ChemNanoMat

CHEMNANOMAT
CHEMISTRY OF NANOMATERIALS FOR ENERGY, BIOLOGY AND MORE
ACES Asian Chemical Editorial Society

REVIEW OPEN ACCESS

Roles of Liquid Interface with Delicate Nanoarchitectonics for Molecular Recognition, Analysis, Reaction, Molecular Machine, Microrobot, and Life

Katsuhiko Ariga[1,2]

[1]Research Center for Materials Nanoarchitectonics, National Institute for Materials Science (NIMS), Tsukuba, Japan | [2]Graduate School of Frontier Sciences, The University of Tokyo, Kashiwa, Japan

Correspondence: Katsuhiko Ariga (ariga.katsuhiko@nims.go.jp)

Received: 13 September 2025 | **Revised:** 16 October 2025 | **Accepted:** 27 October 2025

Keywords: analytical technique | delicate control | liquid interface | molecular recognition | nanoarchitectonics

ABSTRACT

Ultimate functional materials could be functional substances found in biological systems. The organization of biological organizations can be described as a rational entity composed of functional small molecules and objects. It is becoming increasingly evident that liquid interfaces play a pivotal role in the development of sophisticated biological functions. It is therefore the case that the present review discusses the subject of nanoarchitectonics in the context of liquid interfaces as the environment. Specifically, the focus is on molecular recognition, analytical techniques, specific reactions, control of molecular machines and microrobots at liquid interfaces, and regulation of biological phenomena utilizing liquid interface environments. These examples will illustrate the role of liquid interfaces in achieving delicate nanoarchitectonics. The electronic properties are modulated in accordance with the variations in dielectric natures, extending perpendicularly to the interface. This enables delicate control of intermolecular interactions. Concurrently, the application of delicate forces within the interface direction facilitates precise structural adjustments of molecular structures analogous to those observed in biological systems. This theme is positioned as the convergence of chemistry (Chem) and nanotechnology/nanoarchitectonics (Nano) for materials (Mat) creation. This paper reveals the unexpectedly important role liquid interfaces can play within this context.

1 | Introduction

1.1 | Challenge Toward Ideal Materials

First of all, I would like to consider the future of functional materials by looking back. This is not a matter of decades, but the history of the evolution of life, which dates back billions of years. The mechanisms of existing biological organisms are highly sophisticated, with numerous functional elements cleverly arranged [1–3]. Such high functional systems have been built up over billions of years of evolutionary history of life. These elements function in a rational manner to perform a wide range of functions at room temperature and pressure with extremely high efficiency and selectivity. Developing such functional systems could be said to be the ultimate goal of developing future materials. It is our responsibility to establish a methodology for creating the ultimate future materials within a few decades, mimicking the know-how for creating functional systems that has been built up over billions of years of evolutionary history.

Living matter can be described as a rationally organized body of functional small molecules and materials. While this organization is highly sophisticated, it is safe to say that it falls within the realm of materials science. The key to achieving this is

---

(i) developing functional unit materials; (ii) controlling the precise structure, such as the nanostructure, of those unit materials; and (iii) rationally organizing those materials into functional systems. Without the billions of years it took living organisms, humanity is making progress on our historical scale. This progress can be so far roughly summarized as follows. Humans originally processed natural materials to make tools and others. In the 20th century, various material chemistries were developed, enabling them to create materials upon our designs. The creation of functional materials continues today through organic chemistry [4–8], inorganic chemistry [9–14], polymer chemistry [15–19], supramolecular chemistry [20–24], coordination chemistry [25–29], materials chemistry [30–35], biochemistry [36–41], and more [42–44]. Furthermore, it has been discovered that the physical properties and functionality of the same material can be improved by adjusting its precise structure, particularly its nanostructure [45–48]. Nanotechnology has made this progress significantly, which makes it possible to observe structures at the atomic and molecular level [49–52], create and manipulate structures at a nanoscopic level [53–56], and evaluate physical properties in these environments [57–60], which is being actively conducted today.

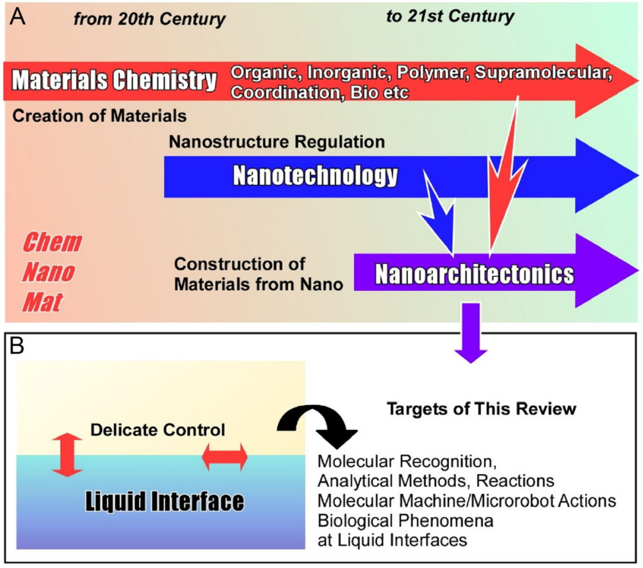

FIGURE 1 | (A) Outline and history of nanoarchitectonics concept. (B) Targets of this review article, roles of liquid interface with delicate nanoarchitectonics.

1.2 | Nanoarchitectonics Has Appeared

Finally, architecting and organizing unit objects with controlled nanostructures into functional materials are required. Indeed, research into creating functional structures through assembly and organization continues to attract attention. Self-assembly and self-organization in supramolecular chemistry [61–65] and thin-film fabrication methods such as, in interface science, self-assembled monolayer (SAM) [66–68], the Langmuir-Blodgett (LB) method [69–71], and layer-by-layer (LbL) assembly [72–74] are being investigated as methods for creating structurally controlled functional materials. Furthermore, methods for creating ordered nanostructures, particularly nanoporous structures, have been developed including metal-organic framework (MOF) in coordination chemistry [75–79], covalent organic frameworks (COF) in polymer chemistry [80–83], and mesoporous/nanoporous materials synthesized by template synthesis [84–86]. While these research approaches each have their own unique value systems, a more comprehensive and integrated concept is needed. Just as nanotechnology has driven the comprehensive development of nano- and micro-science and technology, a more integrated concept of assembling materials is required. This is the post-nanotechnology concept of nanoarchitectonics (Figure 1) [87], which is the concept of constructing functional materials from nano-units such as atoms, molecules, and nanomaterials [88, 89]. Just as nanotechnology was founded by Richard Feynman in the mid-20th century [90, 91], nanoarchitectonics was proposed by Masakazu Aono as the 20th century turned into the 21st century [92, 93]. Now in the 21st century, we are transitioning from nanotechnology to nanoarchitectonics.

Nanoarchitectonics is a very general concept that weaves together various processes to build functional materials from nanoscale. It transcends the boundaries of physics, chemistry, and biology and is applicable to any material and any application. Nanoarchitectonics can incorporate not only expected molecules and molecular association behavior, but also molecular design including covalent bonds [94] and material synthesis such as alloys [95]. Rather than proposing a completely new methodology, nanoarchitectonics breaks down the boundaries between fields to create an integrated concept. Because matter is essentially composed of atoms and molecules, nanoarchitectonics has the potential to become a universal principle applicable to all materials. Just as physics aims to be a theory of everything [96], nanoarchitectonics could become a method for everything as ultimate strategy in materials science [97, 98].

1.3 | Delicate Nanoarchitectonics at Liquid Interface

As mentioned at the beginning of this introduction, the ultimate functional material would be a functional substance like that found in biological systems. Biological tissues can be described as objects composed of functional small molecules and small objects organized in a rational manner. The reason such highly functional objects can be constructed is because they provide a suitable platform for nanoarchitectonics. What is the most important feature that enables biological functions to be constructed and functioned? It is an interface, specifically flexible and fluid interface. For example, the phenomenon of liquid–liquid phase separation (LLPS) has recently attracted attention [99–101]. Liquid–liquid phase separation occurs spontaneously within cells, forming membranes, separated endoplasmic reticular, which serves as a site for various biological functions. Such structures are said to be key to the emergence of life. It is becoming increasingly clear that fluid interfaces, such as liquid interfaces, play an important role in the development of advanced biological functions.

According to these backgrounds, this review paper will discuss nanoarchitectonics using liquid interfaces as an environment. Here, I will introduce several research examples, primarily using the air–water interface as a model system that is easy to analyze for liquid interfaces, and consider the essential role of liquid interfaces in nanoarchitectonics. The review will cover molecular recognition at liquid interfaces, analytical methods, reactions controlled at liquid interfaces, control of molecular machines and microrobots at liquid interfaces, and regulation of biological phenomena using the liquid interface environment. Overall, all of these require extremely delicate control. Liquid interfaces are soft and sometimes ambiguous, allowing for delicate parameter changes and control of electronic and mechanical factors. From this perspective, this review will discuss the role of liquid interfaces in enabling delicate nanoarchitectonics.

2 | Molecular Recognition at Air–Water Interface

This section discusses molecular recognition phenomena at liquid interfaces, particularly the air–water interface. The delicate electronic changes at the air–water interface provide valuable clues to understanding the fundamental mechanisms underlying the function and structure formation of biological systems. The sophisticated functionality of biological systems relies on highly selective molecular recognition and molecular assembly. Molecular recognition is essential for specific molecular signals to be received by receptors and transmitted, and for specific functional components to assemble and function together. Hydrogen bonds play a central role in these processes. Indeed, hydrogen bonds are essential for important functions such as the formation of double helices in DNA and the reception of specific substrates by receptors and enzymes. Hydrogen bonds interact through hydrogen atoms based on electronic uneven distributions. Therefore, hydrogen bonds are difficult to form in polar environments where charge imbalances are alleviated. Water, in particular, is the worst medium for hydrogen bonding. This presents a serious paradox in biological function. Biological systems are essentially aqueous solutions, where hydrogen bonds play a crucial role. However, hydrogen bonds are difficult to form in water. The solution to this contradiction is use of the interface where water comes into contact. Molecular recognition in living organisms does not occur independently in aqueous solution, but happen at interfaces in aqueous solution, such as the surface of cell membranes, the inner wall interface of enzyme active pockets, and the polymer interface formed by DNA. Without interfaces, we cannot discuss biological functions.

What is the secret? What is the role of the interface? Research to clarify this was conducted on monolayers at the air–water interface. Receptor molecules were placed on the monolayer located on the air-phase side, and various biomolecular were dissolved in the aqueous phase [102–104], and molecular recognition phenomena at this interface were examined. The results demonstrated that biomolecular such as sugars [105], nucleic acid bases [106], amino acids [107], peptides [108, 109], and nucleotides [110] are recognized via hydrogen bonds. In other words, it was experimentally demonstrated that, given the appropriate interfacial environment, biomolecules can recognize each other via hydrogen bonds even in the presence of water (Figure 2A).

Furthermore, it was investigated whether the type and shape of the interface affect the strength of molecular recognition. The molecular recognition pair was immobilized on guanidinium and phosphate, and the binding constant was examined under various conditions (Figure 2B) [111]. When guanidinium and phosphate were dispersed in water, the binding constant was only 1.4 M^{-1}. When the molecular recognition site was transferred to the mesoscopic interface in water, such as the surface of a micelle or a lipid bilayer vesicle, the binding constant increased to 10^2–10^4 M^{-1}. Furthermore, when the macroscopic air–water interface was used as the recognition site, the binding constant between guanidinium and phosphate reached 10^6–10^7 M^{-1}. Even with the same molecular recognition pair, the binding constant could increase by a million to ten million times, simply by changing the interfacial environment. Importantly, such a high binding constant was obtained even in the presence of water.

Why does the strength of hydrogen-bonded molecular recognition dramatically increase at interfaces? A molecular model system, such as that shown in Figure 2C, was established, and quantum mechanical calculations were performed while varying the interfacial position and the distance between the guanidinium and phosphate molecules to estimate the binding constant at each interfacial position [112]. In this research, reaction field calculation combined with the AM1 molecular orbital method was applied. A multidielectric system was used to simulate interfacial environment, where a cylindrically shaped dielectric with a dielectric constant of 2 for the lipid phase was embedded in the infinitely extended dielectric with a dielectric constant of 80 for the water phase. However, this simplified calculation system neglected dynamic diffusion effect of interfacial water molecules. The binding constant is large when the system is in a hydrophobic phase and small when in a polar phase. However, the binding constant changes significantly with delicate changes in the interfacial position, and experimental binding constants were reproduced even when the hydrogen-bonding site is in the aqueous phase. Even if the hydrogen-bonding site is in the aqueous phase, which has a high dielectric constant, intermolecular interactions are influenced by the nearby low dielectric constant, allowing hydrogen bonding. This is the secret behind how biological systems enable molecular recognition and assembly through hydrogen bonding in aqueous environments. When considering a liquid interface such as an air–water interface perpendicular to the interface, it can be considered a site where electronic effects can be delicately altered. This not only enables hydrogen-bond molecular recognition in biological systems, but also potentially provides a site for delicate assembly of functional elements containing water-soluble molecules.

The air–water interface has elucidated hydrogen bonding in molecular recognition and has also become a platform for studying different types of molecular recognition events. Ishii, Akamatsu, and coworkers studied the mechanical control of fluoride anion recognition with naphthalenediimide monolayers at the air–water interface as shown in Figure 3 [113]. Mechanical stimuli, specifically monolayer compression and expansion processes, modified the packing of the naphthalenediimide monolayer and repeatedly triggered anion binding and the associated changes in membrane potential. This behavior can be compared to the behavior of mechanosensitive ion channels in biological membranes. The behavior of anion binding or release was dependent on the structure of the alkyl side chains attached to the naphthalenediimide core. Naphthalenediimides with saturated linear alkyl side chains exhibit a strong propensity to interact with one another, thereby enabling molecular packing around the vertically aligned naphthalenediimide core. The packing mode is highly favorable for anion-π interactions, which

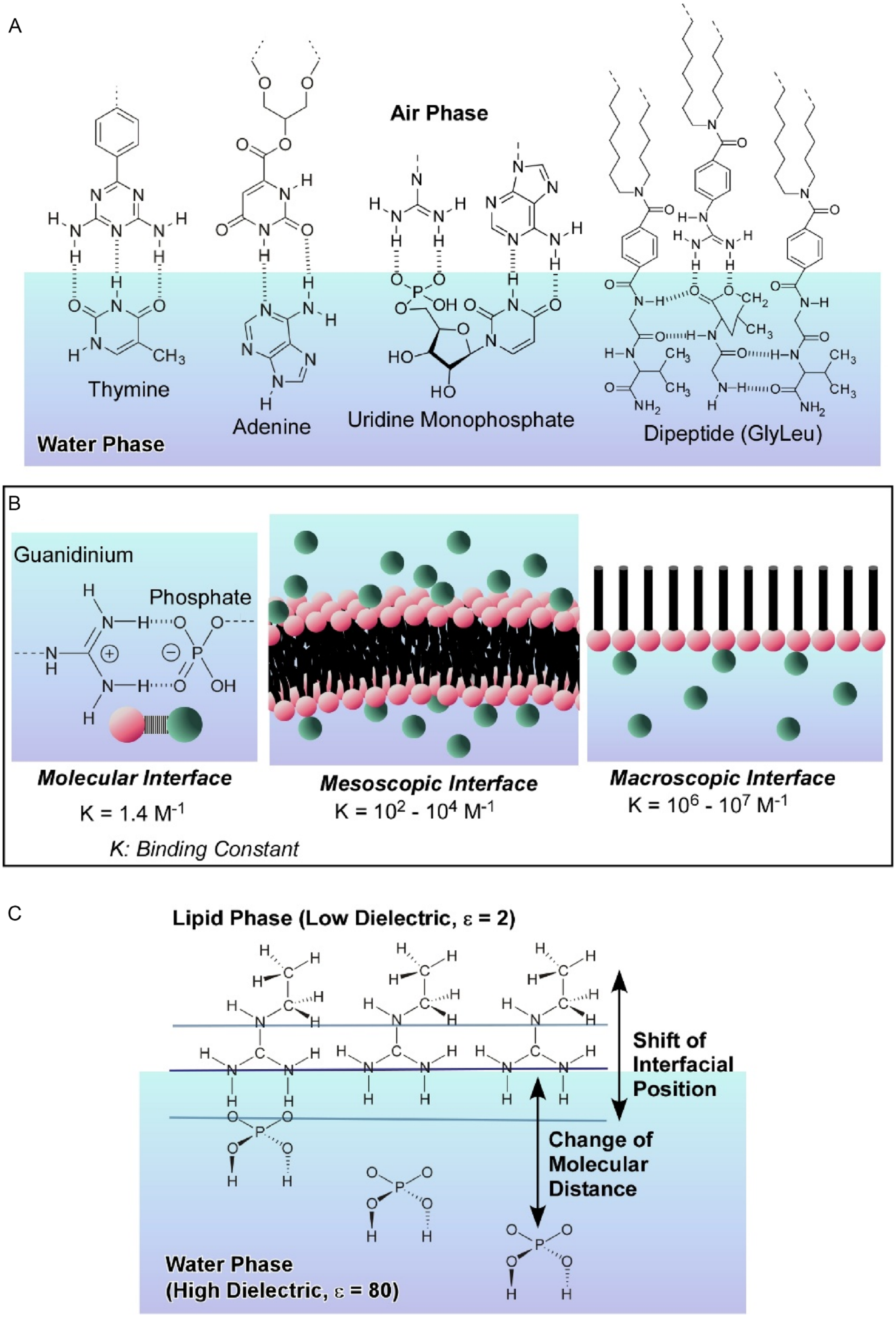

FIGURE 2 | (A) Examples of biomolecular recognitions at the air–water interface. (B) Binding constants of guanidinium and phosphate pairs under various environments. (C) Molecular model system for quantum mechanical calculations with varying the interfacial position and the distance between the guanidinium and phosphate molecules to estimate the binding constant at each interfacial position.

form a charged sandwich structure with a significant molecular footprint within the naphthalenediimide monolayer. Compression of this naphthalenediimide monolayer led to a phase transition, expelling anions and producing a discharge. In contrast, naphthalenediimides with branched alkyl side chains displayed the opposite tendency. The side chains' low packing tendency meant that anions were initially undetected in the densely populated, large molecular areas prior to

