formed in solution, individual molecules align perpendicular to the long axis of the fibers. This leads to long-range excitation energy transfer within the entangled fibers, resulting in significant fluorescence quenching. In contrast, the molecules in nanorods formed by compression at the air-water interface align parallel to the rods' long axis. This nanostructure exhibits significant fluorescence enhancement. Therefore, entangled gel fibers with perpendicularly aligned molecules are favourable for excitation energy transfer, while nanorods with parallel alignment are favourable for charge transport. These findings imply that the molecular organization and nanoscale morphology of self-assembled structures can be altered through mechanical processes at the air-water interface. Controlling excited-state properties could be useful for efficient sensing and imaging applications.

The mechanical action of vortex flow at the air-water interface can induce chiral nanoassemblies from achiral molecules and generate chiral properties. Maeda et al. demonstrated that vortex flow can induce and control the supramolecular chiral structure of aggregates consisting of achiral trans-bis(salicylaldiminato)Pt(II) complexes with hexadecyl chains (Figure 13).[46] The circularly polarized luminescence direction and intensity of these Pt(II) complex aggregates can be precisely tuned by adjusting vortex conditions such as rotation direction and flow velocity. Furthermore, an increase in vortex-induced luminescence was observed with an increase in vortex flow velocity. Circularly polarized luminescence has various applications that have been investigated by controlling its chirality and intensity. This mechanical methodology at the interface is expected to provide important insights into future technologies for forming functional luminescent materials.

Control the assembly of luminescent molecules can be attempted through mechanical processes at the air-water interface in order to achieve specific optical properties. For instance, materials that exhibit both mechano-luminescence and electroluminescence are useful for sensing and optoelectronics applications. Acharya and co-workers used 1,2-bis(4-(1-([1,1-biphenyl]4-yl)-2,2'-diphenylvinyl)phenyl)-1,2-diphenylethene as a luminescent molecule (Figure 14).[47] They obtained aggregation-induced emission (AIE)-active complexes by forming supramolecular spherical aggregates at the air-water interface. These molecules were found to have a highly twisted structure at the air-water interface. The phenyl rings promoted strong benzene-H bonding at the air-water interface, forming stable monolayers of the molecules. Furthermore, the AIE effect significantly enhanced the emission of the luminescent molecules in both monolayer and multilayer Langmuir-Blodgett (LB) films. The obtained results demonstrated the high-contrast, reversible mechano-chromic

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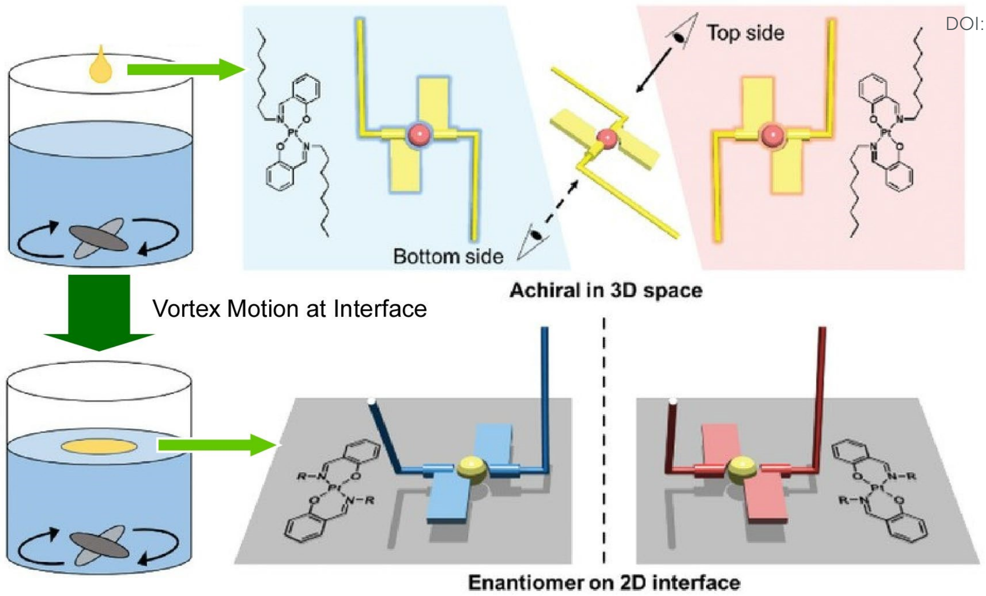

