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Shaping interfaces with light: evolution and application of azobenzene-containing photoresponsive surfactants

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Surfactants are ubiquitous, playing critical roles in biological processes, including those occurring in the lungs, as well as in many consumer and industrial applications. This versatility is further enhanced by stimuli-responsive surfactants, among which photoresponsive systems have garnered considerable research interest, especially in recent years. A thorough analysis of this subject is required because there are not many review papers on photoresponsive surfactants, which has limited our knowledge of this area. This review describes the historical emergence and evolution of photoresponsive surfactants. It provides a detailed analysis of the various photoresponsive moieties (*e.g.*, azobenzene and spiropyran) that are covalently integrated into surfactant architectures, focusing on their distinct types, operational limitations, and applications. A comparative analysis of conventional *versus* gemini photoresponsive surfactants is presented, highlighting key differences in their physicochemical properties. This review elucidates the mechanisms of photoisomerization in azobenzene and its direct impact on molecular-level properties (*e.g.*, polarity and geometry) and their macroscopic physical outcomes (*e.g.*, surface tension and viscosity) in both azo-based and non-azo surfactants. This review comprehensively surveys the diverse applications of these materials, including enhanced oil recovery, environmental remediation, motion manipulation, catalysis, smart materials, and controlled drug delivery.

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1. Introduction

Surfactants or surface-active agents are a class of chemical compounds that act at interfaces, primarily to lower the surface

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Mr Shivank Sharma holds an MSc in Organic Chemistry from Vellore Institute of Technology (VIT), Vellore, India. His research interests during that time included the design, synthesis and characterisation of novel surfactants and their applications. He then completed a GRIP fellowship at Jawaharlal Nehru Centre for Advanced Scientific Research (JNCASR), Bengaluru, India. His research focuses on the design and synthesis of peptide nucleic acids (PNAs) and peptidomimetics to engineer targeted molecular architectures for overcoming antimicrobial resistance.

tension of liquids, which make it easier for them to spread or mix with other substances and facilitates the mixing of otherwise immiscible substances. These properties give rise to various applications, such as their use as stabilizing agents for emulsions[1] and in the relatively newer area of stabilizing microemulsions for nanomaterial synthesis.[2] Surfactants have numerous applications across a wide range of fields, which are discussed in the Applications section. A surfactant molecule has both hydrophilic (water-attracting) and hydrophobic (water-repelling) regions, allowing it to interact with different phases simultaneously, giving rise to stabilization properties. The head of the molecule is the hydrophilic part, and the tail, which is usually a carbon chain, is the hydrophobic part; these two chemically distinct parts act as ''connecting points'', and the rest of the molecule serves as a ''bridge'' between two different phases.[3]

There are different types of surfactants based on the number of head and tail groups. Surfactants with only one head group and one long tail per molecule are called monomeric or conventional surfactants, whereas those with two head groups and two tails are called dimeric or gemini surfactants.[4] Surfactants can also be classified based on the charge of their head group. Thus, they can be categorized as non-ionic, cationic, anionic, and zwitterionic surfactants. Surfactants are present all around us in different capacities, including the human body,[5] the petroleum industry,[6] the food industry,[7] water purification,[8] pharmaceuticals,[9,10] controlled drug delivery systems,[11] soil water remediation,[12] nanoparticle synthesis,[2] and sensors for metal detection.[13]

At low concentrations, surfactants dissolved in water are either adsorbed at the interface or present in a monomolecular state. The lowest concentration at which micelles form in solution is known as the critical micelle concentration (CMC).[14] Switchable surfactants, or stimuli-responsive surfactants, are a special class of surfactants that alter their rheological properties based on one or more external stimuli. In addition to the benefits of conventional surfactants, switchable surfactants also solve the technical issue of surfactant separation from aqueous solution, resulting in significant cost savings. Recently, a wide range of stimuli-responsive surfactants have been discovered, which respond to a variety of stimuli, such as pH,[15,16] redox conditions,[17,18] CO_2 exposure,[19,20] magnetic fields,[21,22] light (photoresponsive),[11] and temperature.[23] The changes or transitions in interfacial properties due to surface-active molecules can be attributed to the deviation of the interface from an equilibrium state to a non-equilibrium state based on the external stimuli provided. This has broad implications in terms of the type of stimulus. As observed in the case of pH- or temperature-responsive surfactants, the timescale of the equilibrium deviation is comparable to that of surfactant diffusion, complicating their practical use and applications.[24]

One stimulus that can reach most of the bulk at an apparently instantaneous rate depending on the bulk characteristics is light. This gives rise to photochromism as a very attractive phenomenon to exploit. Photochromism is the phenomenon in which a molecule reversibly changes between two different forms with different absorption spectra upon exposure to light. In addition to absorption spectra, many other physicochemical properties such as isomeric structure, polarity, magnetic strength, refractive index, quantum yield, and redox potentials can also vary.[25–27] Exploring

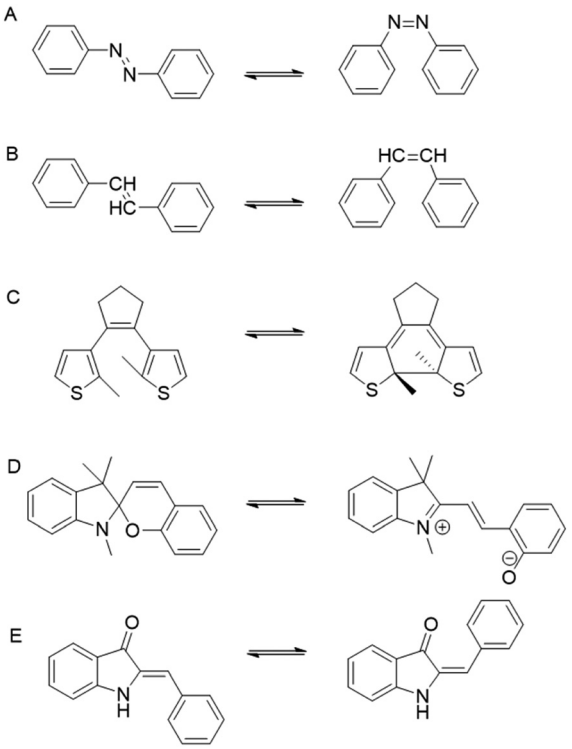

Fig. 1 Photoisomerization in (A) azobenzene, (B) stilbene, (C) diarylethene, (D) spiropyran, and (E) hemiindigo.