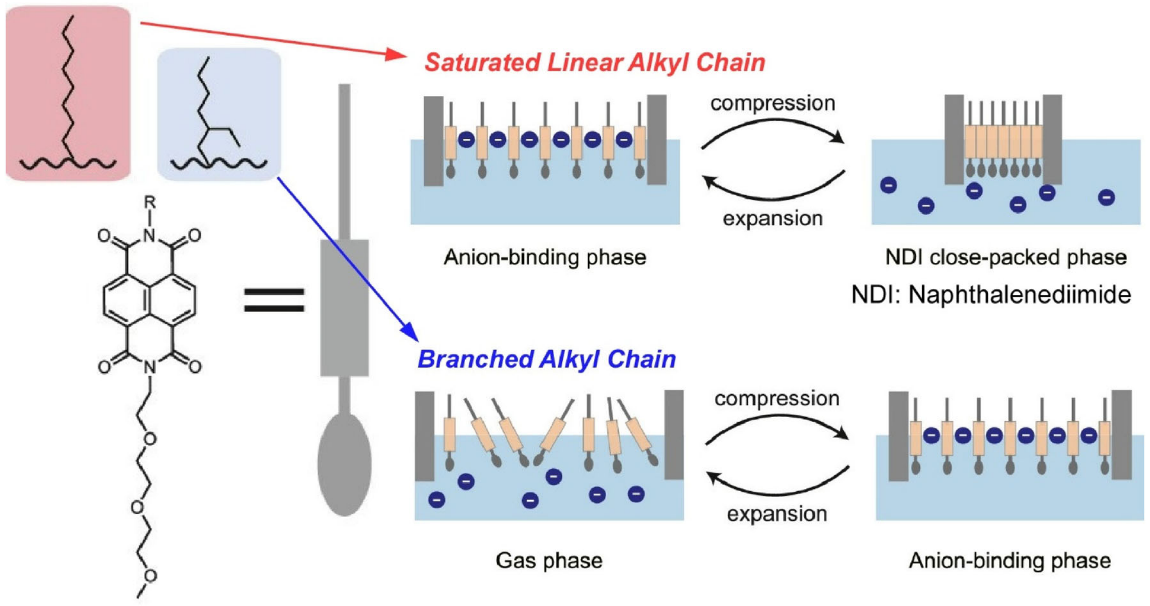

FIGURE 3 | The mechanical control of fluoride anion recognition with naphthalenediimide monolayers at the air–water interface, where mechanical stimuli, specifically monolayer compression and expansion processes, modified the packing of the naphthalenediimide monolayer and repeatedly triggered anion binding and the associated changes in membrane potential. Reproduced with permission [113]. Copyright 2024, American Chemical Society.

compression. The compression of the monolayer formed a sandwich structure, leading to charging that is dependent on anion recognition. These behaviors replicate signal production and transmission processes found in biological systems, where different stimuli and energy are converted into membrane potentials via ion transport and binding. Implementing this concept in artificial systems could result in the development of biomimetic signal transmission devices and energy-gathering systems. Combining two monolayers whose surface potential difference changes sign upon the application of a mechanical stimulus may be used to create an XOR gate in digital circuits. This may lead to the creation of devices based on mechanically induced ion recognition monolayers.

Molecular recognition phenomena of the inclusion type have also been studied at the air–water interface. The air–water interface in a two-dimensional confined space may elevate local concentrations of both host and guest molecules, thus facilitating the inclusion phenomenon. Ishii et al. found that ethoxy-functionalized pillar[5]arene host molecules and amphiphilic straight-chain fatty acid guest molecules create oriented monolayers at the air–water interface via inclusion-type self-assembly (Figure 4) [114]. Here, a pseudorotaxane structure is created, which orients and self-assembles at the air–water interface to form a monolayer. The length of the alkyl chain in the fatty acid guest determines the resulting monolayer's structure according to this principle. Straight-chain fatty acid guests form pseudorotaxane monolayers with ethoxy-functionalized pillar[5]arene cavities oriented toward the water surface. In contrast, fatty acid guests that are incompatible with ethoxy-functionalized pillar[5]arene undergo phase separation. The guest mixing ratio and chain length were found to control the stoichiometry of pseudorotaxane formation and the monolayer composition. Removing guest molecules from pseudorotaxanes consisting of macrocyclic host molecules and linear guest molecules should allow for the formation of two-dimensional arrays of supramolecular pore

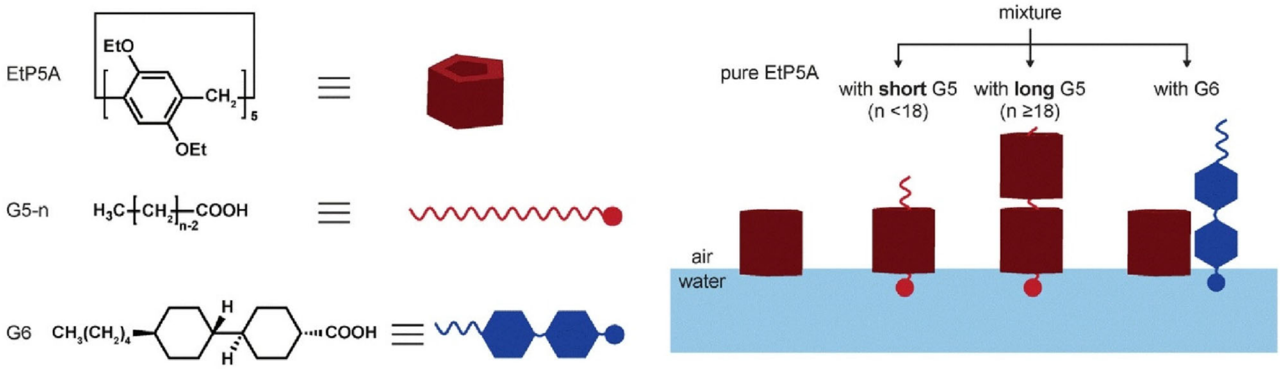

FIGURE 4 | Formation of oriented monolayers with ethoxy-functionalized pillar[5]arene host molecules and amphiphilic straight-chain fatty acid guest molecules create at the air–water interface via inclusion-type self-assembly to result in a pseudorotaxane structure. Reproduced under the terms of the CC-BY license [114]. Copyright 2025, Royal Society of Chemistry.

structures. These materials are anticipated to be employed in sensors and filters.

The mutual recognition within the monolayer described above is also evident in the nanoarchitectonics of the in-plane film structure through chiral recognition. Researchers led by Negi studied the behavior at the air–water surface of a monolayer consisting of 2,2′-bis(octadecyloxy)-1,1′-binaphthyl-6,6′-dicarboxylic acid, a compound with axial chirality resulting from a binaphthyl group with two carboxylic acid groups (Figure 5) [115]. The racemic form gives rise to a solid film, whereas the chiral S-isomer results in a liquid film. The analysis of the atomic force microscope on the monolayer structures showed that the actual lattice arrangements of the two single layers are substantially distinct. The structural differences are attributed to the stereoregularity caused by axial chirality. These behaviors result in distinct intermolecular interaction patterns between the two monolayers. A solid film of the racemic form is created through a cooperative network of π–π interactions between the naphthyl rings and strong van der Waals interactions. In contrast, the S-isomer of the chiral form has a greater intermolecular distance than its racemic counterpart, which makes it challenging for strong intermolecular forces to act, and its long alkyl chains and naphthyl rings are arranged one-dimensionally via van der Waals interactions. As the monolayer is compressed in this case, the dihedral angle of the binaphthyl rings within the one-dimensional columnar structure diminishes, and a phase transition from a liquid-expanded film to a liquid-condensed film takes place. Chiral molecular information can carefully regulate the intermolecular interactions within a monolayer plane and determine its two-dimensional nanoarchitecture.

Several developments have been made in methods for analyzing phenomena at liquid interfaces, as outlined in the following section. Methods such as these have made a significant contribution to elucidating molecular recognition behavior at the air–water interface. Biological membranes interact with small molecules and proteins through different mechanisms, such as molecular recognition, structural stabilization, and transmembrane signaling. Molecular interactions within lipid monolayers, which replicate biological membranes, hold significant relevance. Kim and coworkers employed sum-frequency vibrational spectroscopy with 1,2-dipalmitoyl-3-trimethylammonium propane, featuring a choline group, as depicted in Figure 6A [116]. The sum-frequency signal suddenly reappeared when aromatic molecules were introduced into the aqueous phase due to molecular adhesion. They examined the adsorption process of aromatic ring molecules in the aqueous subphase onto the choline group of 1,2-dipalmitoyl-3-trimethylammonium propane via cation-π interactions. Evidence of π molecule penetration was observed through increased CH signal intensity of the aromatic ring and alterations in surface pressure. Benzylamine molecules, unlike those of phenol and indole, competed with 1,2-dipalmitoyl-3-trimethylammonium propane at the interface, thereby decreasing the number density of cationic surfactant molecules. This approach could be employed to sensitively evaluate the penetration of foreign molecules into in vitro biological membrane models.

Interactions between peptides and anions are crucial to numerous biological processes, including ion transport and signal transduction. Examining them in model systems at the air–water interface has significant implications in physiological contexts. Gunwant and Pandey employed vibrational sum-frequency generation spectroscopy to study the interaction between the amphiphilic peptide melittin and Hofmeister series anions at various pH levels as shown in Figure 6B [117]. In acidic environments with low pH, the various charge states of peptides have a significant impact on their interactions with anions, and electrostatic forces enhance the bonding between the peptide and the anion. Interactions with anions under neutral pH conditions are

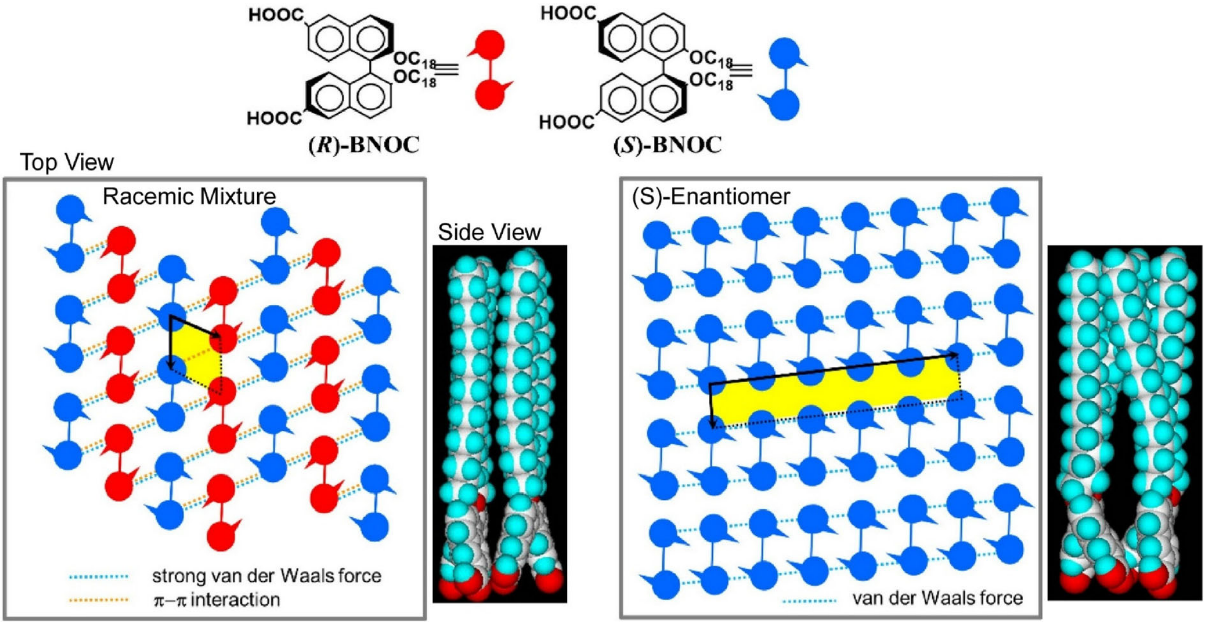

FIGURE 5 | Packings at the air–water surface of a monolayer consisting of 2,2′-bis(octadecyloxy)-1,1′-binaphthyl-6,6′-dicarboxylic acid, a compound with axial chirality resulting from a binaphthyl group with two carboxylic acid groups, where the racemic form gives rise to a solid film and the chiral S-isomer results in a liquid film. Reproduced with permission [115]. Copyright 2023, Oxford University Press.

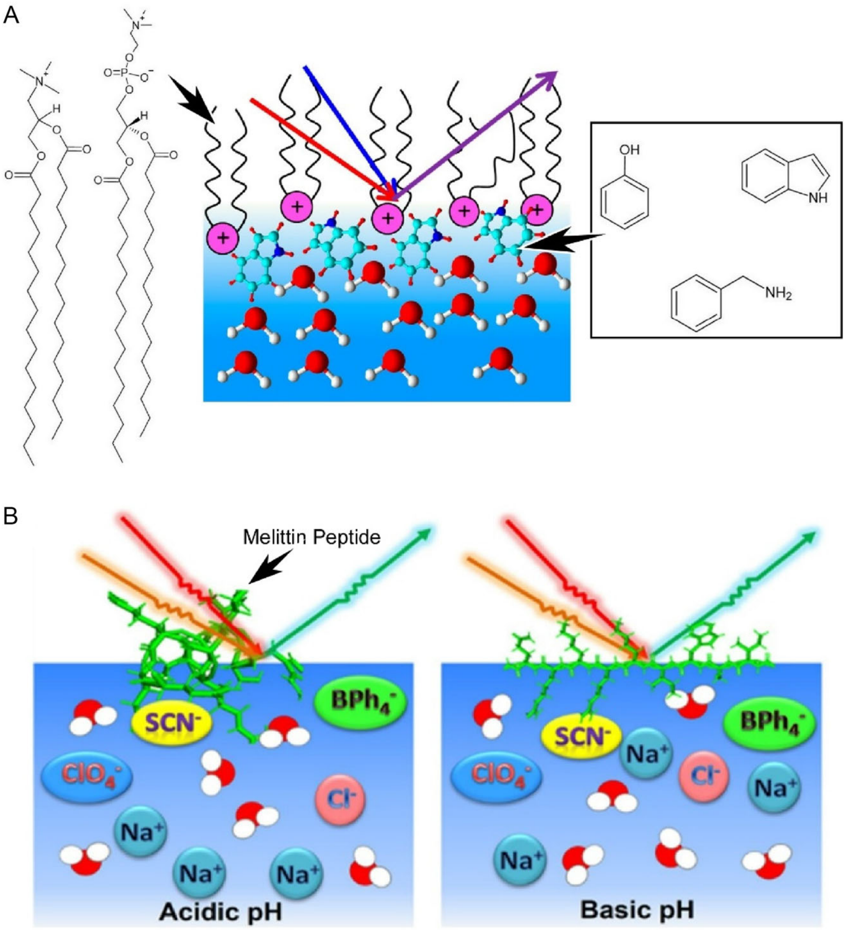

FIGURE 6 | (A) Application of um-frequencyvibrational spectroscopy to 1,2-dipalmitoyl-3-trimethylammonium propane and DPPC with adsorption process of aromatic ring molecules in the aqueous subphase onto the monolayer via cation-π interactions. Reproduced with permission [116]. Copyright 2023, American Chemical Society. (B) Use of vibrational sum-frequency generation spectroscopy to study the interaction between the amphiphilic peptide melittin and Hofmeister series anions at various pH levels. Reproduced with permission [117]. Copyright 2025, American Chemical Society.

primarily driven by the anion's affinity for the peptide surface. Peptides show stronger interactions with anionic species that have high surface activity. As a result, the neutral charge of the peptide leads to a substantial decrease in these interactions. Under basic conditions, peptide-anion interactions become independent of the type of anion. The interaction's strength is determined by the relative affinity and concentration of competing ions. In addition, these findings indicate that water molecules at the interface have a significant impact on controlling these interactions. Clarifying these interactions can offer valuable insights into peptide stability, solubility, and functionality, which will contribute to advancements in drug delivery systems, antimicrobial therapy, and biotechnological applications.

The air–water interface serves as an extremely valuable environment for research into molecular recognition. Hydrogen-bonding molecular recognition is particularly crucial in living organisms, and the fact that hydrogen bond strength is affected by delicate variations in interfacial position is extremely significant. It leads to the crucial conclusion that hydrogen bonding plays a significant role in aqueous biological systems since they effectively utilize the interfacial environment. Inclusion interactions, chiral recognition, and other interactions are also investigated at the air–water interface, in addition to hydrogen bonding. The asymmetrical air–water interface is a sensitive medium for specific spectral analyses. These analytical techniques are crucial instruments for deciphering the intricate physical characteristics of liquid interfaces. The following section provides recent development of analysis at the air–water interface.

3 | Analysis at Air–Water Interface

There are now a lot of interests in ways to analyze what happens at the surface of a liquid, like the air–water interface. The signals used to analyze interfaces are not as strong as the methods used to analyze bulk materials. Also, unlike with solid interfaces, it is hard to use direct atomic and molecular observation methods such as STM and AFM with nonsolid liquid interfaces. When studying liquid interfaces, scientists use techniques such as measuring surface pressure and spectroscopic techniques like reflectance spectroscopy [118–120]. What's more, a type of second-order nonlinear spectroscopy is naturally selective to interfaces and has become a vital spectroscopic method for molecular science research at interfaces [121, 122]. It is also useful to combine these methods with theoretical approaches such as molecular dynamics simulations. This review cannot cover all of these approaches here, but this section gives some examples of ways to analyze things that happen at the air–water interface.

In their study, Nayak and his coworkers employed a combination of conventional interface science techniques and computational science to investigate monolayers of alkylated meta-substituted azobenzene derivatives [123]. This investigation utilized a dual approach of molecular dynamics simulations and surface pressure measurements. The quantification of hydrogen-bonding interactions in their photoisomeric monolayers was the subject of their research. Molecular dynamics simulations were utilized to analyze Z-density profiles, radial distribution functions, order parameters, and hydrogen bonds, thereby confirming experimental observations of changes in monolayer surface pressure, dipole moment, and thickness. The results of the study revealed significant details. Minor alterations in the quantity of hydrogen bonds present within intermolecular and molecule-water interactions have been demonstrated to induce substantial modifications in the properties of monolayers. The position of the substituents on the phenyl ring of azobenzene exerts a significant influence on the stability of the trans- and cis-isomers. The self-assembly properties of azobenzene molecules can be significantly altered by supramolecular aggregation through intermolecular hydrogen bonding. The quantification of hydrogen-bonding interactions is imperative for the rational design and optimization of self-assembled materials for a range of applications. Amphiphilic azobenzene molecules have significant potential for the design of photocontrollable functional supramolecular systems in aqueous media. These analytical results are of fundamental importance for the design of photoresponsive molecules with customized properties for targeted drug delivery and other industrial applications.