Figure 13. Control of supramolecular chiral structure of aggregates consisting of achiral trans-bis(salicylaldiminato)Pt(II) complexes with hexadecyl chains by vortex flow at the aur-water interface. Reprinted with permission from Ref. 46 Copyright 2022 Wiley-VCH.

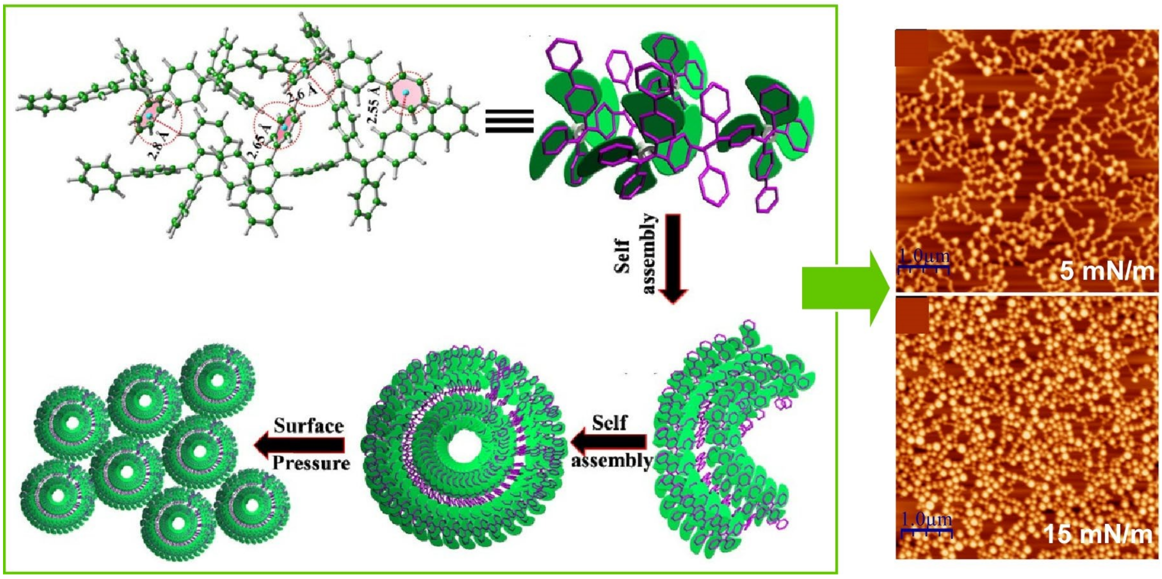

Figure 14. Formation of spherical aggregates of 1,2-bis(4-(1-([1,1'-biphenyl]-4-yl)-2,2-diphenylvinyl)phenyl)-1,2-diphenylethene at the air–water interface: plausible formation mechanisms (left) and AFM topography images of the spherical aggregates (right). Reprinted with permission from Ref. 47 Copyright 2018 American Chemical Society.

behaviour of the luminescent molecules where temperature acts as a mechanical stimulus that alters the molecular packing to switch photoluminescence. These findings underscore the importance of controlling the organization of AIE complexes in interfacial mechanical processes to optimize luminescent properties for efficient lighting and sensing applications.