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photochromism in the context of surfactants has given rise to light-responsive surfactants, which are currently widely studied because of their rapid and tunable isomerization, use of light as a green stimulants, and cyclability.[28] Consequently, they have found applications in many fields such as liquid crystals,[29] molecular switches,[30] micellar catalysts,[31] photofoams,[32] and polymers.[33]

Many functional groups or structural modifications can be introduced in a molecule to induce photoresponsive behavior. Some of these groups include azobenzene, hemiindigo (and hemiindigoids), stilbene, diarylethene and spiropyran (Fig. 1). In these molecules, azobenzene and stilbene undergo photoisomerization upon irradiation, while diarylethene and spiropyran undergo structural isomerization *via* a ring-opening/closing transition.[24,34,35] In contrast, stilbene derivatives find relatively fewer applications because of their low solubility in aqueous media.

Recently, azo group-containing molecules have become very attractive to researchers, with some of their reported properties, including static properties such as equilibrium surface tension, critical micellar concentration (CMC), photoswitching between equilibrium assemblies, formation of complexes, and contact angles,[24] as well as dynamic properties. For instance, Diguet *et al.*[36] studied the manipulation of oil droplets at the water-air interface in the presence of a cationic azo surfactant, whereas Liang *et al.*[37] studied the dynamic manipulation of droplets on liquid-infused surfaces using a spiropyran-based photoresponsive surfactant. Kunitake *et al.*[38] are the earliest reported authors who, in 1981, discussed azobenzene-containing amphiphile structures and the effect of their structures on their aggregate morphologies. Shinkai *et al.*[31] demonstrated that the *trans–cis* isomerization of azobenzene head groups in surfactants affected their aggregation mode. They were the first to demonstrate light-based control of micellar catalysis. The azobenzene group can be present in any part of the surfactant to give it photoresponsive properties, with differing effects, including the head group, tail group, or the spacer region in geminal surfactants.[32] The number of azobenzene groups can also be varied and their effects studied gemini surfactants with an azobenzene group per tail have been synthesized and studied.[39]

This article aims to highlight the importance and evolution of responsive surfactants. Surfactants can be responsive to pH, temperature, CO_2/N_2, and light. Among the responsive surfactants, photoresponsive surfactants are discussed herein due to their versatility upon irradiation with UV-vis light. This includes the effects of molecular substitutions, functional groups, number of head groups, and the position of the azobenzene moiety on the rheological properties of the surfactants. We also summarize the uses of azobenzene-based gemini surfactants in conjugation with conventional surfactants in various applications. We also compile a comprehensive list of the applications of photoresponsive surfactants and study the effects of surfactant structure that make them suitable for a particular application, with emphasis on their future scope in various industries.

2. Classification of surfactants

Surfactants are classified as conventional and gemini surfactants based on the number of head and tail groups per molecule.

2.1 Conventional surfactants

The primary class of surfactants is conventional surfactants. These types of molecules have the unique characteristic of having a singular head and tail group to act as the hydrophilic and hydrophobic part, respectively. When studies on surfactants began, these were the first type of surfactants synthesized in laboratories owing to their simple design. Naturally, the first type of synthetic photoresponsive surfactants were also conventional surfactants.[31] A plethora of experiments were done to induce light-responsive character in molecules. Some methods included the introduction of a moiety that undergoes photoisomerization on the tail end,[26,32] near the head group,[34] or even somewhere in the middle.[24] In most of these compounds, azobenzene was the preferred moiety because of its reversible photoisomerization behavior.

Some efforts have been made to study the effect of the position of the azobenzene groups on the properties of the surfactants.[40] Peng *et al.*[40] synthesized two types of surfactants, one in which azobenzene was placed in the center of the hydrophobic tail and another in which azobenzene was placed at the interface between the hydrophilic head and hydrophobic tail. The first type formed multiple lyotropic liquid crystal (LLC) phases (such as hexagonal and lamellar) depending on the water content, and showed significant photoinduced changes in its self-assembled structure and bulk physical properties. In contrast, the second type was less responsive to light stimuli overall. They concluded that if the azobenzene is too close to the hydrophilic/hydrophobic boundary (interface) in a surfactant structure, steric constraints and packing requirements prevent the molecule from rearranging after it undergoes isomerization. Conventional surfactants can then further be classified based on the charge of their head group, including nonionic,[32,34] cationic,[24,26,34] and anionic[41] surfactants. Nonionic surfactants do not have any charge on their head groups, while cationic surfactants have positively charged head groups, and anionic surfactants have negatively charged head groups.

Many synthetic procedures have been devised to synthesize conventional photoresponsive surfactants. The simplest approach involves attaching a photoresponsive unit towards the end of the tail with a quaternary ammonium salt as the head group. This method is considered to be the easiest way to introduce photoresponsive behavior in conventional surfactants. Other methods, including introducing the photoresponsive moiety near the head group or even between the carbon chains, have also been devised, as discussed previously, and the corresponding synthetic procedures are well established. The resulting structures are presented in Table 2.

From a performance standpoint, conventional photoresponsive surfactants generally exhibit critical micelle concentration (CMC) values in the range of 10^{-3}–10^{-4} M, requiring relatively higher concentrations to achieve micellization. Correspondingly,

their equilibrium surface tension values are typically reduced to 35–45 mN m^{-1}, depending on molecular structure and experimental conditions.[39] However, although these systems are effective, their higher CMC can lead to increased material usage, which may have economic and environmental implications at larger scales.

Despite these limitations, conventional surfactants offer important advantages, including synthetic simplicity, cost-effectiveness, and well-established scalability, which continue to make them relevant for practical applications.

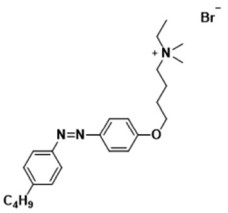
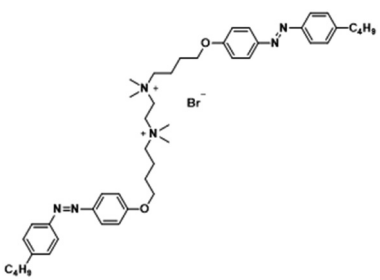
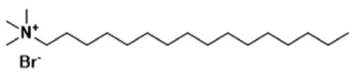
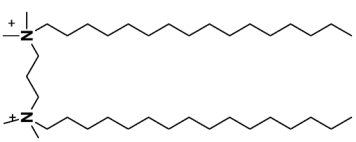
2.2 Gemini surfactants

Gemini surfactants represent an advanced class of surfactants characterized by the presence of two hydrophilic head groups and two hydrophobic tails, typically linked by a spacer unit. This unique architecture leads to markedly different interfacial and aggregation behavior compared to conventional surfactants.[4] As a result, gemini surfactants exhibit significantly lower CMC values ($\sim 10^{-5}$–10^{-6} M), corresponding to reductions of approximately 1–3 orders of magnitude compared to conventional surfactants. In addition, they often achieve greater reductions in surface tension, with typical values reaching $\sim$25–30 mN m^{-1}, indicating enhanced surface activity and adsorption efficiency.[39] These properties make gemini surfactants particularly attractive for applications requiring high efficiency at low concentrations, such as controlled drug delivery, nanostructure templating, and responsive interfacial systems. Furthermore, their lower operational concentrations can reduce the overall chemical load in a system, offering potential environmental benefits. However, it is important to note that gemini surfactants are not universally superior. Their synthesis is often more complex, and factors such as spacer length, flexibility, and molecular symmetry can significantly influence their behavior. Additionally, their higher production costs and challenges in large-scale synthesis may limit their widespread industrial adoption. Therefore, although gemini surfactants demonstrates enhanced interfacial efficiency in terms of lower CMC and improved surface activity, conventional and gemini surfactants offer complementary advantages, and the choice between them should be guided by the specific requirements of the intended application.[42] Table 1 compares the structures, CMC values and surface tensions of two representative conventional and gemini surfactants (Fig. 2).