Acidity is a significant factor in determining chemical reactivity in atmospheric aqueous aerosols and in microdroplets employed in catalytic reactions. It requires to elucidate the differences in acidity between bulk solutions and interface. To this end, de la Puente and Laage conducted neural network-based hybrid molecular simulations of density functional theory, accounting for explicit nuclear quantum effects [124]. These simulations were then combined with analytical models to successfully describe the pH and self-ion concentration of droplets and films ranging in size from nanometers to micrometers (Figure 7). The present study elucidates the mechanisms by which the acidity of water droplets and thin films is regulated by the properties of the air–water interface and the surface area-to-volume ratio. Utilizing molecular dynamics simulations, they elucidated the alterations in the self-dissociation equilibrium of water slabs and the stability of hydronium and hydroxide ions as they approach the air–water interface. Subsequently, these results were incorporated into a general analytical model, thereby revealing the pH and ion concentration profiles of droplets and water films that exhibit size-dependent changes. In system sizes of less than a few tens of nanometers, acidity is found to be strongly dependent on the surface area to volume ratio. The presence of hydroxide ions in the core, along with hydronium stabilization at the interface, results in an increase in pH. The results obtained in pure aqueous systems have significant implications for the understanding of chemical reactivity in atmospheric aerosols and catalysis in aqueous microdroplets. The model can be readily extended to encompass scenarios involving the presence of multiple acidic species. This will contribute significantly to our understanding of the chemical reactivity of atmospheric aerosols, whose properties are fundamentally affected by acidity.

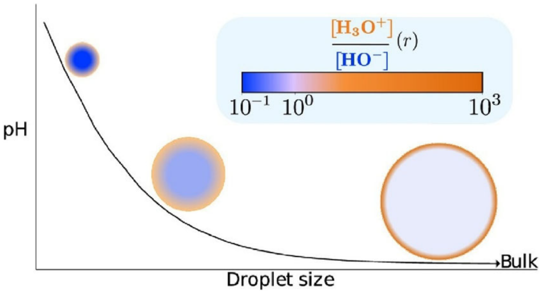

FIGURE 7 | Neural network-based hybrid molecular simulations of density functional theory combined with analytical models to successfully describe the pH and self-ion concentration of droplets and films ranging in size from nanometers to micrometers, where the pH and ion concentration profiles of droplets and water films exhibit size-dependent changes. Reproduced with permission [124]. Copyright 2023, American Chemical Society.

The accurate modeling of aerosol climate impacts is contingent upon the correct characterization of the acid–base behavior and surface protonation states of atmospherically relevant organic acids. Furthermore, such characterization is also conducive to the advancement of our understanding of interfacial reactivity. Rapf and coworkers conducted a study on the protonation states of two medium-chain α-hydroxy acids, namely 2-hydroxyhexanoic acid and 2-hydroxyoctanoic acid [125]. This investigation was conducted at the air–water interface and in bulk (Figure 8). The effective surface pK_a values obtained from conventional surface tension measurements are influenced by contributions from adsorption and desorption processes. Consequently, these values do not directly probe differences in dissociation equilibria at interfaces. In this study, the surface dissociation of α-hydroxy acids was investigated in situ using infrared reflection–absorption spectroscopy (IR-RAS). It has been determined that the measurement of pH in bulk form alone is

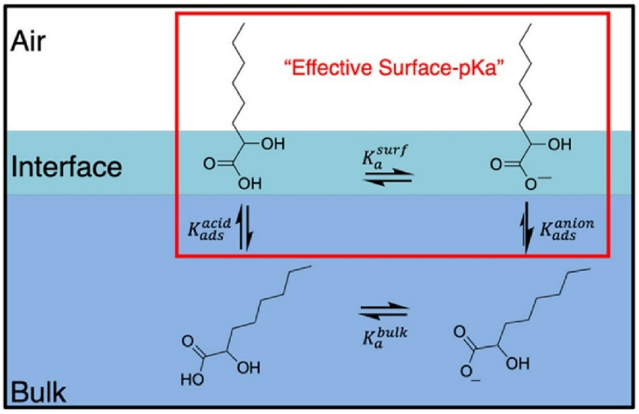

FIGURE 8 | Investigation of the surface dissociation of two medium-chain α-hydroxy acids, namely 2-hydroxyhexanoic acid and 2-hydroxyoctanoic acid by in situ using infrared reflection–absorption spectroscopy (IR-RAS) with consideration on differences in dissociation equilibria at interfaces. Reproduced with permission [125]. Copyright 2025, American Chemical Society.

an inadequate predictor of the distribution of species present at interfaces. Therefore, the importance of fundamental laboratory studies to investigate speciation at the air–water interface as a function of solution conditions has been confirmed. The protonated forms of organic acids exhibit increased surface activity, while the deprotonated, charged anionic forms demonstrate greater solubility in the bulk. Consequently, a discernible alteration was evident at the air–water interface for both 2-hydroxyhexanoic acid and 2-hydroxyoctanoic acid, accompanied by a decline in the degree of dissociation relative to the bulk. It was also observed that surface activity increased with increasing tail length, with the relative magnitude of the effect increasing. It was hypothesized that the discrepancy in surface activity between the acid and anionic species is likely to account for a significant proportion of the observed change in protonation state at the interface.

Elucidation of the energy dissipation process is imperative to comprehension of the various phenomena occurring in nature. In particular, the vibrational relaxation and its timescale at water interfaces where the hydrogen-bonding network is disrupted have been a topic of debate. In their research, Tahara and coworkers centered their attention on the OH stretching of interfacial water at the air–water interface [126]. Utilizing femtosecond time-resolved, heterodyne-detected vibrational sum-frequency generation (TR-HD-VSFG) spectroscopy, they embarked on an investigation into its vibrational relaxation. This finding indicates that the vibrational relaxation of interfacial hydrogen-bonded OH occurs predominantly via an intramolecular relaxation mechanism, analogous to that observed in bulk water. This phenomenon signifies the conversion of excited free OH to excited hydrogen-bonded OH, which is subsequently followed by a relaxation process leading to lower-frequency vibrations. The present study endeavors to derive a unified picture of vibrational relaxation at the air–water interface. It was therefore possible to clarify the overall relaxation dynamics of the OH stretching vibration at the air–water interface. Moreover, the differences and similarities with the relaxation dynamics in bulk liquid water were elucidated. These experimental observations provide a solid foundation for understanding the energy dissipation processes at various water interfaces with broken hydrogen bond networks at the air–water interface.

Conical intersections (CIs) have been shown to play a pivotal role in the manipulation and control of photochemical reaction pathways of molecules at interfaces and surfaces, with the capacity to influence their molecular dynamics. Rao and coworkers developed phase-cycling interface-specific two-dimensional electronic spectroscopy (i2D-ES) as a tool for characterizing CIs at interfaces and surfaces (Figure 9A) [127]. In conjunction with advanced computational modeling, the nonadiabatic CI dynamics of molecules at the air–water interface were investigated. i2D-ES is a fourth-order nonlinear spectroscopy technique in which four related pulses generate polarized light, resulting in the observed interface-specific spectral signal. Specifically, a phase-locked pump pulse pair is integrated with an interface-specific electronic probe to obtain two-dimensional interface-specific responses. It was demonstrated that the nonadiabatic transitions of surface-active azo dye molecules occurring via interfacial CI have kinetic pathways that differ from those in bulk water. The findings of the present study indicate that the orientational configuration of molecules exerts a significant influence on the reaction pathways at interfaces and surfaces. The orientational configurations of molecules, both in the ground and excited states, play an important role in determining the reaction pathways at interfaces and surfaces. The rates of interfacial charge separation and recombination have a significant impact on the efficiency of photovoltaic devices. The present study offers novel insights into the kinetics and lifetime of electronic states implicated in nonadiabatic transitions. It is anticipated that this will facilitate the elucidation of all light-induced interfacial processes, encompassing solar energy conversion, photosynthesis, photocatalysis, and environmental photochemistry.

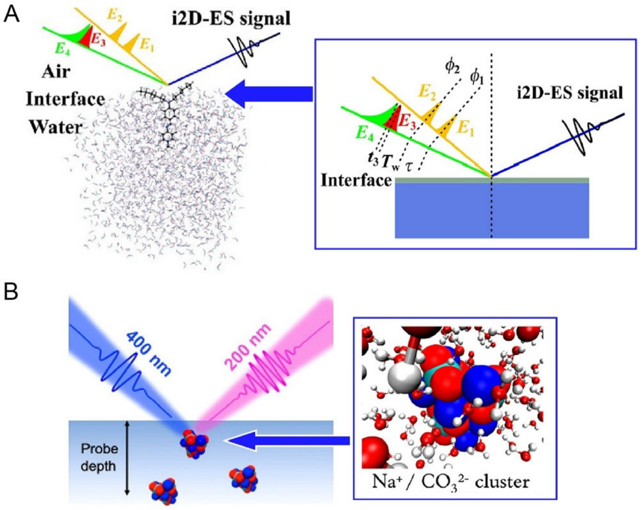

FIGURE 9 | (A) Phase-cycling interface-specific two-dimensional electronic spectroscopy (i2D-ES) for characterizing conical intersections (CIs) of molecules at interfaces and surfaces. Reproduced with permission [127]. Copyright 2023, American Chemical Society. (B) Resonantly enhanced deep-UV second-harmonic generation spectroscopy to investigate adsorption of carbonate (CO_3^{2-}) and bicarbonate (HCO_3^-) ions at the air–water interface. Reproduced with permission [128]. Copyright 2024, American Chemical Society.

Qian, Pascal, Saykally, and coworkers utilized resonantly enhanced deep-UV second-harmonic generation spectroscopy to ascertain the Gibbs free energy of adsorption of carbonate (CO_3^{2-}) and bicarbonate (HCO_3^-) ions at the air–water interface (Figure 9B) [128]. Utilizing resonantly enhanced deep-UV second-harmonic generation (DUV-SHG) spectroscopy, the researchers were able to directly probe carbonate and bicarbonate anions at the air–water interface with a high degree of surface specificity. In contrast to the predictions of classical electrostatic theory, it was found that divalent carbonate anions exhibited a stronger preference for the air–water interface than monovalent carbonate anions. The magnitude of this phenomenon was measured by calculating the respective Gibbs free energies of adsorption. The analysis revealed a high speciation of carbonate species in proximity to the interface, along with the presence of a deep potential minimum. These findings contribute to the advancement of fundamental understanding in the field of ion adsorption chemistry and have implications for significant practical processes, including ocean acidification, the chemistry of seawater spray aerosols, and mammalian respiratory physiology.

At the air–water interface, a multitude of physical properties deviate from those observed in the bulk, not limited to hydrogen-bond molecular recognition. The occurrence of such events is contingent upon delicate environmental changes in the vicinity of the interface. In addition to conventional interface science techniques, spectroscopy based on nonlinear optics has been demonstrated to be effective in analyzing these properties. Moreover, theoretical approaches, including molecular dynamics simulations, provide substantial support for these analyses. In contemplating the future, the analysis of delicate physical properties at liquid interfaces may represent an area in which artificial intelligence (AI) can be effective.

4 | Liquid Surface Reaction

Changes in physical properties at liquid interfaces, including intermolecular interactions such as hydrogen bonding and acid dissociation, have been demonstrated to result in a variety of phenomena. For instance, the reactivity of molecules present at a liquid interface can undergo alteration, which may consequently give rise to reactions that are exclusive to the interface. Consequently, liquid interfaces can also be considered to be sites where unique reactions can occur. This section introduces some of the recent research on the properties and analysis of liquid interfaces.

The gas–liquid interface of micro/nanobubbles has been shown to function as a reaction field for heterogeneous interfacial reactions. Pradeep and coworkers reported the formation of carbon–carbon bonds at the air–water interface of negatively charged aqueous microdroplets at room temperature (Figure 10), resulting in a catalytic carbon dioxide reduction reaction [129]. The reaction is initiated by the generation of a carbanion at the α-carbon of a ketone, which is then followed by a nucleophilic addition to CO_2. The formation of a C—C bond within the charged microparticles is initiated by the chemical inertness of CO_2 and the α-carbon of a ketone. The utilization of online mass spectrometry has led to the identification of the substance as a β-keto acid. The initial step in the reaction sequence is the ionization of the ketone, which occurs via C—H cleavage at the α-position. The newly formed carbanion is concentrated at the interface, thereby promoting nucleophilic attack on the C-center of CO_2, which is the key step in the reaction. The reaction rate is influenced by several factors, including the concentration of the reactants, the pressure of the CO_2 gas, and the distance traveled by the droplets. Preliminary theoretical calculations suggest that the presence of water in the microdroplets is conducive to this unique chemical reaction. Moreover, this microdroplet strategy has been expanded to encompass seven ketones. This work demonstrates a green route to reducing CO_2 to useful carboxylated organic compounds, thus showing great potential for converting environmental CO_2 into useful materials. This has significant ramifications for the comprehension of gas–liquid interfacial chemistry in confined spaces, including clouds, aerosols, raindrops and mists.

Chlorine oxides have been identified as playing a significant role in the depletion of the ozone layer, with the final oxidation step of chlorine oxides resulting in the production of chloric acid ($HClO_3$) or perchloric acid ($HClO_4$). In their investigation, Francisco and colleagues examined the solvation and reactive adsorption of three stable isomers of chlorine trioxide (Cl_2O_3), namely ClOCl(O)O, $ClClO_3$, and ClOOOCl, at the air–water interface [130]. To do so, they employed a combination of classical and hybrid quantum mechanics/molecular mechanics (QM/MM) molecular dynamics (MD) methods, in conjunction with advanced free energy approaches. The free energy profiles for the hydrolysis reactions of the three isomers were calculated, and reactive adsorption of Cl_2O_3 on aerosols was revealed, as well as the formation of ClO_3^- or $HClO_3$. The results provide evidence for the hypothesis that $HClO_3$ functions as a sink for chlorine oxides. Furthermore, they provide theoretical support for the widespread distribution of $HClO_3$ observed in the Arctic.

It has been demonstrated that the chemical reactions of pyruvate occur in microdroplets but are limited in bulk solution under thermal and dark conditions. Continetti, Grassian, and coworkers utilized in situ Raman microscopy as a probe to investigate the chemical reactions of pyruvate in aqueous microdroplets within a relative humidity- and temperature-controlled environmental cell (Figure 11) [131]. In this study, Raman microscopy is

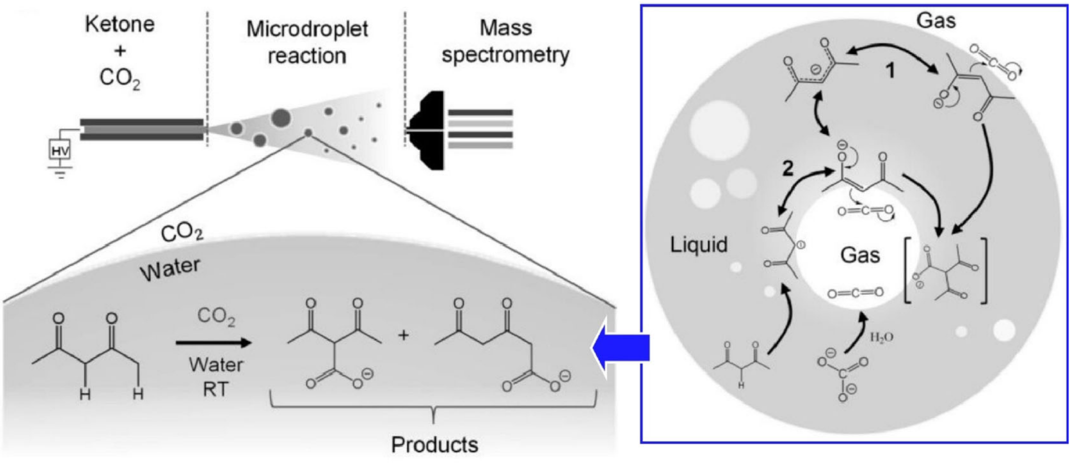

FIGURE 10 | Formation of carbon–carbon bonds at the air–water interface of negatively charged aqueous microdroplets at room temperature, resulting in a catalytic carbon dioxide reduction reaction. Reproduced with permission [129]. Copyright 2024, Wiley-VCH.

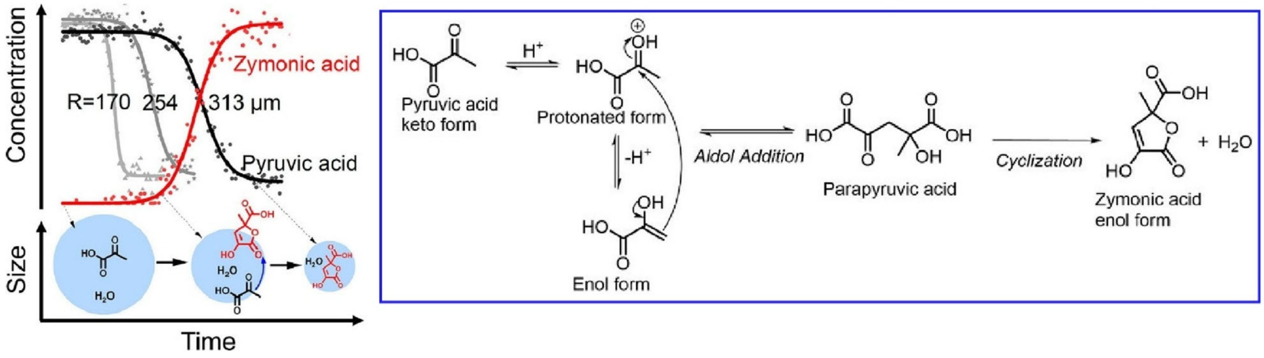

FIGURE 11 | Chemical reactions of pyruvate in aqueous microdroplets within a relative humidity- and temperature-controlled environmental cell, where Raman microscopy is utilized as an in situ probe to investigate the evolution of the chemical composition and size of aqueous pyruvate microdroplets deposited on a hydrophobic substrate. Reproduced under the terms of the CC-BY license [131]. Copyright 2023, American Chemical Society.

utilized as an in situ probe to investigate the evolution of the chemical composition and size of aqueous pyruvate microdroplets deposited on a hydrophobic substrate. As the reaction progresses, the droplet size undergoes dynamic changes in response to alterations in droplet composition and re-equilibration with the relative humidity in the environmental cell. The demonstration of size-dependent sigmoidal kinetics, as observed for the first time in microdroplets, underscores the intricacy of the reaction mechanism and the significance of the air–water interface in the pyruvate condensation reaction. The process of condensation of pyruvate in microdroplets under acidic conditions has been observed to increase with decreasing droplet size and relative humidity. The dynamic properties of droplet composition, size, and surface reactivity, driven in part by the equilibrium pressure of gas-phase HO, produce complex reaction kinetics. Elucidation of these divergent effects within aqueous droplets is imperative to comprehending their reactivity.