As seen in the previous example, mechanical processes at the air-water interface can induce structural changes, such as molecular twisting. These changes can be used to drive molecular machines or optimize the structure of receptor molecules through macroscopic mechanical motion.[48] Figure 15A shows a steroid cyclophane operating as a molecular machine at the interface.[49] This molecular machine has a central cyclophane ring structure to which four steroid moieties are attached via flexible spacers. Specifically, one of the steroid moieties is a cholic acid unit with three hydroxyl groups on one side. Without pressure, the hydrophilic side of the cholic acid unit adheres to the water surface, adopting an open conformation. When pressure is applied by macroscopic compression of the monolayer, the molecular machine forms a three-dimensional cavity. Repeated compression and expansion, the process that changes sizes of the monolayer

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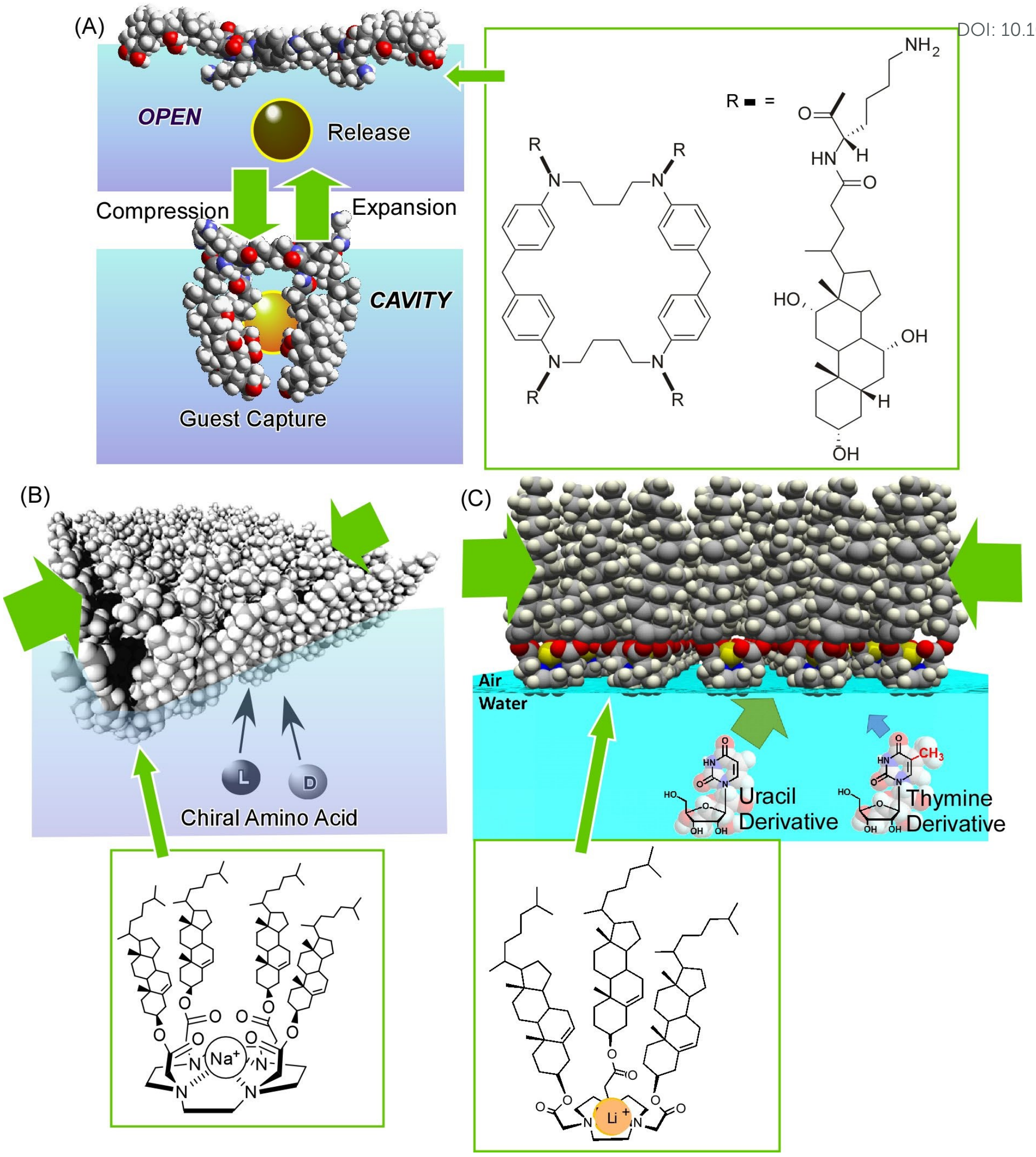

Figure 15. Controls of molecular machine and molecular receptor at the air-water interface: (A) guest capture by steroid cyclophane; (B) chiral recognition of amino acid by molecular receptor with asymmetric twisting; (C) discrimination of uracil and thymine derivatives by armed cyclononane molecular receptor.

macroscopically at a scale of tens of centimeters, results in the repeated capture and release of guest molecules. In an interfacial environment, nanoscale molecules can be grasped and released using bulk mechanical stimuli that can be manipulated by hand.