3. Evolution of photoresponsive surfactants

The first report of a surfactant altering the morphology of its micellar structures was by Kunitake *et al.* in 1981.[38] Their work focused on the formation of stable bilayer assemblies in water from single-chain amphiphiles and the relationship between the amphiphile structure and the aggregate morphology. They incorporated a rigid moiety as the 'spacer' in the amphiphile structures. They discussed azobenzene in the context of introducing 'dynamic' control of the amphiphile aggregate morphology. In their work, it was observed that the *trans*-azobenzene

Table 1 The difference in the CMC and surface tension values of conventional and gemini surfactants[39,89]

S. no.	Structure	CMC (mM)		Surface tension (mN m^{-1})
		Conventional surfactant	Gemini surfactant	
		*Trans*-0.98 mM *Cis*-1.15 mM	—	*Trans*-33.06 mN m^{-1} *Cis*-37.39 mN m^{-1}
1.		—	*Trans*-0.52 mM *Cis*-0.53 mM	*Trans*-28.94 mN m^{-1} *Cis*-31.69 mN m^{-1}
		1 mM		30–40 mN m^{-1}
2.			0.02 mM	35–40 mN m^{-1}

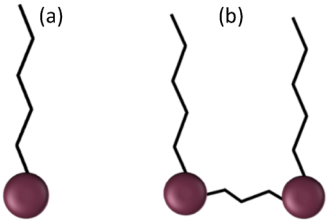


Fig. 2 Structural representation of (a) conventional and (b) gemini surfactants.

spacer unit adopted an extended geometry, and globular aggregates were formed. It was also seen that the *cis*-azobenzene unit possessed a bent geometry and should act as a bent rigid segment. However, the photochemical properties of the aggregates were not thoroughly studied.

Many groups tried manipulating the basic structure of surfactants by varying the number of head groups or tail groups, changing the length of the alkyl chain, and introducing different functionalities in the head group or the tail group including but not limited to ether linkages[13,40] and carbonyl groups in the form of both esters[43] and carboxylic acids.[44] Spacers with varying lengths have been studied by different groups in the case of gemini surfactants[45–48] and their effects are briefly discussed in the following sections. Various photoresponsive moieties have also been employed including azobenzene, stilbene, merocyanine, diarylethene, and spiropyran. In most applications, the azobenzene group has proven to be the most synthetically viable and experimentally versatile group due to its properties such as quick and tunable photoresponsive behavior, good cycling stability, and excellent quantum yield.[49,50]

Many photomechanisms have been considered for introducing photoresponsive behavior. Among them, *cis–trans* photoisomerization is the most widely employed, particularly in the case of azobenzene- and stilbene-based moieties. Spiropyran follows a ring-opening/closing mechanism. An interesting study was reported by Ahmadi *et al.*, who employed the photodimerization properties of coumarin to synthesize photoresponsive polymeric micelles.[51] This work represented an important step toward the synthesis of polymeric surfactants that are not only light-responsive but also pH- and temperature-responsive. These surfactants have evolved from initial investigations as a proof of concept to real-life applications. As demonstrated earlier, azobenzene-containing surfactants initially exhibited major solubility issues, which were later resolved through the introduction of charged heads. After decades of research, stimuli-responsive surfactants have emerged as a rapidly developing field both in science and industry.

4. Mechanism and kinetics of *cis–trans* isomerization of azobenzene

Based on the wavelength of the irradiated light, azobenzene undergoes isomerization, resulting in a change in its chemical structure and, consequently, its properties. The most common type is *cis–trans* isomerization, which is exhibited by moieties such as azobenzene and stilbene. The *cis–trans* isomerization of azobenzene has been thoroughly studied as early as 1954.[52]

In azobenzene-based surfactants, the *trans* isomer absorbs light at about λ = 360 nm and is converted to the *cis* form. Upon illumination, the *cis* isomer absorbs at around λ = 460 nm and relaxes back to the *trans* form. *Cis*-to-*trans* isomerization can also occur thermally. These values vary depending on the other functional groups present in the molecule. In most cases, full conversion to the *cis* or *trans* isomer is not possible.[53] Rather, these systems typically reach a photostationary state consisting of a mixture of *trans* and *cis* isomers, the composition of which depends on the chemical substituents on the chromophore and the wavelength of the incident light used to illuminate the system.[54] Mechanistic insights into light-induced interfacial processes have also been studied.[55] For the azobenzene molecule, two major mechanisms for the photoisomerization process have been proposed, namely the rotational and inversion mechanisms.[56] Additionally, cases where these two mechanisms act in concert have been theorized.[57] Some groups have referred to them as inversion-assisted rotation[58] and concerted inversion[59] (Fig. 3).

The rotational mechanism is based on the evolution of the N═N double bond toward increased single-bond character, allowing bond rotation and reducing the requirement for coplanarity. This causes one of the phenyl rings to move out of the molecular plane and the dihedral angle of C–N–N–C to change from 180° to 0°.[60] The N═N–C bond angle remains at 120°. The inversion process involves changes in the N═N–C bond angle from 120° toward 180°, approaching a linear configuration, accompanied by rotation of the phenyl ring through the N–C single bond.[61] During inversion, the C–N═N–C dihedral angle remains unchanged. In the inversion-assisted rotational pathway, the C–N═N–C dihedral angle decreases, while the N═N–C bond angle also decreases. In contrast, concerted inversion proceeds through an unusual pathway involving a linear transition state, in which both N═N–C bond angles approach 180°.[62]

Rotation, inversion, co-inversion, and inverse-assisted rotation vary in the structural motion around the N═N-azo linkage and in their associated energy barriers. Torsion around the N═N double bond induces isomerization in the rotation mechanism (A), which involves a 180° rotation that temporarily breaks π-conjugation and proceeds through a high-energy twisted transition state in which the C–N═N–C dihedral angle approaches 90°. Alternatively, the inversion process (B) does not involve bond rotation; instead, one of the nitrogen atoms rehybridizes from sp^2 toward a quasi-linear sp-like configuration, inducing a bending motion (change in C–N═N angle) while preserving the π-bond framework. A coordinated geometric rearrangement that avoids complete bond rotation but necessitates a greater cooperative distortion energy is produced by the co-inversion pathways (C), which entails simulations or sequential inversion at both nitrogen atoms. The inversion-assisted rotation mechanism (D) is a hybrid pathway that