The reactive incorporation of bromine nitrate ($BrONO_2$) into aqueous aerosols is a crucial process in atmospheric bromine chemistry. $BrONO_2$ has been observed to form halogen bonds with neighboring water molecules, thereby disrupting the hydrogen-bonding network and potentially resulting in unconventional chemical behavior. Yuan, Wang, and colleagues conducted a study to investigate the solvation and hydrolysis of $BrONO_2$ at the air–water interface [132]. This investigation utilized a combination of quantum chemical calculations, classical molecular dynamics, and ab initio molecular dynamics (AIMD) simulations. Free energy calculations have demonstrated that $BrONO_2$ has a strong tendency to remain at the air–water interface, thereby providing favorable conditions for further hydrolysis. The interplay between halogen bonding and solvation at the air–water interface leads to the spontaneous formation of $H_2OBrONO_2$ from $BrONO_2$. Furthermore, the ion-pair mechanism for the reaction of $H_2OBrONO_2$ was demonstrated to be both kinetically and thermodynamically feasible. These results further emphasize the importance of aerosol interfaces in atmospheric halogen chemistry and provide new insights into the role of halogen bonding in facilitating other molecular reactions at the water interface.

Methane is a potent greenhouse gas, and its atmospheric lifetime is primarily controlled by oxidative reduction. Zhu, Chu, and their coworkers conducted a study on the photooxidation of methane (CH_4) in both gas-phase and microdroplet systems under UV irradiation (Figure 12) [133]. It was reported that the photooxidation of CH_4 at the air–water interface of microdroplets exhibited an order-of-magnitude rate increase compared to the gas-phase reaction. In this study, original N-layer integrated molecular orbital and molecular mechanics calculations were employed to investigate the CH_4 oxidation process, thereby gaining atomic-level insight into the photooxidation mechanism at the air–water interface. One hundred snapshots were uniformly extracted from the last 5 ns of the molecular dynamics simulation trajectory to simulate the absorption spectrum. A combined QM/MM approach was employed: the CH_4 molecule was treated at the quantum mechanical (QM) level and the surrounding water molecules were included in the molecular mechanics (MM) region. The M06–2X functional with the aug-cc-pVTZ basis set was employed in the QM region, while background point charges represented the MM region. All QM/MM calculations were performed using Gaussian09

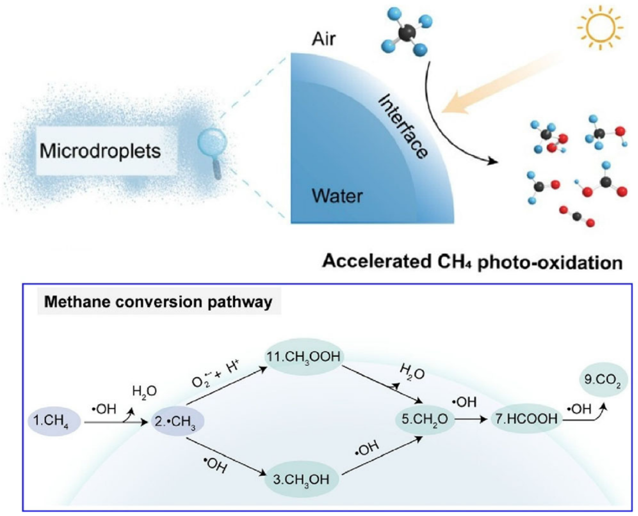

FIGURE 12 | Photooxidation of methane (CH_4) in both gas-phase and microdroplet systems under UV irradiation in which original N-layer integrated molecular orbital and molecular mechanics calculations were employed to investigate the CH_4 oxidation process, thereby gaining atomic-level insight into the photooxidation mechanism at the air–water interface. Reproduced with permission [133]. Copyright 2025, American Chemical Society.

software. The accelerated photochemical oxidation of methane (CH_4) at the air–water interface of microdroplets is primarily attributable to two factors. Firstly, there is the rapid accumulation of hydroxyl radicals ($^{\bullet}OH$) at the interface, which react with methane via a low-energy barrier mechanism to produce methyl radicals ($\bullet CH_3$) with high efficiency. Secondly, there is the spontaneous reaction of methyl radicals at the interface with oxygen (O_2) via a barrierless exothermic process, which ultimately produces significantly higher concentrations of methyl hydroxide (CH_3OOH) compared to the conventional bulk-phase reaction. The identification of a hitherto unrecognized reaction pathway has provided a quantitative understanding of how the collective interactions between photons and microdroplets govern both oxidation rates and product selectivity. These findings provide essential mechanistic evidence for updating methane budget models, particularly with respect to cloud- and aerosol-driven interfacial photochemistry. The transformation of methane (CH_4) at the air–water interface has been shown to be enhanced by microdroplets, a process which has far-reaching implications for a number of atmospheric chemical processes. These include the formation of secondary organic aerosols, the production of acid rain and the mitigation of the greenhouse effect.

The C—F bond is widely regarded as the strongest single bond in organic chemistry. Activation typically necessitates stringent reaction conditions, frequently involving costly transition metal catalysts or aggressive reagents. In stark contrast, Shibata, Banerjee, and coworkers demonstrated a reagent-free defluorination reaction using only water at the air–water interface (Figure 13) [134]. The study observed selective cleavage of Csp^2—F and Csp^3—F bonds when aqueous solutions of organofluorine substrates (ArF and ArCF) were sprayed into air. The generation of highly reactive carbocation species and fluoride anions occurred at the air–water interface. The carbocations that were formed at the air–water interface subsequently reacted with various nucleophiles to generate defluorination products. The one-electron reduction of aromatic substrates has been shown to promote rapid heterolytic cleavage of C—F bonds, occurring in less than 1 ms. The elimination of fluorine from organofluorine compounds by water has been demonstrated to be an effective process. This reaction is characterized by its independence from catalysts or reagents, relying exclusively on water as a solvent. This characteristic opens up a novel pathway for sustainable and environmentally friendly C—F bond activation. This work serves to expand the existing scope of microdroplet chemistry and to establish new avenues for the utilization of aqueous interfaces as catalysts for unconventional transformations of fluoroarenes.

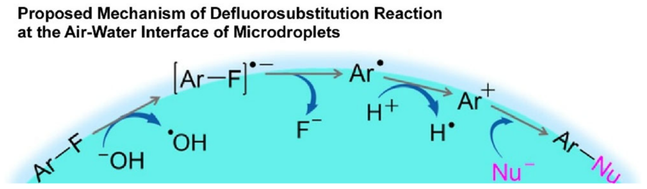

FIGURE 13 | Reagent-free defluorination reaction using only water at the air–water interface. Reproduced with permission [134]. Copyright 2025, American Chemical Society.

Feng and coworkers have proposed a methodology for the emulation of chemical reactions on water surfaces, utilizing micelles (Figure 14) [135]. In this strategy, a hydrophobic core and an aqueous layer form an interface, thereby creating an environment that promotes chemical reactions similar to those on water surfaces. Utilizing the water-surface interface of these micelles, the researchers successfully demonstrated the reactivity and site selectivity of 14 reversible and irreversible chemical reactions. By extending this method of confining monomers with preorganized structures on the micellar surface, the researchers further demonstrated the applicability of the method to two-dimensional polymerization at the micellar interface. The synthesis of thin crystalline 2D polymer layers in aqueous solution was successfully achieved. Uniform circular sheet size distributions with micrometer- and nanometer-scale thicknesses were achieved. This strategy has the potential to considerably extend the applicability of water-surface chemistry to a broad spectrum of chemical syntheses. This approach introduces a method to extend underwater surface chemistry to upscalable aqueous synthesis, thereby broadening the accessibility of underwater surface chemistry to a wide range of chemical and material synthesis.

Chemical reactions occurring on liquid surfaces offer certain advantages, including enhanced reactivity, well-defined selectivity, and a defined reaction space. Localization of reaction species at liquid interfaces also provide precious opportunities to analyze reaction mechanisms with specifying reaction intermediates. In addition, the development of reaction sites that exploit these properties beyond the macroscopic air–water interface is also highly desirable. Such systems can be recognized as good models for environmental situations. Analyses of reaction mechanisms at such microscopic liquid interfaces would give useful information for environmental remediations.

5 | Molecular Machine and Microrobot on Liquid Interface

As demonstrated in the preceding sections, the subtle environmental variations in proximity to an interface have been shown to exert a significant influence on the stability, intermolecular interactions, and intermolecular reactivity of molecules and materials. This phenomenon can be attributed to the presence of electronic effects, such as the substantial change in the dielectric constant of a medium when observed perpendicularly to an environment, such as an air–water interface. In this section, the role of the in-interface (lateral) direction is considered. It is evident that dynamic interfaces, including liquid interfaces, are capable of macroscopic deformation, measured in tens of centimeters, along the film plane direction. This movement can be coupled with molecular-level phenomena in the film thickness direction. Utilizing this fundamental principle enables the manipulation of molecular-level objects at a liquid interface through macroscopic mechanical actions [136, 137].

As illustrated in Figure 15, a molecular machine deployed on the water surface can be controlled with macroscopic movements analogous to those of the hand [138, 139]. The used molecular machine (steroid cyclophane) consists of four hydrophilic cholic acid units, which are connected to a ring substance. The machine

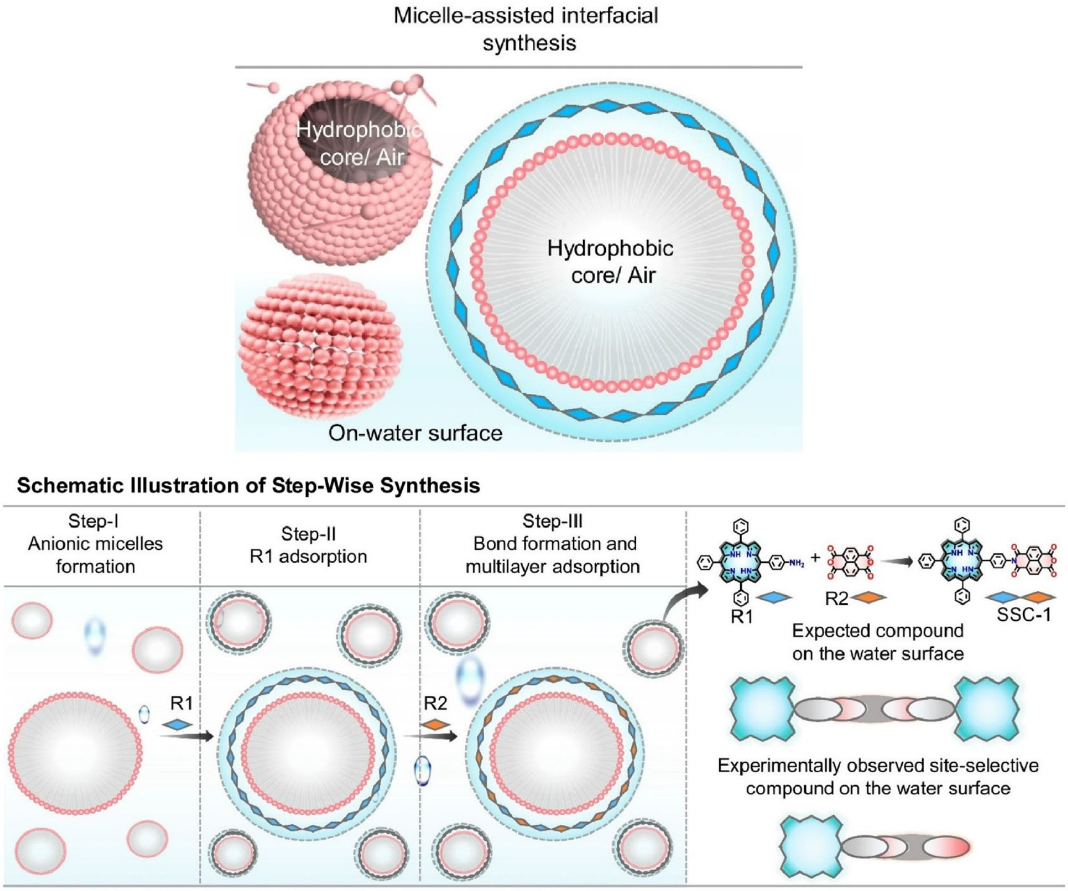

FIGURE 14 | Methodology for the emulation of chemical reactions on water surfaces, utilizing micelles (top), where, a hydrophobic core and an aqueous layer form an interface, thereby creating an environment that promotes chemical reactions similar to those on water surfaces See step-wise synthesis at bottom). Reproduced under the terms of the CC-BY license [135]. Copyright 2024, Springer-Nature.

is spread out and attached to the water surface. In the event of lateral compression of a monolayer composed of the aforementioned molecular machine, the molecular machine undergoes closure, resulting in the formation of a cavity structure. At this time, the molecular machine exhibits the capacity to capture guest molecules that are dissolved in the water phase. The molecular machine monolayer at the air–water interface can be manipulated through macroscopic compression and expansion, enabling the molecular machine to grasp and release guest molecules. The employment of a liquid interface enables the precise control of the molecular machine through macroscopic manual manipulation.

It has been demonstrated that this technique can also be used to adjust the conformation of molecular receptors continuously. The molecular conformation of the receptor can be modified through the application of lateral compression to the monolayer, which is spread at the air–water interface. This technique facilitates the unrestrained modification of the chiral recognition of amino acids [140], and the distinction between thymine and uracil derivatives that differ by a single methyl group [141]. It can be concluded that the configuration of molecular receptors can be adjusted at liquid interfaces in order to function in an optimal manner.

Figure 16 is a classification of molecular recognition modes [142]. The most prevalent approach entails the identification of a single state in which the host–guest bond structure exhibits the greatest degree of energetic stability. This is the basis of supramolecular chemistry, for which the Nobel Prize in Chemistry was awarded in 1987. An advanced version of this is the switching of host structures in response to external stimuli. The process entails the generation of multiple states, which facilitate the transition between molecular recognition. This fundamental principle underlies the operation of molecular machines, a field that was recognized with the Nobel Prize in Chemistry in 2016. Conversely, the conformational control of molecular receptors on liquid surfaces can be regarded as a form of tuning [143]. By tuning to generate all of the infinite possible molecular structures, it is possible to select the desired structure from among the many possibilities. This is distinct from the previous single-state and several-state modes.

A re-evaluation of the force required to effect changes in molecular structure is considered. Molecular structure changes, such as those caused by photoisomerization, which are utilized in organic chemistry, require forces of the order of several thousands of pN. In contrast, molecular changes in biological systems, such as myosin walking, are caused by extremely delicate forces of several to several tens of pN. The force required for molecular manipulation at the air–water interface is calculated to be in the range of 0.5–35 pN, which is comparable to the delicate mechanical manipulation of molecular deformation in biological systems

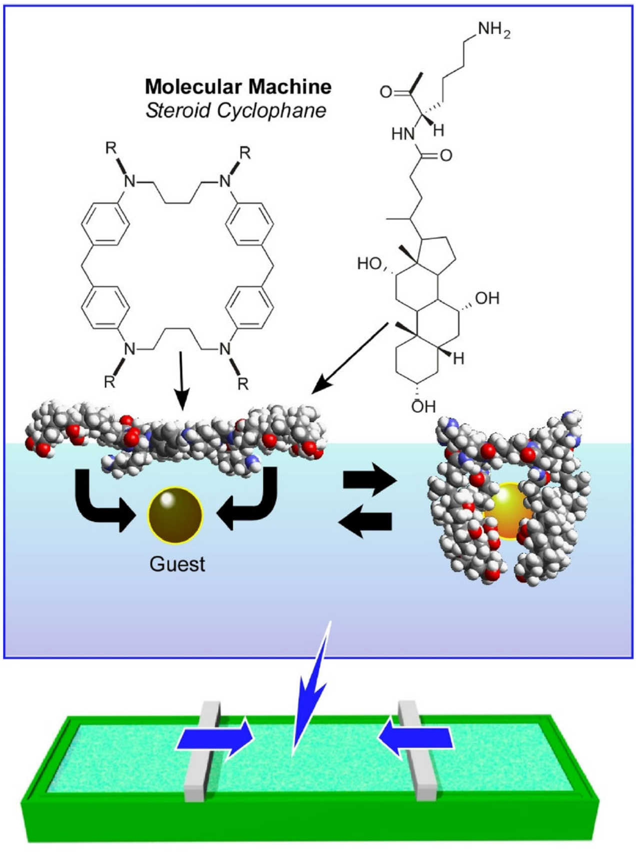

FIGURE 15 | Molecular machine (steroid cyclophane) on water surface exhibits the capacity to capture guest molecules dissolved in the water phase upon lateral compression of its monolayer.

[144]. Consequently, the function of the liquid interface in relation to the membrane surface is to facilitate precise mechanical manipulation.

The manipulation of matter at liquid interfaces is not limited to molecular machines or molecular receptors; it can also be applied to a variety of objects. The following examples are provided to illustrate this phenomenon. A further investigation was conducted into the effect of interfacial tension imbalance on microgel monolayers. Karg and coworkers conducted a systematic study of the behavior of monolayers at the air–water interface, controlled by the addition of sodium dodecyl sulfate or linear poly-*N*-isopropylacrylamide homopolymer (Figure 17) [145]. The fabrication of freely floating core–shell microgel monolayers can be achieved by means of direct injection of a microgel-containing spreading solution into the air–water interface. Depending on the interfacial tension imbalance, the monolayer undergoes a change in morphology, either expanding, contracting, or maintaining its original form. The structural and dynamic characteristics of freely suspended core–shell microgel monolayers at the air–water interface were found to be governed by the external interfacial tension, defined as the interfacial tension external to the region encompassed by the monolayer. This finding may provide a new strategy for controlling the interparticle distance of microgel monolayers at fluid interfaces, replacing conventional Langmuir trough devices, which control the state of the monolayer by compression through a movable barrier.