The molecular receptor depicted in Figure 15B is capable of generating an asymmetric twist and selectively absorbing chiral molecules due to its chiral environment upon compression of the receptor monolayer.[50] By aligning this receptor at the air-water interface, the monolayer system successfully reversed the binding selectivity of chiral isomers of amino acids through macroscopic compression. This enabled the recognition of chiral amino acid molecules through hand movements. Figure 15C shows a molecular receptor called armed cyclononane aligned as a monolayer on the water surface.[51] By gradually applying lateral pressure to the membrane and tuning its structure, it was found that, under optimal conditions, the receptor could selectively recognize uracil over thymine derivative with 64-fold accuracy. In this example, the selective recognition of nucleic acid bases was achieved by simply mechanically pushing the membrane. Thus, mechanical processes at the air-water interface allow for the manipulation of molecular machines and receptors through movements similar to hand movements.

Furthermore, another investigation quantitatively analysed molecular deformation at interfaces using a simpler molecular machine. Using binaphthyl-type amphiphilic molecules as openable molecular pliers, this research investigated the extent to which macroscopic mechanical energy is used for molecular deformation (Figure 16).[52] First, the system positioned the molecular pliers at an air-water interface and gradually compressed them. The dihedral angle of the binaphthyl group

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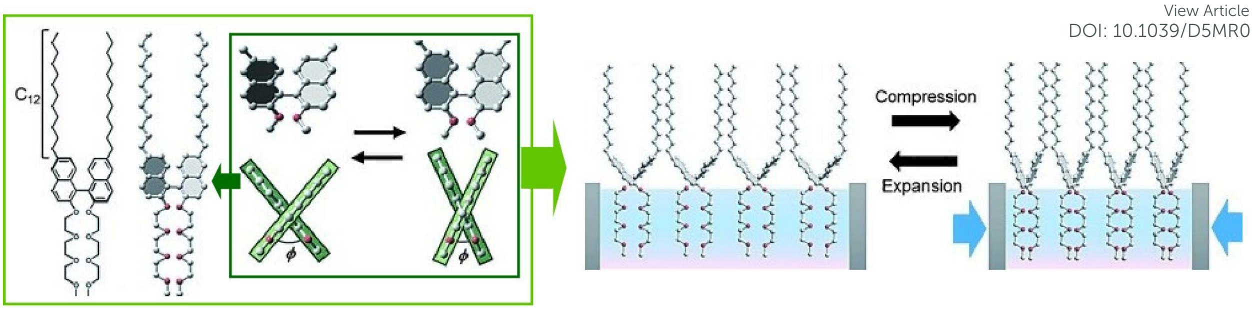

Figure 16. Mechanochemical tuning of the binaphthyl conformation at the air–water interface. Reprinted with permission from Ref. 52 Copyright 2015 Wiley-VCH.

was then measured to determine the degree to which the pliers were closed. Based on this data, this research used quantum chemical calculations to determine the energy required for molecular deformation. This approach also thermodynamically estimated the energy value for the mechanical deformation of a macroscopic monolayer. The two values were very similar. As surface pressure increases from 0 to 10 mN/m, approximately 0.2 kcal/mol of mechanical energy is stored and the same amount of molecular deformation energy is used. At higher pressures, however, the molecular deformation energy becomes smaller than the mechanical energy. Nevertheless, it has been suggested that, in interfacial systems, macroscopic mechanical energy can be used very efficiently to deform molecular machines. The results presented here pertain to the air-water interface, but it is highly intriguing that this analytical method can be applied to other interfaces possessing degrees of freedom. For instance, extending it to systems such as liquid-solid interfaces would allow for a more generalized understanding of the role of interfaces in mechanochemistry.