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Table 2 Structure of azobenzene containing surfactants and their various applications

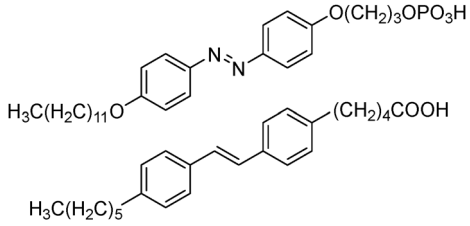
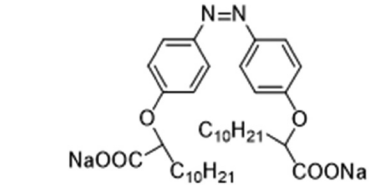
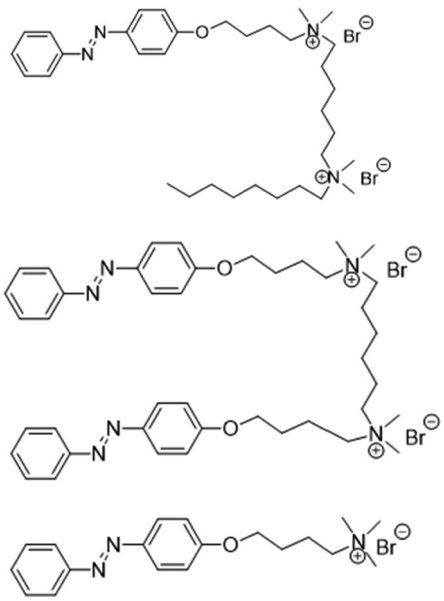
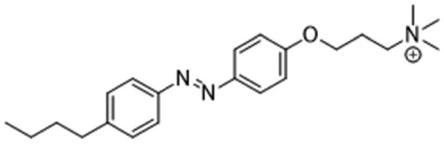
S. no.	Structure	Key points	Applications	Ref.
1.		Compares azobenzene and stilbene surfactants as dopants in DHP bilayer membranes. They produce opposite effects on potassium ion (K^+) permeability.	Model systems for photoregulated ion channels. Controlled release of ions or drugs. Creating nanodevices with opposing, light-controlled functions.	41
2.		*Trans* state promotes the formation of a high-viscosity (gel-like) fluid. *Cis* state converts system to a sol state.	Photorheological fluids (light-controlled viscosity). Switchable "smart" materials.	44
3.		The spacer length is crucial. The largest light-induced change in CMC was seen with moderate spacer length (n = 3, 4), not the shortest one (n = 2).	Light-controlled surface tension modification.	120
4.		Demonstrates a "pumping-out" mechanism where light irreversibly removes surfactant from the air–water interface.	Light-induced, irreversible destabilization of foams and emulsions.	24

Table 2 (*continued*)

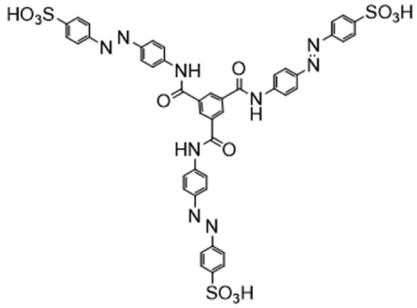
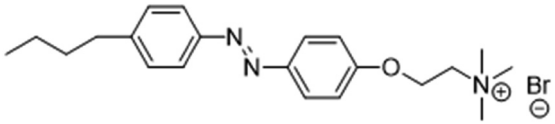
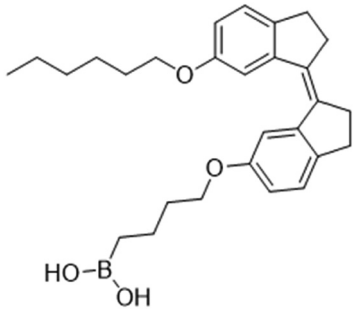
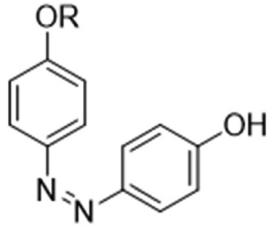
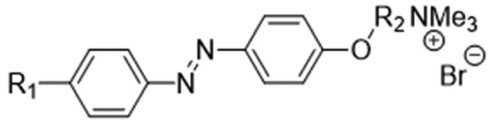
S. no.	Structure	Key points	Applications	Ref.
5.		The surfactant acts as both a stabilizer (capping agent) and a shape-directing agent during the synthesis of gold nanoparticles (AuNPs).	Photocontrol of nanoparticle morphology. Fabricating materials with tunable plasmonic properties.	2
6.		Demonstrates reversible "catch-and-release" of polycyclic aromatic hydrocarbons (PAHs).	Removing toxic PAH pollutants from water. Controllable release systems for hydrophobic "guest" molecules.	1
7.		The *trans* molecule forms heterodimers, which effectively transport protons through a lipid bilayer. The *cis* isomer disrupts this dimerization, significantly inhibiting proton transport.	Artificial, light-controlled ion channels. Photoregulation of pH inside vesicles or liposomes.	30
8.		The tail length is crucial. Longer-chain surfactants (C_{10}, C_{12}) showed the most significant and effective light-control over foam stability.	Light-controlled foaming and defoaming agents for industrial processes.	32
9.		Spacer length is more dominant than tail length in controlling the *trans* isomers self-assembly. Short spacer (n = 3) forms small, spherical micelles regardless of tail length. Long spacer (n = 11) forms large, ellipsoidal/wormlike micelles, especially with longer tails.	Smart materials with predictable, light-controlled nanostructures.	121

Table 2 (*continued*)