The prospect of manipulating hydrogel microrobots through the utilization of multiphysical fields has garnered significant attention, particularly with regard to their potential applications in the domains of biomedical engineering and environmental exploration. Luo, Liu, Yang, and coworkers conducted a study on the multimodal locomotion and dynamic interactions of hydrogel microrobots at the air–water interface under magnetic and optical stimuli (Figure 18) [146]. The synthesis of magnetically and photoresponsive disk-shaped hydrogel microrobots was accomplished through the utilization of a prepolymer solution and Fe_3O_4 nanoparticles. Through the fabrication of hydrogel microdisks with varying dimensions and magnetic particle

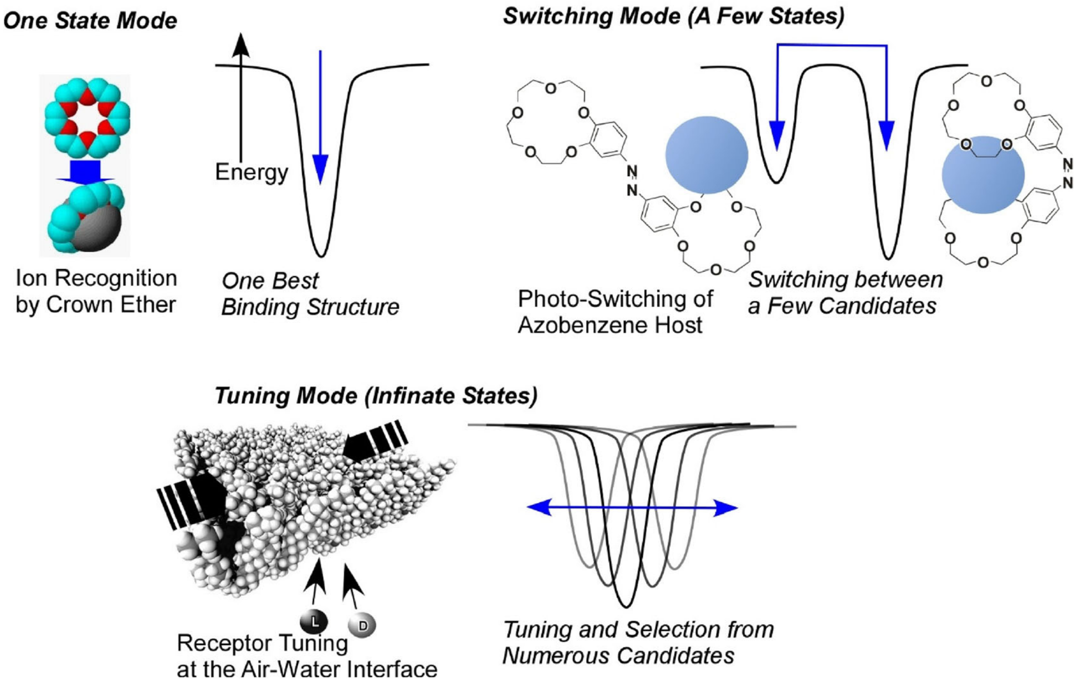

FIGURE 16 | Classification of molecular recognition modes: one state modes, switching mode, and tuning mode.

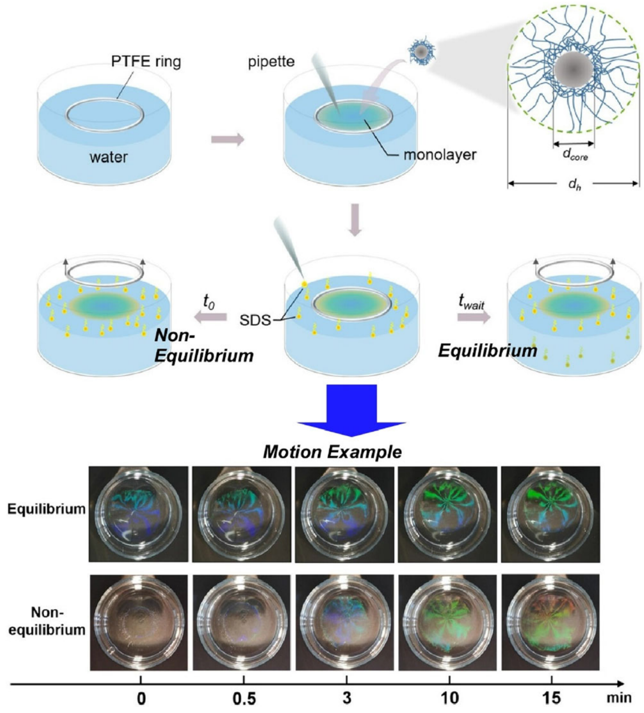

FIGURE 17 | Freely floating core–shell microgel monolayers formed by means of direct injection of a microgel-containing spreading solution into the air–water interface undergo a change in morphology, either expanding, contracting, or maintaining its original form depending on the interfacial tension imbalance: preparation (top) and motion examples (bottom). Reproduced under the terms of the CC-BY license [145]. Copyright 2025, American Chemical Society.

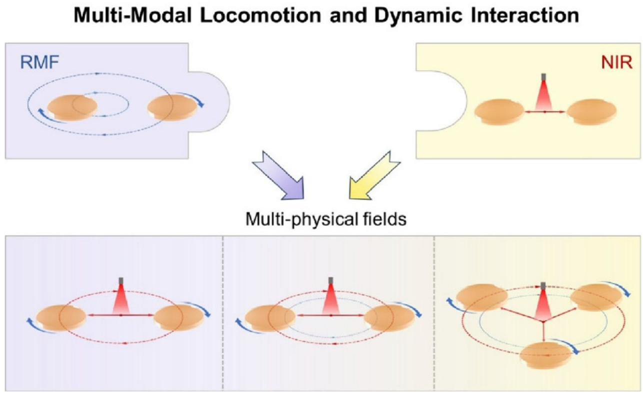

FIGURE 18 | The multimodal locomotion and dynamic interactions of hydrogel microrobots at the air–water interface under a rotating magnetic field (RMF) and near-infrared (NIR) light. Reproduced with permission [146]. Copyright 2024, American Chemical Society.

concentrations, a range of behaviors was observed and substantiated. In particular, the hydrogel microdisks exhibited remarkable responsiveness to both a rotating magnetic field (RMF) and near-infrared (NIR) light. The influence of the RMF on the behavior of a pair of hydrogel microrobots located at the air–water interface was observed to result in a transition from a coordinated complex rotation to an interactive behavior involving both rotation and revolution. Concurrently, under NIR illumination, a transition from attraction to separation was observed, thereby demonstrating dynamic behavioral changes in response to divergent stimuli. Specifically, the behavioral patterns exhibited by multiple hydrogel microrobots under a multiphysical field demonstrate the capacity of NIR light to enhance interactive motion under RMF, thereby extending the range of motion trajectories. The utilization of the characteristics of multiphysical field driving facilitates a transition from cooperative motion to interactive motion among multiple hydrogel microdisks, thereby enhancing the manipulation ability and efficiency of these hydrogel microrobots.

The conception and development of intelligent robots capable of unrestricted movement on water, emulating the behavior of aquatic organisms, represents a highly appealing objective. Xie and coworkers have developed an underwater robot resembling a water strider that does not require assembly (Figure 19) [147]. This robot is composed of a single layer of light-programmable liquid-crystal elastomer. The light-programmable liquid-crystal elastomer was designed as a four-legged structure, with the front and back legs individually programmable by light. The strider demonstrated self-oscillation and self-propulsion behavior on water with a temperature gradient at the air–water interface, owing to an imbalance in contact area and tension between the legs and the water. Following the implementation of selective coating, the light-programmable liquid-crystal elastomer strider was subjected to local and remote heating, thereby enabling the modulation of its trajectory. The trajectory was successfully modeled, with the navigation process being replicated with precision. This mimics the fundamental survival technique employed by natural water striders. This design method will provide a foundation for the conceptualization of other types of soft intelligent machines.

The capacity of mobile microrobots to navigate, sense, and interact with their environments is well documented. This finding has the potential to transform the fields of biomedicine and environmental remediation. The utilization of self-organizing swarms of microrobots has been demonstrated to be advantageous for operating in complex environments. Wang, Petersen, Sitti, and coworkers conducted a study into a microrobot assembly system that is capable of on-demand reconfiguration [148]. This system is able to adapt and exploit its environment in order to perform a variety of functions at the air–water interface (Figure 20). A group of approximately 120 microdisks was subjected to an externally applied magnetic field that varied over time. The system demonstrated a range of modes, transitioning between isotropic and anisotropic states, and between globally driven and self-propelled behaviors. These behaviors are enabled by several particle–particle and hydrodynamic, capillary, and magnetic dipole–dipole interactions, which are controlled by an external magnetic field. The potential exists for its eventual transformation into a three-dimensional system for utilization in microscale packaging and medical applications, such as targeted delivery of active drugs or other cargoes. Its use as a reconfigurable micromachine in biomedical and environmental applications is also valuable.

Dynamic interfaces, such as liquid interfaces, exhibit macroscopic deformation in the film plane. This movement can be coupled with molecular-level phenomena in the film thickness direction. The utilization of this fundamental principle enables

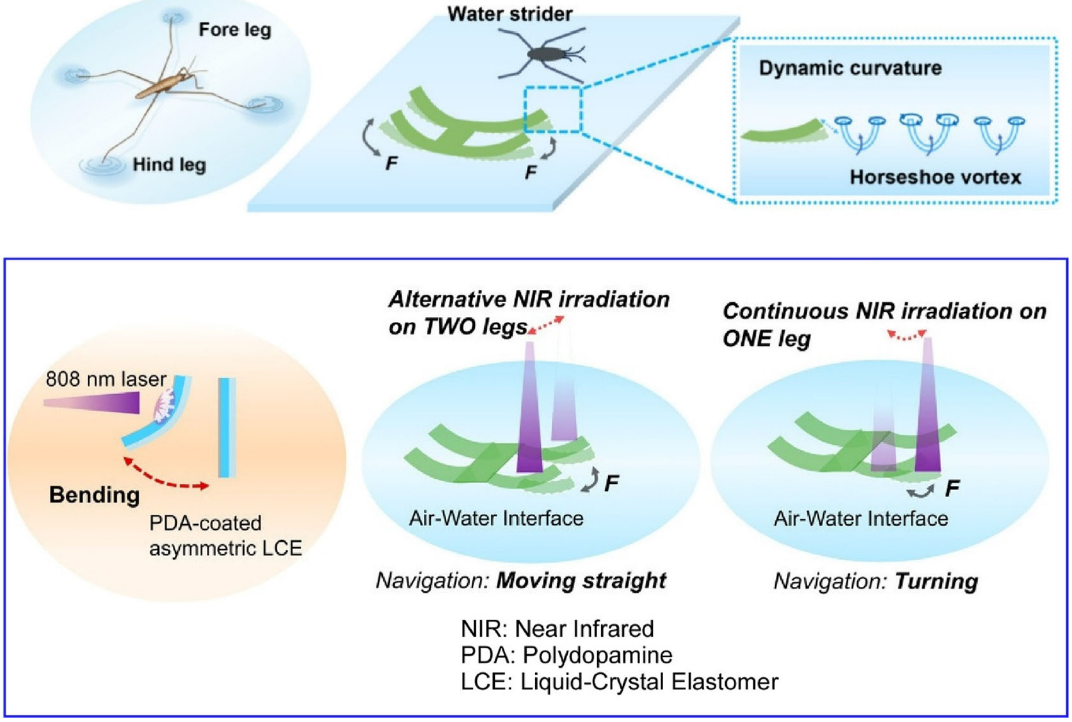

FIGURE 19 | Underwater robot resembling a water strider composed of a single layer of light-programmable liquid-crystal elastomer, in which the light-programmable liquid-crystal elastomer was designed as a four-legged structure, with the front and back legs individually programmable by light. Reproduced with permission [147]. Copyright 2025, American Chemical Society.

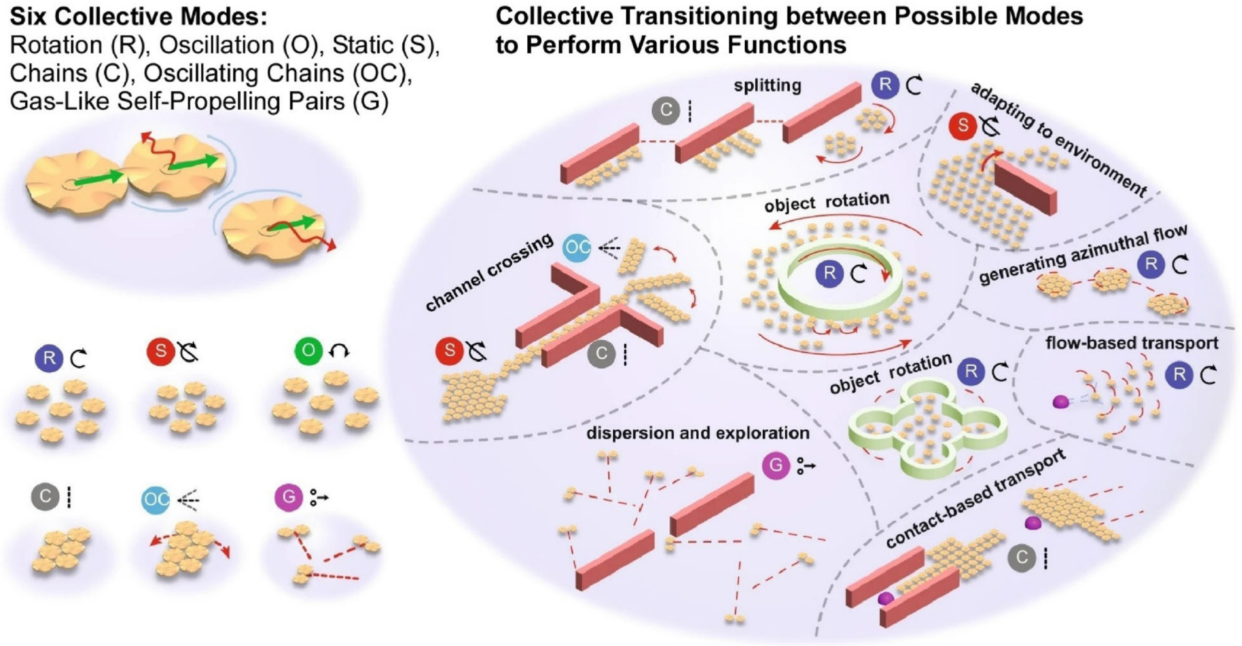

FIGURE 20 | Microrobot assembly systems capable of on-demand reconfiguration to perform a variety of functions at the air–water interface where a group of approximately 120 microdisks is subjected to an externally applied magnetic field with a range of modes, transitioning between isotropic and anisotropic states, and between globally driven and self-propelled behaviors. Reproduced under the terms of the CC-BY license [148]. Copyright 2022, Springer-Nature.

the manipulation of molecular-level objects through macroscopic movement at a liquid interface. The forces required for molecular manipulation at an air–water interface are analogous to the delicate mechanical manipulation of molecular deformation in biological systems. It can thus be concluded that the function of a liquid interface in the film plane direction is to facilitate precise mechanical control. Macroscopic movements, such as those of a hand, have been shown to be capable of manipulating molecular machines when they are deployed on the water surface. The manipulation of objects at liquid interfaces is not limited to molecular machines or molecular receptors, but is also applicable to a variety of other objects. The movement of microgels, microrobots, or their assemblies can be controlled. The function of a liquid interface in the lateral direction is to regulate delicate mechanical manipulation.

6 | Life at Liquid Interface

The forces required for molecular manipulation at the air–water interface are analogous to the delicate mechanical manipulation of molecular deformation in biological systems. It is hypothesized that this may also be useful for the control of more complex biological systems. Indeed, liquid interfaces, including those between liquid and liquid or liquid and gas, have the potential to contribute to a variety of biological control applications, such as cell culture and the associated control of cell differentiation. The interface between two immiscible liquids is well utilized as a cell culture environment. In particular, perfluorocarbons, defined as an organic solvent that does not adversely affect cells, are utilized as an organic solvent phase [149]. Furthermore, it has been determined that at such liquid–liquid interfaces, spontaneous nanoarchitectonics are formed, resulting in structures analogous to molecular membranes of proteins, which in turn influence cell differentiation.

For instance, Jia et al. investigated the effects of two-dimensional protein nanofibril networks spontaneously formed at the liquid–liquid interface between aqueous culture medium and fluorocarbons on stem cells [150]. The researchers discovered that the processes of lipid raft formation and focal adhesion kinase (FAK) phosphorylation specifically impacted stem cell differentiation at the liquid–liquid interface. The integration of downstream FAK-mediated signaling within lipid rafts in membrane microdomains results in the induction of stem cell neurogenesis. In the context of cell culture at the liquid–liquid interface, the utilization of cell culture on small oil droplets emerges as an environmentally sustainable alternative to the employment of microplastics. Gautrot and coworkers fabricated highly elastic protein nanosheets by coassembling supercharged albumin with the surfactant pentafluorobenzoyl chloride [151]. It was found that the protein nanosheets formed at this liquid interface mediated the adsorption of extracellular matrix proteins and cell adhesion. These proteins act as structural elements to define the liquid–liquid interface and as a substrate to capture extracellular matrix molecules. The methodology outlined in this study has resulted in the development of a system that functions without the use of plastics or microplastics.

The lung epithelium is exposed to various atmospheric pollutants through the process of respiration. The utilization of a robust in vitro lung epithelial model is imperative for the elucidation of the mechanisms and treatments of respiratory diseases caused by these pollutants. Tanabe and Ishikawa developed an in vitro alveolar epithelial model using primary human pulmonary alveolar epithelial cells at the air–liquid interface (Figure 21) [152]. Primary human pulmonary alveolar epithelial cells were successfully cultured at the air–liquid interface for a period of 28 days in a medium that had been supplemented with three small molecules. A multitude of cells expressed alveolar type 2 cell markers, including surfactant protein B and prosurfactant protein C. This air–liquid interface culture model, incorporating differentiated alveolar type 2 cells, serves as a valuable tool for elucidating the mechanisms and treatments of respiratory diseases. It has been established that alveolar type 2 cells are of significant importance in the development of various respiratory diseases. Therefore, this differentiated alveolar type 2 cell model has the potential to serve as a valuable instrument in the study of the mechanisms and treatments of respiratory diseases.