Figure 17A provides a rough comparison of the operating efficiency of machines and mechanisms in systems of various sizes.[53] The typical energy is calculated by multiplying the force acting on the system by its size. This value is then divided by the system's weight (mass) and plotted against its size. The data are compiled based on how much weight is manipulated per unit of energy. Despite their significant size differences, macroscopic machines, such as automobiles and human movement; molecular machines in biological systems; and the molecular machines at interfaces discussed here, manipulate large amounts of weight per unit of energy. Conversely, these are systems that move things with little energy. Conversely, artificial molecular machine systems that use photoisomerization and other mechanisms use large amounts of energy per unit weight. It is also known that photoisomerization and other processes consume energy in processes other than isomerization, resulting in inefficient operation. Mechanically driven molecular machine systems at interfaces can simply and efficiently drive molecular machines, much like scissors in macroscopic systems. It is experimentally known that processes such as photoisomerization involve significant waste. This is because such processes often include steps where absorbed energy is not fully utilized and is instead lost in other processes. In contrast, in mechanical deformation, energy is used efficiently. For example, as seen in the deformation of a spring, mechanical processes utilize all energy efficiently when there are no energy losses due to friction. There is still room for improvement in molecular machines. Mechano-nanoarchitectonics at interfaces will make this possible.

Figure 17B summarizes the relationship between different phenomena and applied forces.[54] Many biological phenomena are achieved through molecular deformation and movement due to weak forces. In contrast, stimuli such as light or conventional mechanochemistry only operate within a much larger range of forces. The delicate and sophisticated functions observed in biological systems, which operate within a weaker range of forces, remain unexplored in artificial stimulus systems. The force exerted at the air-water interface is calculated to be equivalent to the force in this unexplored area. Future development of advanced molecular functions must be based on interface science. The special dielectric environment and soft molecular deformation at the interface can greatly enhance the molecules' inherent potential.

This section demonstrated that the air-water interface is an ideal place to mechanically adjust nanostructures and molecular structures. Our work in this area is largely pioneering and has yet to be systematized. However, changes in aggregate and molecular structures are linked to various interactions, as well as to chemical, physical, and optoelectronic effects. As we expand our understanding of these interactions, we anticipate that this methodology will be applicable to a variety of systems. When considered fundamentally, the more important concept to emphasize is the mechanical confinement effect. Thermochemistry is mechanochemistry under conditions of low configurational order (high entropy). Mechanics promotes reactions by either reducing entropy or increasing configurational order. The gas-liquid interface is one method for orienting and constraining diverse reactive structures. This essence is expected to become clearer as techniques for analyzing various phenomena at the gas-liquid interface at the molecular level advance. While observation techniques on solid surfaces are currently highly developed, we also hope to see their development targeting the gas-liquid interface in future.

Mechanical processes at interfaces are interesting because they are highly efficient and driven by delicate forces similar to those found in biological systems. Biological systems function at soft interfaces, such as membrane surfaces, protein and receptor pockets, and biopolymer surfaces, rather than in