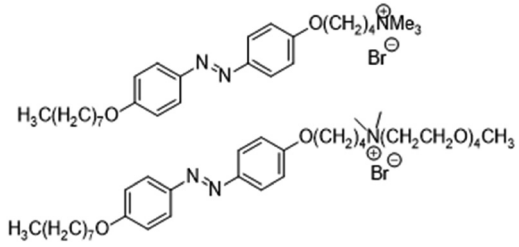
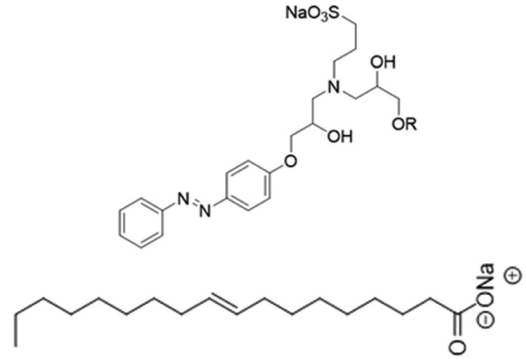
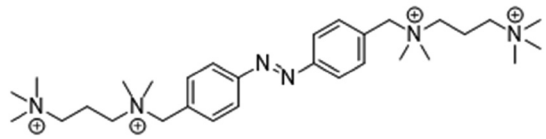
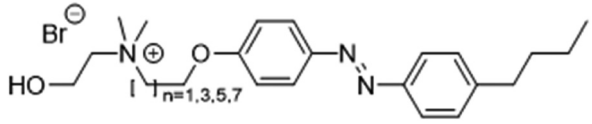
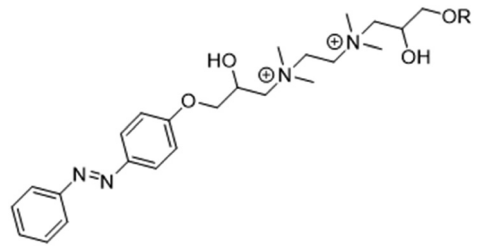
S. no.	Structure	Key points	Applications	Ref.
10.		Instead of affecting the main, strong π–π* band (~350 nm), Hg^{2+} binding specifically interacts with and enhances the very weak n–π* transition band (~436 nm).	Selective and colorimetric detection of Hg^{2+} ions in aqueous solutions.	13
11.		Incorporated an azobenzene group and a sulfonic acid headgroup. The series varies the hydrophobic tail length.	Photo-switchable foaming agents.	34
12.		Used a light-switchable "pseudo-tetramer" assembly as the builder for the wormlike micelles, offering a new route to photorheological fluids.	Switchable "smart" gels and materials.	122
13.		A Pickering emulsion, stabilized by solid nanoparticles ($Pd@SiO_2$), and light-responsive surfactants.	Phase-transfer catalysis, acting as a nanoreactor.	116
14.		The gemini structure results in exceptionally low CMCs compared to similar conventional photosurfactants.	High-efficiency, photo-switchable wetting agents.	26

Table 2 (*continued*)

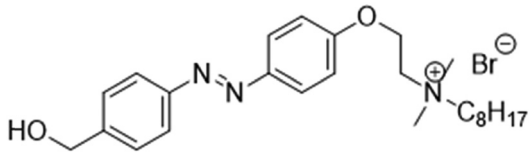
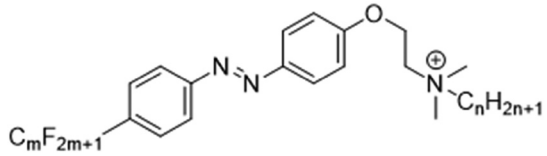
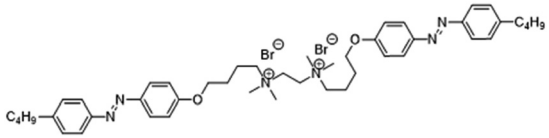
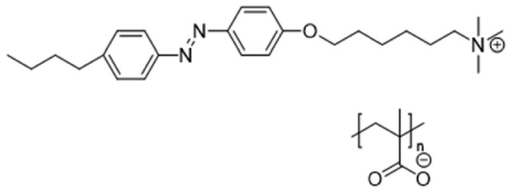
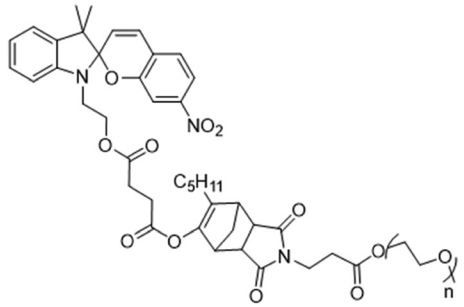
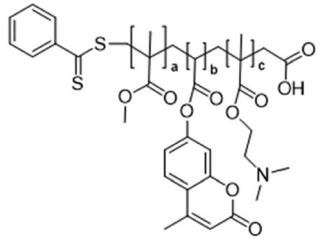
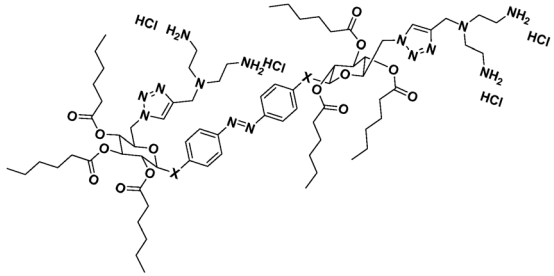
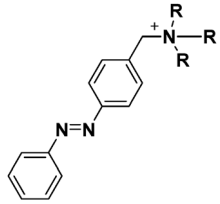
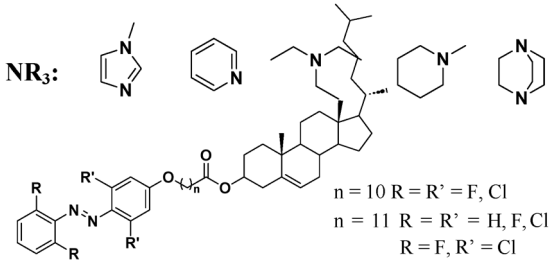
S. no.	Structure	Key points	Applications	Ref.
15.		Forms vesicles (bilayers), not just simple micelles in solution. These are further loaded with anticancer drug doxorubicin (DOX).	On-demand, site-specific drug release.	11
16.		The hybrid surfactant shows superior surface activity (lower CMC and lower surface tension) than either the pure hydro or pure fluoro chain versions due to the unique packing of the two different tails.	Light-controlled foaming and emulsification (especially for fluorinated systems).	91
17.		Demonstrates reversible, high-efficiency solubilization of PAHs.	Remediating PAH-contaminated groundwater.	4
18.		Provides multi-stage, stepwise photocontrol (*trans–trans* to *cis–trans* to *cis–cis*). This stepwise isomerization allows for a wider and more tunable range of properties.	Photo-switchable foaming agents with tunable foam stability.	39
19.		Uses a photoresponsive surfactant inside an aqueous droplet placed on a liquid-infused surface (LIS/SLIPS). Achieves precise, dynamic manipulation (movement) of the droplet using only light employing the Marangoni effect.	Microfluidics/lab-on-a-chip with light-based pumps. Micro-cargo transport using droplets as light-guided carriers.	37
20.		A triple-stimuli-responsive polymer, responding to temperature, pH, and light, that forms reversibly cross-linked micelles.	Smart nanocarriers for targeted cancer therapy.	51

Table 2 (*continued*)