Interfaces between water-immiscible hydrophobic fluids, such as perfluorocarbons and silicones, could be an alternative to conventional plastic dishes for cell attachment and growth. In this regard, it is imperative to deliberate the array of mechanical and chemical stimulants to which cells are subjected. Nakanishi, Sodeyama, and coworkers utilized a data-driven approach to identify noncytotoxic ionic liquids as fluid culture platforms (Figure 22) [153]. The objective is to guarantee repeated utilization due to their elevated thermal stability prior to biological applications. The employment of data-driven methodologies has been demonstrated to expedite research processes by facilitating the identification of noncytotoxic ionic liquids through a guided experimental design approach. This approach is characterized by its ability to minimize the number of experiments required, thereby reducing the time and resources necessary for research and development. In particular, they successfully cultured human mesenchymal stem cells at trihexylethylammonium trifluoromethylsulfonylimide interfaces, allowing for repeated use after solvent extraction and heat sterilization. An exploration was conducted into the potential for new sustainable cell culture systems that fulfill the three Rs (Reduce, Recycle and Replace). It is anticipated that such liquid interface-based methodologies will supersede petroleum-derived products in the context of a sustainable society.

Life activity controls at liquid interfaces are not limited to the above-mentioned examples. Delicate nanoarchitectonics at liquid interfaces enables delicate multicomponent interaction, cell communications, and cell fate modulations. Many examples have been reported in these years. These developments may contribute well to biomedical applications and further understanding of bioprocesses. Liquid interfaces provide nice media for formation of cell communities and organoid construction. Ma, Li, and coworkers reported generation of human endometrial assembloids with a luminal epithelium at the air–liquid interface [154]. The investigated system can create transcriptionally defined endometrial cell subpopulations, which may be useful for further studies on embryo implantation, and endometrial growth, differentiation, and disease. Similarly, liquid interfacial systems are useful for forming intestinal organoid structures, including the regulation of organoid polarity and cell apoptosis. Kishikawa and coworkers successfully fabricated model systems for intestinal epithelial function and polarity using human induced pluripotent stem cells (iPSCs) with air–liquid interface culture systems [155]. They regenerated polarized monolayers of intestinal epithelium with key features of the human intestine, including absorptive and secretory cell types. The liquid interfacial system

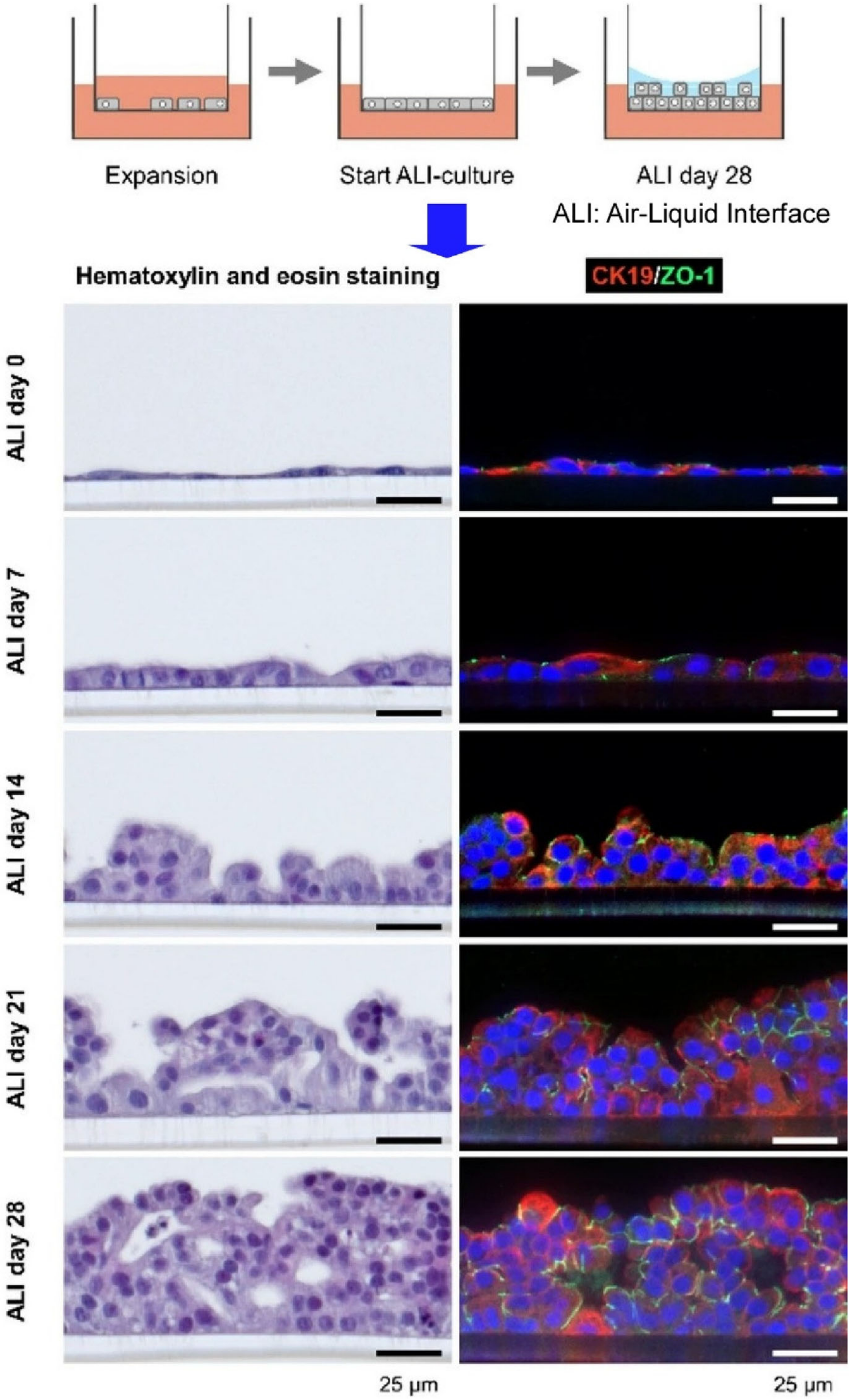

FIGURE 21 | An in vitro alveolar epithelial model using primary human pulmonary alveolar epithelial cells at the air–liquid interface in which primary human pulmonary alveolar epithelial cells were successfully cultured at the air–liquid interface for a period of 28 days in a medium that had been supplemented with three small molecules. Reproduced under the terms of the CC-BY license [152]. Copyright 2025, Springer-Nature.

enables delicate control of cell treatment from both sides. Indomethacin treatment from the basal side induced significant mucosal injury and inflammatory gene expression, in contrast to treatment from the apical side. Dunn and coworkers investigated the effect of the air–liquid interface on cultured human intestinal epithelial cells [156]. Compared with other systems, the human intestinal monolayer at the air–water interface exhibited a morphology more similar to normal intestinal epithelium. This is probably due to the ability to control surface tension delicately; fewer apoptotic cells were observed in air–liquid conditions. The monolayer of cells expressed higher levels of secretory cell lineage genes at the air–liquid interface. Nanoarchitectonics at liquid interfaces can be advantageous for controlling interfacial topological structures and spatial regulations, which can then be used to further modulate various cell activities and intercellular communications. Singh and coworkers investigated the effects of

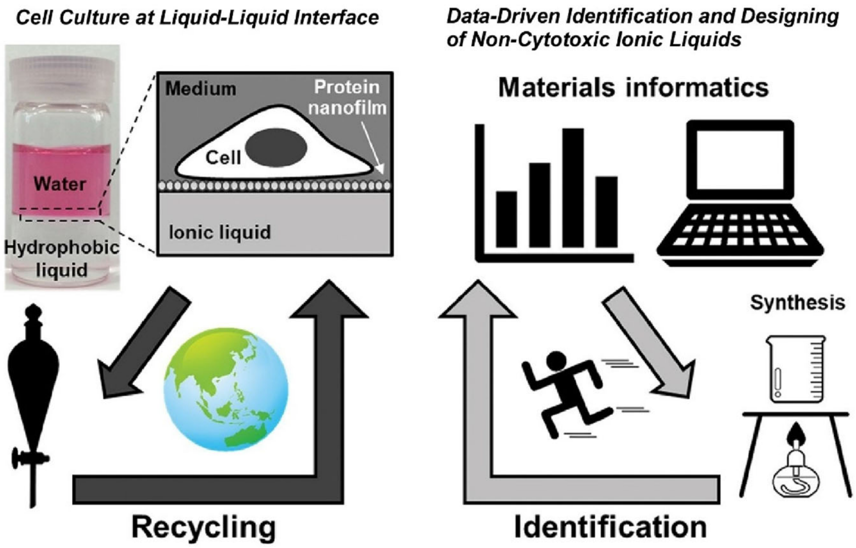

FIGURE 22 | Data-driven approach to identify noncytotoxic ionic liquids as fluid culture platforms for liquid–liquid interfacial cell culture. Reproduced under the terms of the CC-BY license [153]. Copyright 2024, Taylor & Francis.

lipid-rich domains in human colonoid monolayers at the air–liquid interface [157]. Exposure of normal human colonoid monolayers to the air–liquid interface increased acid sphingomyelinase activity and enhanced colonoid differentiation, as well as basal and forskolin-stimulated downregulation in adenoma activities. Wang, Huang, and coworkers reported enhanced transmembrane delivery in interactive protocell communities through liquid–liquid phase separation using liquid droplet systems [158]. Delicate control of protocell interfacial morphologies can regulate the pathway for the transmembrane transport of biomacromolecules. This could be a powerful technique for inducing higher-order behavior in synthetic protocell communities.

The investigation encompasses cell culture at air–liquid and liquid–liquid interfaces. Despite the fact that only a limited number of examples are displayed individually, a variety of complex biocontrols, such as cell differentiation control, are being carried out. It is hypothesized that delicate intermolecular interactions are at work at the liquid interface. The utilization of liquid interfaces for the purpose of cell control is of both academic and practical significance. This technology will also lead to the elimination of unnecessary plastic materials, which will contribute to the solution of the microplastic material problem.

7 | Future Perspectives

This review paper examines a number of research examples and consider the role of liquid interfaces. In summary, as demonstrated in Figure 23, it is evident that liquid interfaces assume varied functions, contingent upon their orientation relative to the interface. Perpendicular to the interface, electronic properties are delicately tuned in response to changes in the dielectric constant. This enables control of intermolecular interactions. This finding elucidates the basis for the occurrence of hydrogen-bond-based functions within biological systems, which are, in essence, aqueous solutions. Concurrently, delicate forces can be applied in the direction within the liquid interface, thereby enabling fine structural adjustment of molecular structures analogous to those found in living organisms. As was outlined in the introduction, the ultimate functional material, which could be considered a material of the future, will be one in which functional elements similar to those found in biological systems are rationally and softly assembled. While highly ordered materials or controlled structures, such as crystals, are often considered to be high-performance materials, the ultimate high-performance material may be one with low order and low symmetry, similar to those found in biological systems. This process gives rise to asymmetric energy transfer and information transmission. Liquid interfaces, which function through mechanisms analogous to those observed in biological systems, may provide optimal conditions for the creation of such future materials.

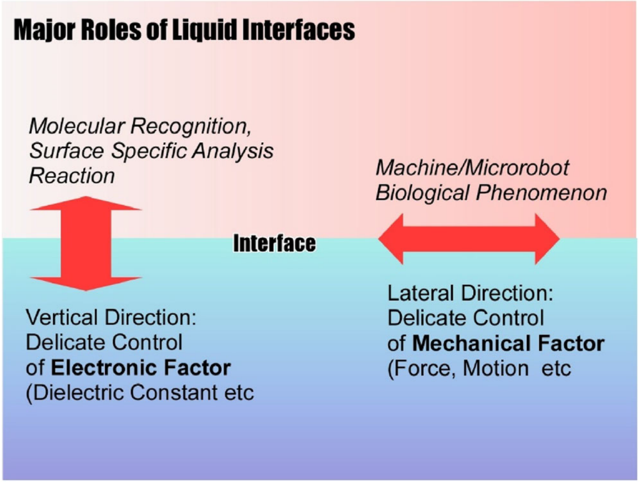

FIGURE 23 | Roles of liquid interfaces for delicate nanoarchitectonics: delicate control of electronic factor (vertical direction) and delicate control of mechanical factor (lateral direction).

In addition to the examples presented here, a variety of nanoarchitectonics can be conducted at liquid interfaces. For instance, the synthesis of fullerene assemblies can be achieved by employing an environment comprising immiscible liquids of varying polarities. Furthermore, the morphology can be varied by combining liquids. This phenomenon can be regarded as a form of delicate structural control at liquid interfaces [159, 160]. Moreover, the application of heat treatment to these aggregates enables the control of the carbon frame structure. The creation of carbon pentagon structures and control of spin behavior can enhance the oxygen reduction reaction (ORR) [161, 162]. Furthermore, the coupling of chemical equilibrium at liquid interfaces enables precise regulation of the doping of organic semiconductors [163]. Liquid interfaces are places where delicate nanoarchitectonics and functional expression can occur, not limited to the examples shown in this paper.

The paucity of material development in these directions is currently due to the inability to successfully incorporate multiple components. Should a significant proportion of functional elements be capable of assembly in a rational and soft manner, it may be feasible to create functional materials that exhibit comparable properties to those found in living organisms. The creation of multicomponent structures using limited theories and experience constitutes a challenging endeavor. The potential solution to this predicament may be found in the utilization of AI [164–168]. The synthesis of extant experimental data and the utilization of AI may facilitate the development of functional systems with rational multicomponent structures at liquid interfaces. Use of AI has various potentials to analyze and design interfacial systems. AI can predict interface reaction products. AI can optimize structural parameters of dynamic functional systems including molecular machine operations at the air–water interface. AI can specify design of liquid interface for desirable cell culture systems. However, several current technical bottlenecks of AI applications such as insufficient data volume and difficulties in modeling multicomponent systems have to be eventually solved. Especially, the scarcity of interface interaction data for multicomponent systems have to be fixed through the establishment of cross-laboratory dataset standards for active assembly of useful data. Using AI to predict the assembly morphology of new molecules at the gas–liquid interface based on existing interface reaction data, communication systems between multiple functional components can be designed with rational expectation. Such approaches can potentially create artificial signal transduction systems at liquid interfaces. More sophisticated interfacial organization of multiple components would establish interfacial assemblies with organized electron and energy transfers, which may be used for artificial photosynthesis. Such systems could potentially result in the creation of future materials with functions comparable to those of living organisms. This initiative employs chemistry as its primary technological framework to synthesize materials at the nanoscale. The subject can be considered to represent a fusion of chemical (Chem) and nanotechnological (Nano) into materials (Mat) creation. This paper sets out the unexpectedly important roles that liquid interfaces can play in this context.

Acknowledgments

This study was partially supported by Japan Society for the Promotion of Science KAKENHI (grant numbers JP23H05459 and JP25H00898).

Funding

This work was supported by Japan Society for the Promotion of Science KAKENHI (JP23H05459 and JP25H00898).

Conflicts of Interest

The author declares no conflicts of interest.

References

1. I. R. Vetter and A. Wittinghofer, “The Guanine Nucleotide-Binding Switch in Three Dimensions,” *Science* 294 (2001): 1299.

2. T. M. Iverson, K. Maghlaoui, J. Barber, and S. Iwata, “Architecture of the Photosynthetic Oxygen-Evolving Center,” *Science* 303 (2004): 1831.

3. C. Xu, X. Pi, Y. Huang, et al., “Structural Basis for Energy Transfer in a Huge Diatom PSI-FCPI Supercomplex,” *Nature Communications* 11 (2020): 5081.

4. M. Sugiyama, M. Akiyama, Y. Yonezawa, et al., “Electron in a Cube: Synthesis and Characterization of Perfluorocubane as an Electron Acceptor,” *Science* 377 (2022): 756.

5. K. Shibuya, A. Chikamatsu, K. Hisano, and H. Nishino, “Synthesis of Dioxapropellanes Using Mn(III)-Based Oxidative Tandem Cyclization of Tetracarbonyl Compounds with Diarylethenes,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae011.

6. K. Fuchibe, T. Fujita, and J. Ichikawa, “Pinpoint-Fluorinated Polycyclic Aromatic Hydrocarbons (F-PAHs) and Their Heteroaromatic Analogs: Syntheses, Reactivities, and Properties,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoad024.

7. Y. Inokuma, “Discrete Polyketones: Synthesis, Derivatization, and Potential Applications,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae072.

8. H. Shudo, P. Wiesener, E. Kolodzeiski, et al., “Thiophene-Fused Aromatic Belts,” *Nature Communications* 16 (2025): 1074.

9. A. H. Khan, S. Ghosh, B. Pradhan, et al., “Two-Dimensional (2D) Nanomaterials towards Electrochemical Nanoarchitectonics in Energy-Related Applications,” *Bulletin of the Chemical Society of Japan* 90 (2017): 627.

10. G. Chen, S. K. Singh, K. Takeyasu, J. P. Hill, J. Nakamura, and K. Ariga, “Versatile Nanoarchitectonics of Pt with Morphology Control of Oxygen Reduction Reaction Catalysts,” *Science and Technology of Advanced Materials* 23 (2022): 413.

11. K. Sasaki, “Prospects for β-Ga_2O_3: Now and into the Future,” *Applied Physics Express* 17 (2024): 090101.

12. M. Minakshi, A. Samayamanthry, J. Whale, et al., “Phosphorous – Containing Activated Carbon Derived From Natural Honeydew Peel Powers Aqueous Supercapacitors,” *Chemistry – An Asian Journal* 19 (2024): e202400622.

13. M. Ghosh, R. Sarma, M. Kamiyama, T. Kawawaki, S. Biswas, and Y. Negishi, “Next-Generation CO_2 Electroreduction: The Role of Atomically Precise Nanoclusters and Emerging Catalytic Strategies,” *Nanoscale Horizons* 10 (2025): 1250.

14. J. Kim, J. H. Kim, and K. Ariga, “Redox-Active Polymers for Energy Storage Nanoarchitectonics,” *Joule* 1 (2017): 739.

15. T. Yamamoto, A. Takahashi, and H. Otsuka, “Mechanochromic Polymers Based on Radical-Type Dynamic Covalent Chemistry,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoad004.