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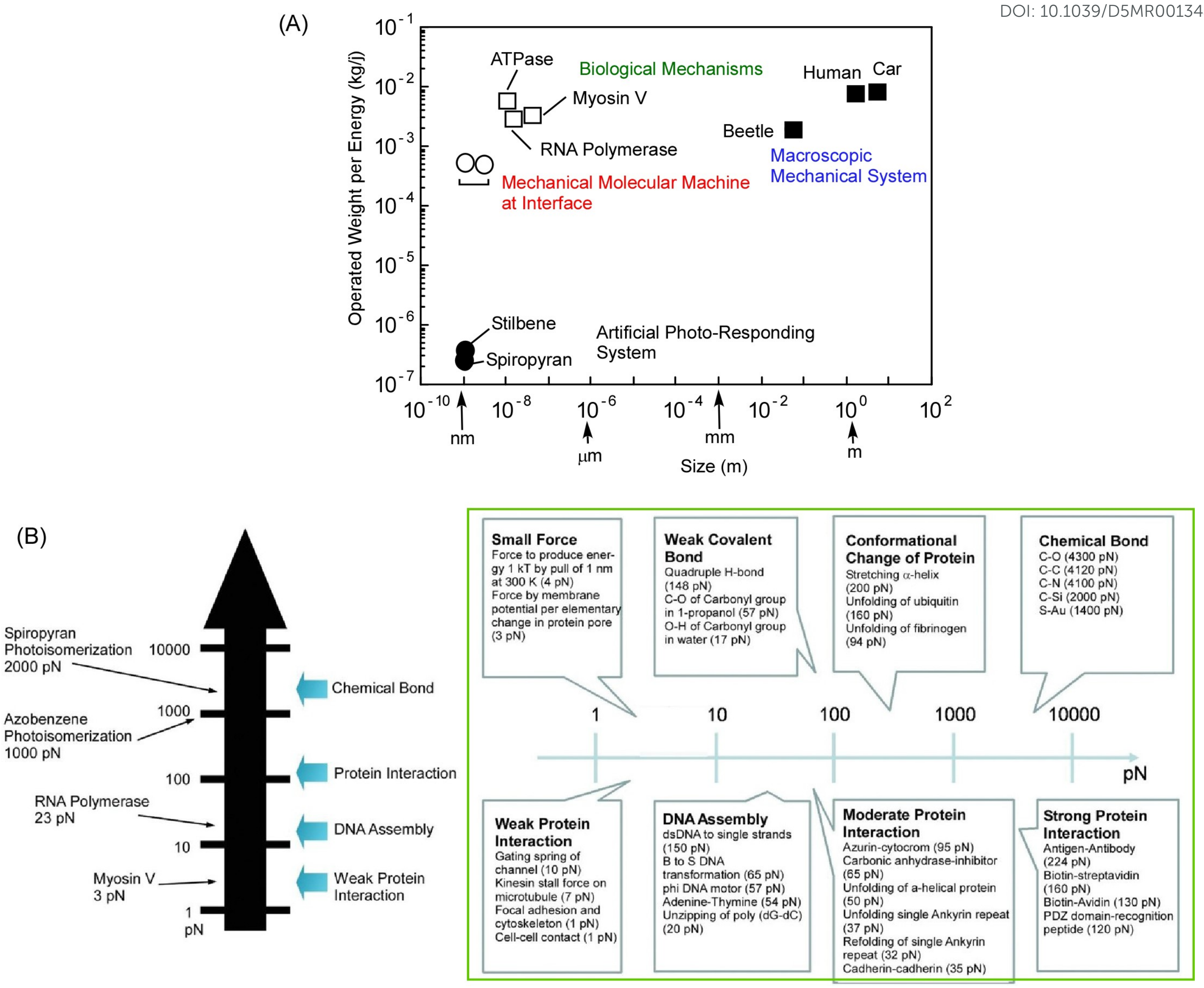

Figure 17. (A) Universal comparisons of machines: operated weight (mass) per energy at different scales; (B) Forces required for various molecular events varied over wide range from 1 to 10 000 pN level. Reprinted with permission from Ref. 54 Copyright 2022 Springer-Wiley-VCH.

solution. Biomolecules move flexibly in interfacial environments, demonstrating high efficiency and functionality. To artificially mimic such systems, the concept of mechano-nanoarchitectonics at interfaces is likely important. Of course, there are many thin film fabrication methods besides the LB method. However, as demonstrated in this section, LB thin films are particularly well-suited for studying specific phenomena at the solid-liquid interface relevant to biological systems.

5. Summary and future perspectives

Humans have long used mechanical processes to gather and process materials. Advances in material chemistry have made it possible to produce diverse materials in a rational way. Advances in nanotechnology and nanoarchitectonics now allow us to analyse and control these structures at the nanoscale. This process requires more advanced optical and electronic techniques. However, growing interest in mechanochemistry and mechanobiology suggests an integration of ancient mechanical processes and cutting-edge nanoprocesses. There is a sense of rational unification in the development of materials. Mechanical processes have significant advantages over more sophisticated optical and electronic processes because almost all materials can respond to mechanical stimuli. With the exception of those using probe microscopes, mechanical stimuli are macroscopic and require direct contact. Therefore, interfaces are necessary to link macroscopic mechanical processes to nanoscale structural and functional control. From this perspective, this paper introduces recent research examples through the concept of mechano-nanoarchitectonics at interfaces.