S. no.	Structure	Key points	Applications	Ref.
21.		Each glucose scaffold is functionalized with specific components to facilitate gene delivery: ionizable headgroups that electrostatically interact with pDNA, and lipophilic tails that drive self-assembly. This "Janus" design having distinct polar and non-polar faces enables the molecule to form stable, single- component nanocomplexes with nucleic acid.	Light-responsive, single-component molecular vectors for the spatiotemporally controlled and organ-selective delivery of plasmid DNA (pDNA) in gene therapy	106
22.		Ionic groups are added to the structure to make the fuel water-soluble. Internal cation–π interactions help the fuel store more energy for over 30 days.	Water-soluble solar thermal fuels show great potential for fabricating advanced solar energy storage devices that capture and store solar energy to be later released as heat.	119
23.		The molecular design incorporates an alkyl spacer of varying lengths (n = 10, 11) connecting the two moieties, along with *ortho*-substitutions on the azobenzene ring to modulate the material properties and changing efficiency.	These liquid crystal-based solar-thermal fuels can be integrated as coatings for solar blankets, deicing systems, and functional fibers to provide on-demand heat release	117

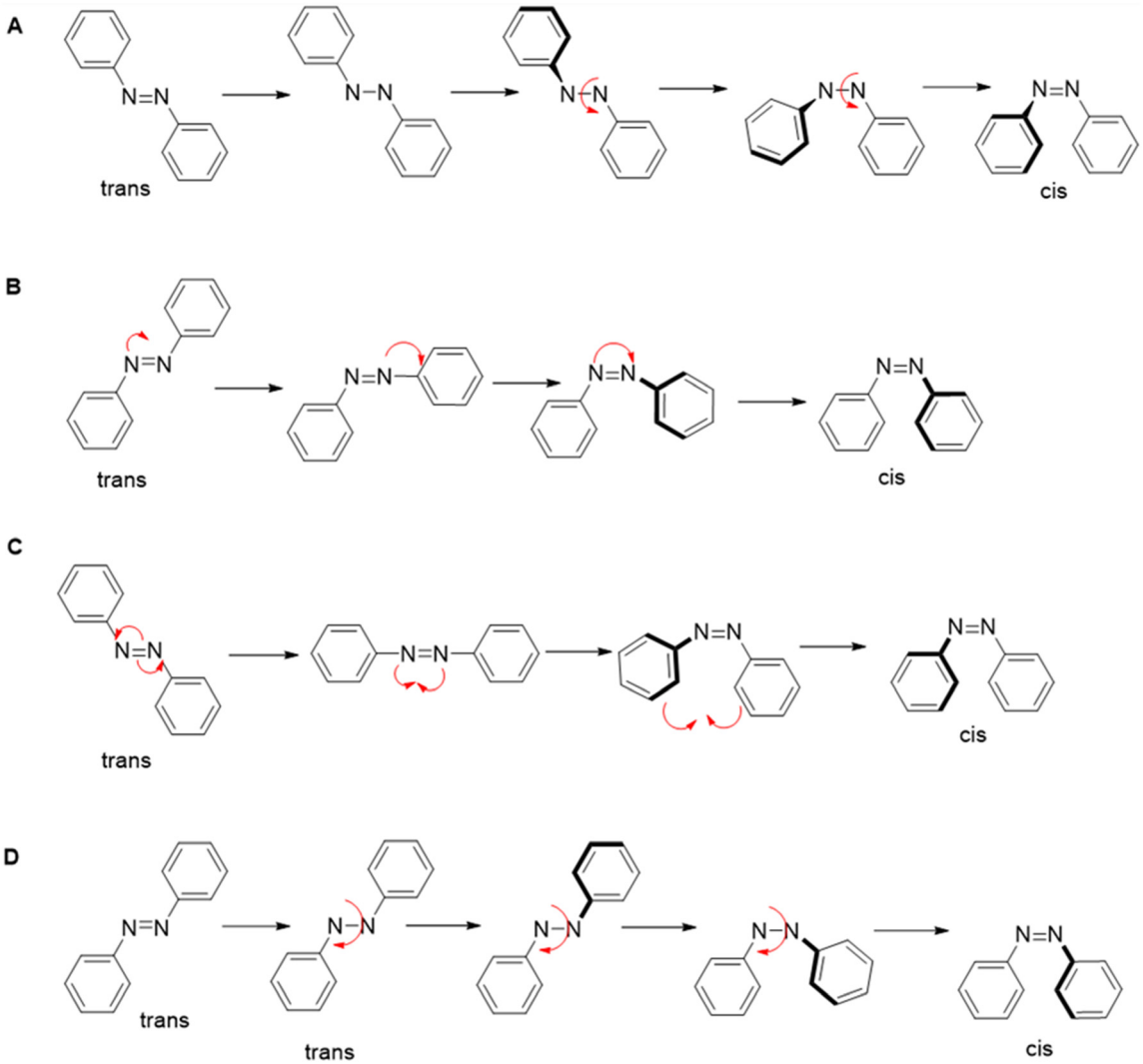


Fig. 3 Mechanism of *cis–trans* isomerization of azobenzene by (A) rotation, (B) inversion, (C) concerted inversion, and (D) inversion-assisted rotation.

facilitates rotation about the N=N bond by lowering the rotational barrier through partial inversion at one nitrogen atom. This pathway is frequently thought to be energetically advantageous under specific excited-state conditions. Depending on the electronic state (n to π* or π to π* excitation), solvent environment, and substitution pattern, these mechanisms can be selectively favored; rotation typically predominates in the excited state, whereas inversion contributes predominantly in ground-state thermal isomerization.

Depending on the direction of isomerization, the reaction conditions, and the substituents involved, azobenzene-containing molecules undergo isomerization *via* one of the above-mentioned paths. For azo compounds with a strong dipole moment, the rotational pathway has been observed, especially in polar solvents.[63] Due to the increased single-bond character of the N=N bond in the excited or transition state, it can be considered as partially weakened, which leads to the formation of the NH–N=hydrazone group. This process is called azo-hydrazone tautomerism. The formation of the hydrazone group dramatically increases the rate of *cis–trans* isomerization by significantly increasing the N–N bond rotational freedom. This process is especially observed in polar solvents such as ethanol.[57,64] The kinetics of surfactant isomerization differ significantly below and above the critical micelle concentration (CMC) of the *trans* isomer, primarily due to steric constraints within tightly packed micelles. However, this behavior is largely unaffected by variations in spacer length.[65]

Studies have investigated the effect of different substituents on the isomerization process. In one such study, Konieczkowska *et al.*[66] investigated 4-hydroxy and 6-hydroxy-4-hexyloyl azobenzene derivatives. They concluded that when a hydroxy group is attached directly to the azo moiety, it increases the rate of thermal *cis–trans* isomerization compared to the hydroxyalkoxy group. They also concluded that intermolecular interactions strongly influence *cis–trans* isomerization. The push–pull effect is also known to affect the *cis–trans* back reaction kinetics, particularly by increasing the rate. There have even been reports of compounds reverting to their *trans* state within

microseconds or faster after conversion to the *cis* state upon photoirradiation.[63,67,68] This makes it a suitable candidate for applications requiring fast-switching molecules.[69–72] Bichot *et al.*[73] synthesized 4,4′-disubstituted azobenzene compounds, reporting three model compounds with $t_{1/2}$ values of 57 h, 5 min, and 9 ms, respectively, highlighting the effect of substituents. Here, $t_{1/2}$ refers to the half-life for reappearance of the *trans* isomer after irradiation.