16. S. Zheng, N. Ning, Y. Hua, and Z. Gao, “A Novel Side-Chain Type Double-Cation Grafted Poly (Binaphthyl Triphenyl Piperidine) Membranes for Anion Exchange Membrane Fuel Cells,” *ChemNanoMat* 10 (2024): e202300542.

17. M. Kamigaito, “Step-Growth Irreversible Deactivation Radical Polymerization: Synergistic Developments with Chain-Growth Reversible Deactivation Radical Polymerization,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae069.

18. T. Ikai, T. Matsumoto, S. Takeda, K. Oki, and E. Yashima, “Tunable Synthesis of One-Handed Helical Ladder Polymers with a Helical Cavity or Helical Grooves for Advanced Chiral Recognition,” *Macromolecules* 2025 (58): 6943.

19. Y. Yan, Y. Chen, Z. Mao, et al., “Bridging the Thermal Divide: Nano-Architectonics and Interface Engineering Strategies for High-Performance 2D Material-Based Polymer Composites,” *ACS Nano* 19 (2025): 20357.

20. H. Fujimoto, T. Hirao, and T. Haino, “Supramolecular Polymerization Behavior of a Ditopic Self-Folding Biscavitand,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoad016.

21. A. T. N. Doan and K. Sakurai, “Unraveling the Molecular Mechanism of the Interaction Between Cyclodextrin-Based Nanoparticles (CDNPs) and Paroxetine Hydrochloride,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae042.

22. T. Kaneda, K. Kato, S. Ohtani, and T. Ogoshi, “Incorporation of ESIPT into Pillar[5]arene for Solid-State Dual Emission Responsive to n-Hexane Vapor,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae093.

23. S. Datta, H. Itabashi, T. Saito, and S. Yagai, “Secondary Nucleation as a Strategy towards Hierarchically Organized Mesoscale Topologies in Supramolecular Polymerization,” *Nature Chemistry* 17 (2025): 477.

24. M. Yoshida, K. Yasuda, T. Matsumoto, et al., “Vortex-Flow-Directed Chiral Macroscopic Ordering of Platelet Nanostructures Formed via the Supramolecular Assembly of Platinum Complexes with Bis(phenylisoxazolyl)benzene,” *Journal of the American Chemical Society* 147 (2025): 30674.

25. F. H. Saboor, S. Shahsavari, M. Zandjou, and M. Asgari, “From Structure to Catalysis: Advances in Metal-Organic Frameworks-Based Shape-Selective Reactions,” *ChemNanoMat* 10 (2024): e202400049.

26. A. Egi, H. Tanaka, T. Nakamura, K. Arashiba, Y. Nishibayashi, and K. Yoshizawa, “Computational Screening of PCP-Type Pincer Ligands for Mo-Catalyzed Nitrogen Fixation,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae041.

27. J. Yang, J. Li, C. Xu, et al., “Recent Advancements in Metal-Organic Frameworks-Derived Metal Phosphides as Anode for Lithium-Ion Batteries,” *ChemNanoMat* 11 (2025): e202400504.

28. S. Miyazaki, N. Ogiwara, C. A. Nagasaka, K. Takiishi, M. Inada, and S. Uchida, “Pore Design of POM@MOF Hybrids for Enhanced Methylene Blue Capture,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae105.

29. B.-N. T. Nguyen, T. T. Y. Tan, K.-I. Otake, S. Kitagawa, and J. Y. C. Lim, “MOF Catalysts for Plastic Depolymerizatio,” *Angewandte Chemie International Edition* 64 (2025): e202504017.

30. T. Nakamura, Y. Kondo, N. Ohashi, et al., “Materials Chemistry for Metal Halide Perovskite Rhotovoltaics,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoad025.

31. Y. Hirotsu, R. Sekiguchi, Y. Takeoka, M. Rikukawa, and M. Yoshizawa-Fujita, “Phase Transition and Ionic Conductivity of Pyrrolidinium-Based Ionic Plastic Crystals with Magnesium Salts,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae101.

32. K. Ariga, S. Akakabe, R. Sekiguchi, et al., “Boosting the Ionic Conductivity of Pyrrolidinium-Based Ionic Plastic Crystals by LLZO Fillers,” *ACS Omega* 9 (2024): 22203.

33. Y. Cheng, T. Araki, N. Kamimura, et al., “Accelerated Development of Novel Biomass-Based Polyurethane Adhesives via Machine Learning,” *ACS Applied Materials & Interfaces* 17 (2025): 15959.

34. T. S. Kang, M. Morikawa, M. Singh, and N. Kimizuka, “Electric Field-Driven Long-Range Order and Enhanced Polarization Switching in High-Dipole Ionic Liquids,” *Journal of the American Chemical Society* 147 (2025): 8809.

35. J. Chen, B. Liu, H. Cai, S. Liu, Y. Yamauchi, and S. C. Jun, “Covalently Interlayer-Confined Organic–Inorganic Heterostructures for Aqueous Potassium Ion Supercapacitors,” *Small* 19 (2023): 2204275.

36. M. Komiyama, K. Yoshimoto, M. Sisido, and K. Ariga, “Chemistry Can Make Strict and Fuzzy Controls for Bio-Systems: DNA Nanoarchitectonics and Cell-Macromolecular Nanoarchitectonics,” *Bulletin of the Chemical Society of Japan* 90 (2017): 967.

37. B. Roy and T. Govindaraju, “Enzyme-Mimetic Catalyst Architectures: The Role of Second Coordination Sphere in Catalytic Activity,” *Bulletin of the Chemical Society of Japan* 97 (2024): bcsj.20230224.

38. T. Miura, T. R. Malla, L. Brewitz, et al., “Cyclic β_2,3-Amino Acids Improve the Serum Stability of Macrocyclic Peptide Inhibitors Targeting the SARS-CoV-2 Main Protease,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae018.

39. W. Yospanya, A. Matsumura, Y. Imasato, et al., “Design of Cyborg Proteins by Loop Region Replacement with Oligo(ethylene Glycol): Exploring Suitable Mutations for Cyborg Protein Construction Using Machine Learning,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae090.

40. Y. Hata, H. Miyazaki, S. Okamoto, T. Serizawa, and S. Nakamura, “Nanospiked Cellulose Gauze That Attracts Bacteria with Biomolecules for Reducing Bacterial Load in Burn Wounds,” *Nano Letters* 25 (2025): 1177.

41. Y. Luo and Y. Gao, “Potential Role of Hydrogels in Stem Cell Culture and Hepatocyte Differentiation,” *Nano Biomedicine and Engineering* 16 (2024): 188.

42. S. d. Kalyana Sundaram, M. M. Hossain, M. Rezki, K. Ariga, and S. Tsujimura, “Enzyme Cascade Electrode Reactions with Nanomaterials and Their Applicability towards Biosensor and Biofuel Cells,” *Biosensors* 13 (2023): 1018.

43. H. Sumida, Y. Kimura, and T. Endo, “Role of Conformational Entropy in Low Melting Point of Ionic Liquids: A Molecular Dynamics Simulation Study,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoad008.

44. A. Kuzume and K. Yamamoto, “Dendrimer-Induced Synthesis of Subnano Materials and Their Characterization: Establishing Atom Hybrid Science,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae022.

45. K. Saitow, “Bright Silicon Quantum Dot Synthesis and LED Design: Insights into Size–Ligand–Property Relationships From Slow- and Fast-Band Engineering,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoad002.

46. Y. Tamai, M. Akiyama, L. Vallan, et al., “Energy Migration, Charge Transfer, and Charge Dissociation in Self-Assembling Nonfullerene Acceptor Aggregates with Zincporphyrin-Nonfullerene Acceptor Dyads,” *Chemical Science* 16 (2025): 15378.

47. Y. Itoh, T. Fu, P.-L. Champagne, et al., “Electric Double-Layer Synthesis of a Spongelike, Lightweight Reticular Membrane,” *Science* 389 (2025): 73.

48. W. He, Y. Yu, K. Iizuka, H. Takezawa, and M. Fujita, “Supramolecular Coordination Cages as Crystalline Sponges through a Symmetry Mismatch Strategy,” *Nature Chemistry* 17 (2015): 653.

49. Y. Sugimoto, P. Pou, M. Abe, et al., “Chemical Identification of Individual Surface Atoms by Atomic Force Microscopy,” *Nature* 446 (2007): 64.

50. Z. Diao, L. Hou, and M. Abe, “Probe Conditioning via Convolution Neural Network for Scanning Probe Microscopy Automation,” *Applied Physics Express* 16 (2023): 085002.

51. T. Nakamuro, “High-Speed Imaging and Quantitative Analysis of Nonequilibrium Stochastic Processes Using Atomic Resolution Electron Microscopy,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae082.

52. K. Sun, N. Cao, O. J. Silveira, et al., “On-Surface Synthesis of Heisenberg Spin-1/2 Antiferromagnetic Molecular Chains,” *Science Advances* 11 (2025): eads1641.

53. Y. Okawa and M. Aono, “Nanoscale Control of Chain Polymerization,” *Nature* 409 (2001): 683.

54. S. Kawai, O. Krejč, T. Nishiuchi, et al., “Three-Dimensional Graphene Nanoribbons as a Framework for Molecular Assembly and Local Probe Chemistry,” *Science Advances* 6 (2020): eaay8913.

55. T. Seki, “Surface-Mediated Dynamic Cooperative Motions in Azobenzene Polymer Films,” *Bulletin of the Chemical Society of Japan* 97 (2024): bcsj.20230219.

56. Y. Kim, K. Iimura, and N. Tamaoki, “Mechanoresponsive Diacetylenes and Polydiacetylenes: Novel Polymerization and Chromatic Functions,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae034.

57. K. Kimura, K. Miwa, H. Imada, et al., “Selective Triplet Exciton Formation in a Single Molecule,” *Nature* 570 (2019): 210.

58. N. Oyamada, H. Minamimoto, T. Fukushima, R. Zhou, and K. Murakoshi, “Beyond Single-Molecule Chemistry for Electrified Interfaces Using Molecule Polaritons,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae007.

59. P. Koviri, H. Komori, H. Tian, et al., “Single-Photon Level Ultrafast Time-Resolved Measurement Using Two-Color Dual-Comb-Based Asynchronous Linear Optical Sampling,” *Applied Physics Express* 17 (2024): 022001.

60. N. Amamizu, K. Sasaki, M. Nishida, et al., “Theoretical Study on Single-Molecule Electron Conductivity of Paddlewheel-Type Dichromium(II, II) Tetracarboxylate Complexes,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae130.

61. K. Ariga, M. Nishikawa, T. Mori, J. Takeya, L. K. Shrestha, and J. P. Hill, “Self-Assembly as a Key Player for Materials Nanoarchitectonics,” *Science and Technology of Advanced Materials* 20 (2019): 51.

62. S. Datta, Y. Kato, S. Higashiharaguchi, et al., “Self-Assembled Poly-Catenanes From Supramolecular Toroidal Building Blocks,” *Nature* 583 (2020): 400.

63. O. Oki, O. Krejčí, T. Nishiuchi, et al., “Synchronous Assembly of Chiral Skeletal Single-Crystalline Microvessels,” *Science* 377 (2022): 673.

64. N. Bäumer, S. Yamada, S. Ogi, and S. Yamaguchi, “Interfacing Flexible Design and Social Self-Sorting Enables Comprehensive Control over Photophysical and Self-Assembly Properties of Supramolecular Polymers,” *Journal of the American Chemical Society* 147 (2025): 8300.

65. S. Maekawa, L. Verstraete, H. S. Suh, et al., “High-Fidelity Directed Self-Assembly Using Higher-χ Polystyrene-Block-Poly(Methyl Methacrylate) Derivatives for Dislocation-Free Sub-10 Nm Features,” *Advanced Functional Materials* 35 (2025): 2421066.

66. M. Zharnikov, Y. Shoji, and T. Fukushima, “Tripodal Triptycenes as a Versatile Building Block for Highly Ordered Molecular Films and Self-Assembled Monolayers,” *Accounts of Chemical Research* 58 (2025): 312.

67. T. Hayashi and G. V. Latag, “Self-Assembled Monolayers as Platforms for Nanobiotechnology and Biointerface Research: Fabrication, Analysis, Mechanisms, and Design,” *ACS Applied Nano Materials* 8 (2025): 8570.

68. O. Vu, J. Song, H. Sugimoto, and M. Fujii, “Polymer-Shell Coating of Mie-Resonant Silicon Nanospheres for Controlled Fabrication of Self-Assembled Monolayer,” *ACS Applied Nano Materials* 8 (2025): 14802.

69. O. N. Oliveira Jr., L. Caseli, and K. Ariga, “The Past and the Future of Langmuir and Langmuir–Blodgett Films,” *Chemical Reviews* 122 (2022): 6459.

70. R. Terui, Y. Otsuki, Y. Shibasaki, and A. Fujimori, “Metal-Desorption and Selective Metal-Trapping Properties of an Organized Molecular Film of Azacalixarene-Containing Copolymer with Spherulite-Forming Ability,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae050.

71. J. Song, A. Jancik-Prochazkova, K. Kawakami, and K. Ariga, “Lateral Nanoarchitectonics From Nano to Life: Ongoing Challenges in Interfacial Chemical Science,” *Chemical Science* 15 (2024): 18715.

72. G. Rydzek, Q. Ji, M. Li, et al., “Electrochemical Nanoarchitectonics and Layer-by-Layer Assembly: From Basics to Future,” *Nano Today* 10 (2015): 138.

73. K. Ariga, J. Song, and K. Kawakami, “Layer-by-Layer Designer Nanoarchitectonics for Physical and Chemical Communications in Functional Materials,” *Chemical Communications* 60 (2024): 2152.

74. K. Ariga, “Layer-by-Layer Nanoarchitectonics: A Method for Everything in Layered Structures,” *Materials* 18 (2025): 654.

75. K. Adpakpang, P. Ponchai, L. Pukdeejorhor, K. Faungnawakij, and S. Bureekaew, “Confined Space of a Nickel-Triazole Metal–Organic Framework Responsible for High Product Selectivity and Enantiospecific Yield of Lactic Acid Converted From Sugar in a Water-Based System,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae016.

76. K. Sugamata, S. Kobayashi, A. Shirai, N. Amanokura, and M. Minoura, “Synthesis, Structural Analysis, and Gas-Adsorption Properties of a Dibenzothiophene-Based Hydroxamate/Zinc Metal-Organic Framework,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae017.

77. T. Kitao, “Precise Synthesis and Assembly of π-Conjugated Polymers Enabled by Metal–Organic Frameworks,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae103.

78. K. Ariga, M. Rezki, K. Suzuki-Nagata, T. Mikawa, and S. Tsujimura, “MOF-Based Hybrid Electrodes for Multi-Enzyme Cascade Reactions with Stabilized Mediator Immobilization,” *Chemical Communications* 61 (2025): 12309.

79. Y. Yin, X. Zhang, H. N. Nam, et al., “Enhanced Efficiency and Stability of Tin Halide Perovskite Solar Cells Through MOF Integration,” *Small* 21 (2025): 2411346.

80. D. Jiang, X. Xu, Y. Bando, et al., “Sulfonate-Functionalized Covalent Organic Frameworks for Capacitive Deionization,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae074.

81. T. Irie, S. Das, Q. Fang, and Y. Negishi, “The Importance and Discovery of Highly Connected Covalent Organic Framework Net Topologies,” *Journal of the American Chemical Society* 147 (2025): 1367–1380.

82. P. Singh and P. N. Dave, “Covalent Organic Frameworks (COFs) as Advanced Carriers for Corrosion Prevention: Cutting-Edge Advances and Promising Future Opportunities,” *ChemNanoMat* 11 (2025): e202400523.

83. Z. Li, R. P. Paitandi, Y. Tsutsui, et al., “Rolling Two-Dimensional Covalent Organic Framework (COF) Sheets into One-Dimensional Electronic and Proton-Conductive Nanotubes,” *Proceedings of the National Academy of Sciences* 122 (2025): e2424314122.

84. V. Malgras, Q. Ji, Y. Kamachi, et al., “Templated Synthesis for Nanoarchitectured Porous Materials,” *Bulletin of the Chemical Society of Japan* 88 (2015): 1171.

85. A. Alowasheeir, M. Eguchi, Y. Fujita, et al., “Extraordinary ^{99}Mo Adsorption: Utilizing Spray-Dried Mesoporous Alumina for Clinical-Grade Generator Development,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae099.

86. M. Han, T. Nagaura, J. Kim, et al., “Mesoporous Materials 2.0: Innovations in Metals and Chalcogenides for Future Applications,” *Bulletin of the Chemical Society of Japan* 98 (2025): uoae136.

87. K. Ariga, “Nanoarchitectonics: What’s Coming Next after Nanotechnology?,” *Nanoscale Horizons* 6 (2021): 364.

88. K. Ariga, J. Li, J. Fei, Q. Ji, and J. P. Hill, “Nanoarchitectonics for Dynamic Functional Materials From Atomic-/Molecular-Level Manipulation to Macroscopic Action,” *Advanced Materials* 28 (2016): 1251.

89. K. Ariga, X. Jia, J. Song, et al., “Nanoarchitectonics Beyond Self-Assembly: Challenges to Create Bio-Like Hierarchic Organization,” *Angewandte Chemie International Edition* 59 (2020): 15424.

90. R. P. Feynman, “There’s Plenty of Room at the Bottom,” *California Institute of Technology Journal of Engineering Science* 4 (1960): 23.

91. M. Roukes, “Plenty of Room, Indeed,” *Scientific American* 285 (2001): 48.

92. K. Ariga, Q. Ji, W. Nakanishi, J. P. Hill, and M. Aono, “Nanoarchitectonics: A New Materials Horizon for Nanotechnology,” *Materials Horizons* 2 (2015): 406.

93. K. Ariga, J. Song, and K. Kawakami, “From Inception to Innovation: 20 Years of Nanoarchitectonics,” *Chemistry – An Asian Journal* 20 (2025): e00836.

94. M. Matsumoto, L. Sutrisno, and K. Ariga, “Covalent Nanoarchitectonics: Polymer Synthesis with Designer Structures and Sequences,” *Journal of Polymer Science* 61 (2023): 861.

95. Y. Yao, S. Wang, C. Wang, et al., “Prussian Blue Analogue-Derived High-Entropy Alloy Nanoarchitectonics for Efficient Fenton-Like Catalysis,” *Journal of Materials Chemistry A* 12 (2024): 10081.