Although not exhaustive or representative, this paper has highlighted several examples of mechanochemistry and mechanobiology research emphasizing interfacial phenomena.

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Advances in scanning probe microscopy have enabled mechanochemistry researchers to analyse various nanoscale phenomena in conjunction with the application of mechanical stimuli. In addition to these cutting-edge technologies, active research is being conducted on mechanochemistry, which is more closely related to controlling functional materials through modifying surface structure and state. In particular, interface control is essential for structural control and the resulting expression of physical properties. Several mechanobiology examples demonstrate that the mechanical properties of simple interfaces can control advanced biological properties by inducing mechanical effects on surfaces. This applies to a wide range of targets, from controlling cell differentiation to understanding disease mechanisms. This is due to the highly organized functional morphology of cells. By constructing organized functional structures using nanoarchitectonics, simple mechanical stimuli on surfaces can drive highly complex functions. Furthermore, this review has demonstrated that the air-water interface is an ideal location for adjusting nanostructures and molecular structures mechanically. Mechanical processes at this interface are highly efficient and driven by delicate forces similar to those in biological systems. Advanced functional systems that mimic living organisms may emerge from these environments. Integrating these findings can lead to a comprehensive concept such as mechano-nanoarchitectonics at interfaces.

Moving forward, two additional directions are also worth considering. The first is the integration of multiple components. Biological systems, such as photosynthetic and signal transduction systems, exhibit sophisticated properties in which numerous functional elements are organized within a single membrane and function together.[55] Drawing from nature, one could employ nanoarchitectonics as a means of designing processes that take advantage of interconnected mechanical signaling. For instance, responses to mechanical stimuli could be transmitted between multiple functional units, resulting in directional functionality. Alternatively, responses could be aggregated to produce greater functionality. Creating such systems requires the rational control of the arrangement and organization of functional units within interfaces. Two-dimensional nanoarchitectonics technologies[56] that can assemble nanostructures within two-dimensional interfacial surfaces as designed are crucial to achieving this. Promising technologies include the formation of molecular patterns at the air-water interface[57] and two-dimensional metal-organic frameworks (MOFs)[58] and covalent organic frameworks (COFs).[59] Of course, this review does not cover every single case or concept. For instance, it does not mention “catch bond,”[60] one of the most intriguing features of biomacromechanics. Catch bond refers to the phenomenon where bond strength increases as applied force rises, just before the bond is ultimately broken. Although not discussed in this review, such processes should also be incorporated into the above considerations. Similarly many research efforts in mechanochemistry and related fields are continuously made. Therefore, recent papers are additionally cited as references for readers.[61]

Another direction is toward larger surface areas. Interfacial mechanochemistry and mechanobiology greatly contribute to elucidating mechanisms through discussions at a nanoscopic scale. Linking these findings to large-scale industrial outputs may present a future challenge. Although the interface environment is nanoscale in thickness, it can be infinitely expanded in the in-plane direction. Even if each mechanically driven unit is nanofunctional, nanofunctions can be generated as massive outputs if an extremely large number of them can be integrated on a two-dimensional surface and simultaneously mechanically driven. Advances in thin-film technology are needed to create large, highly reliable two-dimensional structures in huge areas to achieve this. Until now, nanofunctions have primarily been the subject of basic research, thought of as sophisticated yet minute. However, if nanofunctions could be expanded across a large two-dimensional surface and driven by mechanical stimuli, functions such as catalysis, signal transduction, and pollutant removal could be realized with nanoscale precision on a practical scale.

Author contributions

K.A.: conceptualization, writing, review & editing, funding acquisition.

Conflicts of interest

There are no conflicts to declare.

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