5. Effect of *cis*–*trans* isomerization on surfactant properties

Photoresponsive surfactants are important because of their isomerization properties in the presence of light. Hence, it is imperative to understand the effects of this isomerization. Miyata *et al.*[74] discussed the differences in the CMC and electrical conductivity between the *cis* and *trans* forms. According to them, there is a difference in the hydrophilic-lipophilic balance (HLB) between the *trans* and *cis* forms, and this difference leads to significant variations in the CMC values and electrical conductivities of the surfactant upon *cis–trans* isomerization. They found that surfactants with moderate alkyl chain lengths, such as ethyl and butyl groups, exhibited larger changes in CMC values of up to 5.9 mmol L^{-1} upon *cis–trans* isomerization. The photoresponsive function was similarly impacted by the substitution of the tail chain species. This included both the length and the type of tail chain. When compared to a structurally similar surfactant with an ethyl unit as its tail group, the surfactant with the *p*-ethoxy group as the tail chain was found to form stable micelle aggregation. Additionally, it showed a significant shift in CMC (5.3 mmol L^{-1}) when exposed to UV light.

The introduction of an azobenzene group in the tail has been a particular point of interest for both application and proof-of-concept points of view. In the case of charged surfactants, which constitute a large fraction of synthesized and commercially available surfactants, the charged head group has a relatively high effective cross-sectional area. As a result, the influence of the tail groups on the surface tension is reduced, as changes in tail conformation upon *cis–trans* isomerization have only a minor effect on surfactant packing.[75]

A very important change that occurs upon photoisomerization is the change in surface tension of the solution, which can be observed as a change in the contact angle of a droplet of the solution on a hydrophobic surface. The contact angle can be calculated using Young's eqn (1).

$$\gamma \cos\theta = \gamma_{SV} - \gamma_{SL} \quad (1)$$

where θ is the contact angle, γ_{SV} is the surface energy at the solid–vapour interface and γ_{SL} is the surface energy at the solid–liquid interface.

Another important change in surfactants upon light irradiation is the change in CMC. As shown by Shang *et al.*[75] for nonionic surfactants, their *trans* isomers show remarkably lower CMC than their corresponding *cis* isomers. This can be attributed to the greater packing efficiency of the *trans* form, which facilitates micellar formation at lower concentrations, whereas their *cis* isomers are not as well packed and require higher concentrations to come close enough to form micelles. In some cases, the difference is even as much as 15-fold from the *trans* to *cis* forms. A similar result was reported by Hayashita *et al.*[74] for ionic photoresponsive surfactants. Both Shang *et al.* and Hayashita *et al.* interestingly reported that, for a given surfactant with varying spacer or tail chain lengths, the maximum difference in the CMC values between the *cis* and *trans* forms was observed in the case of moderate alkyl spacer or tail chain lengths. In the case of azobenzene-based surfactants, it was also reported that the adsorption coefficient, *k*, was lower for their *cis* isomers. This was attributed to the more polar nature of the azobenzene group.

Shang *et al.* also demonstrated another interesting phenomenon. The quantum efficiencies of *cis–trans* isomerization and *trans–cis* isomerization were different. Their experiments on $C_4AzoOC_6E_2$ indicated that, for the *trans–cis* conversion, the quantum efficiency was close to unity, that is, $\phi \approx 1$. However, for *cis*-to-*trans* conversion, the quantum efficiency was found to be around 0.69. Their explanation for this was based on the geometrical differences in both the excited and ground states, suggesting that the differing quantum efficiencies may represent distinct reaction mechanisms in these two photoisomerization processes. This is consistent with the proposed mechanisms in which *trans*-azobenzene converts to the *cis* form *via* a rotation pathway, whereas the *cis* form reverts to the *trans* form through a slower inversion mechanism.[76]

The ground-state absorption spectrum of azobenzene shows two bands in the UV-vis region. The symmetry-forbidden S_1 ($n\pi^*$) ← S_0 transition appears as a weak band at about 450 nm, while the symmetry-allowed S_2 ($\pi\pi^*$) ← S_0 absorbs at around 320 nm.[59] Upon excitation, both the S_1 and S_2 states lead to *trans–cis* isomerization, but S_1 relaxes with a higher quantum yield of isomerization.[77] It has been shown by femtosecond time-resolved spectroscopic studies that internal conversion from S_2 to S_1 occurs with a quantum yield close to unity, indicating that it occurs before S_2 can relax to the ground state.[78] Azobenzene with steric constraints that block N–N bond rotation also exhibits a quantum yield independent of the excitation wavelength, suggesting that photoisomerization dominates from S_1 irrespective of the initial excited state.[79,80]

The S_1 state generated by S_1 ← S_0 excitation has a lifetime of 2.6 ps,[81] whereas that generated by S_1 ← S_2 is 500 fs.[78] The S_1 ← S_2 process creates a vibrationally excited S_1 state, leading to its shorter lifetime, and it relaxes rotationally to the *trans* form, resulting in a lower quantum yield of isomerization. While experimental and theoretical data agree that isomerization proceeds mainly from the S_1 state, when the S_2 state relaxes to the S_1 state, other isomerization pathways become possible. Theoretical studies suggest that these alternative pathways are a concerted inversion mechanism, defined by the simultaneous change on both C–N═N and N═N–C angles.[82,83]

Shang *et al.*[75] synthesized a new class of photoresponsive surfactants, $C_4AzoOC_nE_2$ (n = 2, 4, 6, 8). They conducted a series

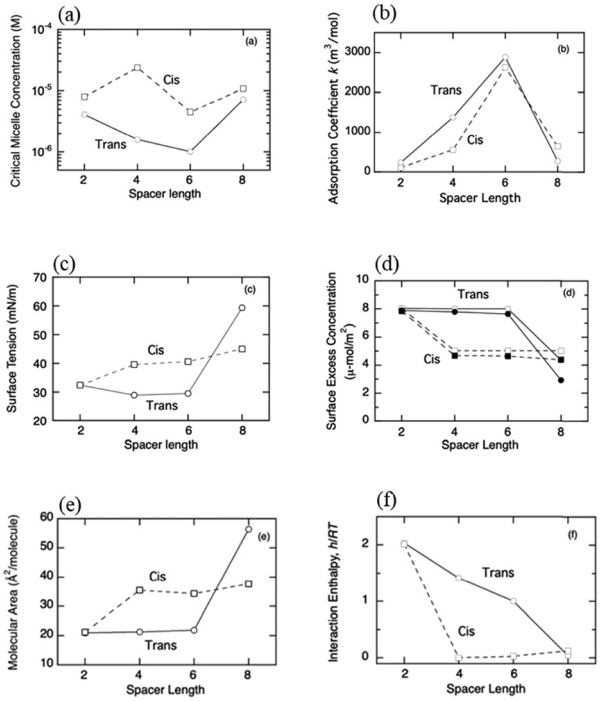


Fig. 4 Variation in photoresponsive surfactant solution properties with spacer length. Circles represent properties under visible illumination (*trans* isomer) and squares represent *cis* isomer properties under UV irradiation. (a) Critical micelle concentration; (b) adsorption coefficient, k; (c) surface tension, γ; (d) surface excess concentration, Γ_{cmc} (solid symbols) and Γ_∞ (open symbols); (e) molecular area at the CMC, $A_{s,cmc}$; and (f) interaction enthalpy, h/RT. (Reused with permission).