96. R. B. Laughlin and D. Pines, “The Theory of Everything,” *Proceedings of the National Academy of Sciences* 97 (2000): 28.

97. K. Ariga and R. Fakhrullin, “Materials Nanoarchitectonics From Atom to Living Cell: A Method for Everything,” *Bulletin of the Chemical Society of Japan* 95 (2022): 774.

98. K. Ariga, “Nanoarchitectonics: The Method for Everything in Materials Science,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoad001.

99. J. Song, K. Kawakami, and K. Ariga, “Localized Assembly in Biological Activity: Origin of Life and Future of Nanoarchitectonics,” *Advances in Colloid and Interface Science* 339 (2025): 103420.

100. T. Nakabayashi, S. Tahara, and S. Kajimoto, “Liquid–Liquid Phase Separation From the Viewpoint of Molecular Crowding Environment: A Raman Imaging Study,” *Journal of Physical Chemistry B* 129 (2025): 8087.

101. G. Li, C. Yuan, and X. Yan, “Peptide-Mediated Liquid–Liquid Phase Separation and Biomolecular Condensates,” *Soft Matter* 21 (2025): 1781.

102. K. Ariga and T. Kunitake, “Molecular Recognition at Air−Water and Related Interfaces: Complementary Hydrogen Bonding and Multisite Interaction,” *Accounts of Chemical Research* 31 (1998): 371.

103. K. Ariga, H. Ito, J. P. Hill, and H. Tsukube, “Molecular Recognition: From Solution Science to Nano/Materials Technology,” *Chemical Society Reviews* 41 (2012): 5800.

104. K. Ariga, “Molecular Recognition at the Air–Water Interface: Nanoarchitectonic Design and Physicochemical Understanding,” *Physical Chemistry Chemical Physics* 22 (2020): 24856.

105. K. Kurihara, K. Ohto, Y. Tanaka, Y. Aoyama, and T. Kunitake, “Molecular Rrecognition of Sugars by Monolayers of Resorcinol-Dodecanal Cyclotetramer,” *Journal of the American Chemical Society* 113 (1991): 444.

106. K. Kurihara, K. Ohto, Y. Honda, and T. Kunitake, “Efficient, Complementary Binding of Nucleic Acid Bases to Diaminotriazine-Functionalized Monolayers on Water,” *Journal of the American Chemical Society* 113 (1991): 5077.

107. Y. Ikeura, K. Kurihara, and T. Kunitake, “Molecular Recognition at the Air-Water Interface. Specific Binding of Nitrogen Aromatics and Amino Acids by Monolayers of Long-Chain Derivatives of Kemp’s Acid,” *Journal of the American Chemical Society* 113 (1991): 7342.

108. X. Cha, K. Ariga, M. Onda, and T. Kunitake, “Molecular Recognition of Aqueous Dipeptides by Noncovalently Aligned Oligoglycine Units at the Air/Water Interface,” *Journal of the American Chemical Society* 117 (1995): 11833.

109. X. Cha, K. Ariga, and T. Kunitake, “Molecular Recognition of Aqueous Dipeptides at Multiple Hydrogen-Bonding Sites of Mixed Peptide Monolayers,” *Journal of the American Chemical Society* 118 (1996): 9545.

110. D. Y. Sasaki, K. Kurihara, and T. Kunitake, “Specific, Multiple-Point Binding of ATP and AMP to a Guanidinium-Functionalized Monolayer,” *Journal of the American Chemical Society* 113 (1991): 9685.

111. M. Onda, K. Yoshihara, H. Koyano, K. Ariga, and T. Kunitake, “Molecular Recognition of Nucleotides by the Guanidinium Unit at the Surface of Aqueous Micelles and Bilayers. A Comparison of Microscopic and Macroscopic Interfaces,” *Journal of the American Chemical Society* 118 (1996): 8524.

112. M. Sakurai, H. Tamagawa, Y. Inoue, K. Ariga, and T. Kunitake, “Theoretical Study of Intermolecular Interaction at the Lipid−Water Interface. 1. Quantum Chemical Analysis Using a Reaction Field Theory,” *Journal of Physical Chemistry B* 101 (1997): 4810.

113. M. Ishii, Y. Nakai, S. Kaneko, et al., “Mechanoelectrical Transduction through Anion Recognition with Naphthalenediimide Monolayers at the Air–Water Interface,” *Langmuir* 40 (2024): 27040.

114. M. Ishii, Y. Nakai, Y. Yamashita, et al., “Pseudorotaxane Monolayers of Pillar[5]arene and Linear Fatty Acids at the Air–Water Interface,” *Chemical Communications* 61 (2025): 10138.

115. S. Negi, M. Hamori, Y. Kubo, H. Kitagishi, and K. Kano, “Monolayer Formation and Chiral Recognition of Binaphthyl Amphiphiles at the Air–Water Interface,” *Bulletin of the Chemical Society of Japan* 96 (2023): 48.

116. S. Sam, S. Sung, and D. Kim, “Sensitive Detection of Biomolecular Adsorption by a Low-Density Surfactant Layer Using Sum-Frequency Vibrational Spectroscopy,” *Langmuir* 39 (2023): 17412.

117. V. Gunwant and R. Pandey, “The Molecular Mechanism for the Interactions of Melittin Peptide with Hofmeister Anions at Air–Water Interfaces,” *Journal of Physical Chemistry B* 129 (2025): 7896.

118. J. F. Neal, W. Zhao, A. J. Grooms, A. H. Flood, and H. C. Allen, “Arginine–Phosphate Recognition Enhanced in Phospholipid Monolayers at Aqueous Interfaces,” *Journal of Physical Chemistry C* 122 (2018): 26362.

119. J. F. Neal, W. Zhao, A. J. Grooms, et al., “Interfacial Supramolecular Structures of Amphiphilic Receptors Drive Aqueous Phosphate Recognition,” *Journal of the American Chemical Society* 141 (2019): 7876.

120. A. J. Grooms, J. F. Neal, K. C. Ng, W. Zhao, A. H. Flood, and Heather C. Allen, “Thermodynamic Signatures of the Origin of Anti-Hofmeister Selectivity for Phosphate at Aqueous Interfaces,” *Journal of Physical Chemistry A* 124 (2020): 5621.

121. M. Okuno, S. Yamada, T. Ohto, et al., “Hydrogen Bonds and Molecular Orientations of Supramolecular Structure Between Barbituric Acid and Melamine Derivative at the Air/Water Interface Revealed by Heterodyne-Detected Vibrational Sum Frequency Generation Spectroscopy,” *Journal of Physical Chemistry Letters* 11 (2020): 2422.

122. T. Tahara, “Working on a Dream: Bringing up the Level of Interface Spectroscopy to the Bulk Level,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae012.

123. P. P. Samal, A. Maiti, S. Patel, et al., “Quantifying Hydrogen-Bonding Interactions in the Self-Assembly of Photoresponsive Azobenzene Amphiphiles at the Air–Water Interface,” *Journal of Physical Chemistry Letters* 15 (2024): 9193.

124. M. de la Puente and D. Laage, “How the Acidity of Water Droplets and Films Is Controlled by the Air–Water Interface,” *Journal of the American Chemical Society* 145 (2023): 25186.

125. B. E. Rugeley, K. R. Holt, E. B. Peterson, S. Moulai-Khatir, C. F. N. Koltun, and R. J. Rapf, "Changes in Protonation State of Atmospherically Relevant α-Hydroxyacids at the Air–Water Interface Measured by Surface Tension and IR-RAS," *Journal of Physical Chemistry A* 129 (2025): 7170.

126. W. Sung, K. Inoue, S. Nihonyanagi, and T. Tahara, "Unified Picture of Vibrational Relaxation of OH Stretch at the Air/Water Interface," *Nature Communications* 15 (2024): 1258.

127. Z.-C. Huang-Fu, N. V. Tkachenko, Y. Qian, et al., "Conical Intersections at Interfaces Revealed by Phase-Cycling Interface-Specific Two-Dimensional Electronic Spectroscopy (i2D-ES)," *Journal of the American Chemical Society* 146 (2024): 20996.

128. S. W. Devlin, S. Jamnuch, Q. Xu, et al., "Agglomeration Drives the Reversed Fractionation of Aqueous Carbonate and Bicarbonate at the Air–Water Interface," *Journal of the American Chemical Society* 145 (2023): 22384.

129. P. Basuri, S. Mukhopadhyay, K. S. S. V. Prasad Reddy, et al., "Spontaneous α-C−H Carboxylation of Ketones by Gaseous CO_2 at the Air-Water Interface of Aqueous Microdroplets," *Angewandte Chemie International Edition* 63 (2024): e202403229.

130. Y.-G. Fang, L. Wei, J. S. Francisco, C. Zhu, and W.-H. Fang, "Mechanistic Insights into Chloric Acid Production by Hydrolysis of Chlorine Trioxide at an Air–Water Interface," *Journal of the American Chemical Society* 146 (2024): 21052.

131. M. Li, C. Boothby, R. E. Continetti, and V. H. Grassian, "Size-Dependent Sigmoidal Reaction Kinetics for Pyruvic Acid Condensation at the Air–Water Interface in Aqueous Microdroplets," *Journal of the American Chemical Society* 145 (2023): 22317.

132. Y.-G. Fang, C. Yuan, S. Wang, C. Zhu, and W.-H. Fang, "Mechanistic Insights into the Reactive Uptake of Bromine Nitrate at the Air–Water Interface: Interplay Between Halogen Bonding and Solvation," *Journal of Physical Chemistry Letters* 16 (2025): 2957.

133. X. Zheng, Y.-G. Fang, M. Tan, et al., "Accelerated Methane Photo-Oxidation at the Air–Water Interface," *Journal of the American Chemical Society* 147 (2025): 26635.

134. A. Nandy, A. Rana, N. Shibata, and S. Banerjee, "Breaking the Strongest Organic Bonds by Water: Defluorosubstitutions at the Air–Water Interface of Microdroplets," *Journal of the American Chemical Society* 147 (2025): 22542.

135. A. Prasoon, S. Ghouse, N. N. Nguyen, et al., "Mimicking on-Water Surface Synthesis through Micellar Interfaces," *Nature Communications* 15 (2024): 10495.

136. K. Ariga, T. Mori, and J. P. Hill, "Mechanical Control of Nanomaterials and Nanosystems," *Advanced Materials* 24 (2012): 158.

137. K. Ariga, T. Mori, S. Ishihara, K. Kawakami, and J. P. Hill, "Bridging the Difference to the Billionth-of-a-Meter Length Scale: How to Operate Nanoscopic Machines and Nanomaterials by Using Macroscopic Actions," *Chemistry of Materials* 26 (2014): 519.

138. K. Ariga, "The Evolution of Molecular Machines through Interfacial Nanoarchitectonics: From Toys to Tools," *Chemical Science* 11 (2020): 10594.

139. K. Ariga, J. Song, and K. Kawakami, "Molecular Machines Working at Interfaces: Physics, Chemistry, Evolution and Nanoarchitectonics," *Physical Chemistry Chemical Physics* 26 (2024): 13532.

140. T. Michinobu, S. Shinoda, T. Nakanishi, et al., "Mechanical Control of Enantioselectivity of Amino Acid Recognition by Cholesterol-Armed Cyclen Monolayer at the Air-Water Interface," *Journal of the American Chemical Society* 128 (2006): 14478.

141. T. Mori, K. Okamoto, H. Endo, et al., "Mechanical Tuning of Molecular Recognition To Discriminate the Single-Methyl-Group Difference Between Thymine and Uracil," *Journal of the American Chemical Society* 132 (2010): 12868.

142. K. Ariga, "Nanoarchitectonics for Analytical Science at Interfaces and with Supramolecular Nanostructures," *Analytical Sciences* 37 (2021): 1331.

143. K. Ariga, "Molecular Tuning Nanoarchitectonics for Molecular Recognition and Molecular Manipulation," *ChemNanoMat* 6 (2020): 870.

144. K. Ariga, "Mechano-Nanoarchitectonics: Design and Function," *Small Methods* 6 (2022): 2101577.

145. Y. Zhou, J. J. Crassous, and M. Karg, "Core–Shell Microgels at Air/Water Interfaces: Role of Interfacial Tension in Monolayer Evolution," *Langmuir* 41 (2025): 9274.

146. Y. Cheng, S. Zhu, H. Ma, et al., "Multimodal Locomotion and Dynamic Interaction of Hydrogel Microdisks at the Air–Water Interface under Magnetic and Light Stimuli," *ACS Applied Materials & Interfaces* 16 (2024): 61633.

147. Q. Chen, J. Huang, X. Feng, H. Xie, and S. Zhou, "Controlling Self-Oscillation of a Single-Layer Liquid Crystal Elastomer at the Air–Water Interface via Light Programming for Water Strider-Inspired Aquatic Robots," *ACS Applied Materials & Interfaces* 17 (2025): 17433–17444.

148. G. Gardi, S. Ceron, W. Wang, K. Petersen, and M. Sitti, "Microrobot Collectives with Reconfigurable Morphologies, Behaviors, and Functions," *Nature Communications* 13 (2022): 2239.

149. K. Ariga, "Liquid–Liquid Interfacial Nanoarchitectonics," *Small* 20 (2024): 2305636.

150. X. Jia, J. Song, W. Lv, J. P. Hill, J. Nakanishi, and K. Ariga, "Adaptive Liquid Interfaces Induce Neuronal Differentiation of Mesenchymal Stem Cells through Lipid Raft Assembly," *Nature Communications* 13 (2022): 3110.

151. A. Chrysanthou, H. Kanso, W. Zhong, L. Shang, and J. E. Gautrot, "Supercharged Protein Nanosheets for Cell Expansion on Bioemulsions," *ACS Applied Materials & Interfaces* 15 (2023): 2760.

152. I. Tanabe and S. Ishikawa, "Comprehensive Characterization of Human Alveolar Epithelial Cells Cultured for 28 Days at the Air-Liquid Interface," *Scientific Reports* 15 (2025): 22995.

153. J. Nakanishi, T. Ueki, S. Dieb, H. Noguchi, S. Yamamoto, and K. Sodeyama, "Data-Driven Optimization of the in Silico Design of Ionic Liquids as Interfacial Cell Culture Fluids," *Science and Technology of Advanced Materials* 25 (2024): 2418287.

154. J. Tian, J. Yang, T. Chen, et al., "Generation of Human Endometrial Assembloids with a Luminal Epithelium Using Air–Liquid Interface Culture Methods," *Advancement of Science* 10 (2023): 2301868.

155. E. Tanaka, T. Kishikawa, Y. Miyakawa, et al., "Modeling Intestinal Epithelial Function and Polarity Using Human iPSCs under Air–Liquid Interface Culture," *Biochemical and Biophysical Research Communications* 781 (2025): 152547.

156. A. Sabapaty, P.-Y. Lin, and J. C. Y. Dunn, "Effect of Air–Liquid Interface on Cultured Human Intestinal Epithelial Cells," *FASEB Bioadvances* 6 (2024): 41.

157. C. M. Tse, Z. Zhang, R. Lin, R. Sarker, M. Donowitz, and V. Singh, "The Air–Liquid Interface Reorganizes Membrane Lipids and Enhances the Recruitment of Slc26a3 to Lipid-Rich Domains in Human Colonoid Monolayers," *International Journal of Molecular Sciences* 24 (2023): 8273.

158. Y. Huang, H. Chen, X. Qiao, et al., "Liquid-Liquid Phase Separation-Boosted Transmembrane Delivery in Interactive Protocell Communities," *Nature Communications* 16 (2025): 5231.

159. P. Bairi, K. Minami, J. P. Hill, et al., "Supramolecular Differentiation for Construction of Anisotropic Fullerene Nanostructures by Time-Programmed Control of Interfacial Growth," *ACS Nano* 10 (2016): 8796.

160. G. Chen, F. Sciortino, K. Takeyasu, et al., "Hollow Spherical Fullerene Obtained by Kinetically Controlled Liquid-Liquid Interfacial Precipitation," *Chemistry – An Asian Journal* 17 (2022): e202200756.

161. G. Chen, M. Isegawa, T. Koide, et al., “Pentagon-Rich Caged Carbon Catalyst for the Oxygen Reduction Reaction in Acidic Electrolytes,” *Angewandte Chemie International Edition* 63 (2024): e202410747.

162. G. Chen, T. Koide, J. Nakamura, and K. Ariga, “Nanoarchitectonics for Pentagon Defects in Carbon: Properties and Catalytic Role in Oxygen Reduction Reaction,” *Small Methods* 9 (2025): 2500069.

163. M. Ishii, Y. Yamashita, S. Watanabe, K. Ariga, and J. Takeya, “Doping of Molecular Semiconductors through Proton-Coupled Electron Transfer,” *Nature* 622 (2023): 285.

164. W. Chaikittisilp, Y. Yamauchi, and K. Ariga, “Material Evolution with Nanotechnology, Nanoarchitectonics, and Materials Informatics: What Will Be the Next Paradigm Shift in Nanoporous Materials?,” *Advanced Materials* 34 (2022): 2107212.

165. L. R. Oviedo, V. R. Oviedo, M. O. Martins, S. B. Fagan, and W. L. da Silva, “Nanoarchitectonics: The Role of Artificial Intelligence in the Design and Application of Nanoarchitectures,” *Journal of Nanoparticle Research* 24 (2022): 157.

166. T. Sadakane, K. Nakata, K. Suda, and D. Yokogawa, “Feature Attributions for Water–Solubility Predictions Obtained via Artificial Intelligence Methods and Chemists,” *Bulletin of the Chemical Society of Japan* 97 (2024): uoae121.

167. B. V. Parakhonskiy, J. Song, and A. G. Skirtach, “Machine Learning in Nanoarchitectonics,” *Advances in Colloid and Interface Science* 343 (2025): 103546.

168. H. Bae, H. Ji, K. Konstantinov, et al., “Artificial Intelligence-Driven Nanoarchitectonics for Smart Targeted Drug Delivery,” *Advanced Materials*. (2025): e10239.

Biographies

Katsuhiko Ariga received his Ph.D. degree from the Tokyo Institute of Technology in 1990. He joined the National Institute for Materials Science (NIMS) in 2004 and is currently a group leader of the Supermolecules Group and a Senior Scientist with Special Missions of Research Center for Materials Nanoarchitectonics for Materials Nanoarchitectonics (MANA). He is also appointed as a professor at The University of Tokyo. His expertise is in supramolecular chemistry and material nanoarchitectonics.