of studies on these compounds, including variations in the surfactant solution properties with varying spacer length. They used the Frumkin isotherm to relate the surface excess concentration to the true bulk concentration, employing the following eqn (2):

$$kC_b = \frac{\Gamma}{\Gamma_\infty - \Gamma}\exp\left[-2\left(\frac{h}{RT}\right)\frac{\Gamma}{\Gamma_\infty}\right] \quad (2)$$

where k is the limiting adsorption coefficient for the surfactant between the interface and the bulk solution at low surface pressure, C_b is the Frumkin adsorption isotherm, h is the infinite dilution surface partial molar heat of mixing of the surfactant at the interface, R is the gas constant, T is the absolute temperature, Γ_∞ is the saturation surface excess concentration and Γ is the surface excess concentration.

Further, the Gibbs adsorption isotherm was employed to relate the surface excess concentration to the slope of the surface tension curve in relation to the true bulk concentration, employing the following eqn (3):

$$\Gamma = -\frac{1}{RT}\frac{d\gamma}{d\ln c} \quad (3)$$

To determine the surface tension as a function of the surface excess concentration, the above equation was integrated and the Frumkin isotherm was further used to eliminate the true bulk concentration to get the following eqn (4):

$$\gamma = \gamma_0 + \Gamma_\infty RT\left[\ln\left(1 - \frac{\Gamma}{\Gamma_\infty}\right) + \frac{h}{RT}\left(\frac{\Gamma}{\Gamma_\infty}\right)^2\right] \quad (4)$$

where γ_0 is the surface tension of pure water.

The area occupied per molecule at the surface is calculated using the following eqn (5):

$$A_s = \frac{1}{\Gamma N_A} \quad (5)$$

where N_A is Avogadro's number.

Fig. 4 shows the trends observed for this new class of surfactants, derived using the above-mentioned equations and other experimental procedures, highlighting the effect of spacer length on photoresponsive surfactant properties.

6. Applications of photoresponsive surfactants

Surfactants are widely used in everyday life, as discussed before. Introducing sensitivity to light transforms them into a niche class of compounds with applications across a broad range of fields, from foams to drug delivery and metal detection to molecular machines.

6.1. Industrial applications

A huge number of surfactants have been synthesized for stabilizing foams and emulsions. Obviously, emulsion stabilization is an inherent property of surfactants and comes with their amphiphilic nature, which is again an inherent part of surfactants. Introducing a photoswitch gives us the ability to stabilize and destabilize emulsions, giving us a "switch" to turn the form "on or off." Many examples of these surfactants are available.[84–86] Wang *et al.*[32] synthesized 4-hydroxy-4′-oxoalkyl azobenzene (HC_nAzo, n = 4, 8, and 12), a class of compounds specifically to act as photofoam switches. They concluded that both photoresponsive qualities and the controllability of foam stability were impacted by the hydrophobic chain length. They reported higher foam stability for the *trans* form compared to the *cis* form. This dynamic behavior offers significant advantages in industrial applications where external, non-invasive triggers are essential, such as in controlled drug delivery,[87] oil recovery, smart coatings,[88] and responsive formulations in cosmetics and cleaning agents. Recent advances in the design of photoresponsive surfactants have focused on optimizing structural features to enhance their switching behavior and interfacial performance under light, providing new tools for precisely controlling multiphase systems.

Kang *et al.*[4] synthesized new surfactants with two azobenzene groups in their structure and studied their foam- and emulsion-stabilizing abilities. The two surfactants synthesized were qDAZDA-0 and qDAZDA-1. For 0.1% solutions of the *trans* forms of qDAZDA-0 and qDAZDA-1, after 5 min the foam volumes were 30 and 68 mL and 29 and 65 mL, respectively. However, for the *cis* forms, the foam volumes were 28 and 48

mL originally and 27 and 45 mL after they were left standing for 5 min, respectively. They also reported higher foam stability for the *trans* form compared to the *cis* form. Similarly recent articles also show the foaming ability of azobenzene as a core moiety for surfactants.[89,90]

Kondo *et al.*[91] reported a "hybrid" surfactant, where one hydrophobic chain was a hydrocarbon alkyl chain and the other hydrophobic chain was a fluorocarbon chain, and all hydrogens of the normal alkyl chain were replaced by fluorine atoms. They reported a tunable superhydrophobic nature due to the fluorocarbon chain and a central azobenzene group. One interesting result from their work was the effect of photoisomerization on surface tension, which was measured as surface tension variation ($\Delta\gamma$). They reported a surface tension variation of up to 30 mN m^{-1} for the surfactant containing an octylfluorocarbon chain.

Jia *et al.*[92] explored the design and control of photoresponsive complex emulsions comprising hexane and perfluorooctane, stabilized in water by a binary surfactant system of fluorosurfactant (Zonyl FS-300) and a synthesized azobenzene-based surfactant (C_4AZOC_2TAB). The emulsion morphology, including double and Janus emulsions, was reversibly modulated by varying the surfactant ratios and applying UV or blue light. The Marangoni effect explains how light-induced changes in interfacial tension resulting from surfactant photoisomerization drive these transitions. The system provided a remote way to modulate multiphase processes by acting as a light-controlled switch to control hexane evaporation. This method offers a straightforward, expandable framework for producing stimuli-responsive emulsions that may find use in chemical processing, drug delivery, and sensing.

An important application of photoresponsive surfactants is as phase-change materials (PCMs), specifically called photoresponsive phase change materials (PPCMs). Phase-change materials are substances that change their phase in response to stimuli, with the most commonly used stimulus being heat. However, heat cannot be used as a reliable stimulus in all applications, especially where heat can alter the properties of the surrounding system as well. Azo-based compounds are highly useful in these applications due to their reversible light-induced *trans–cis* isomerization, which can enable transitions between solid and liquid-like states.[93]

6.2. Medical applications

Due to light being a non-invasive stimulus, photoresponsive surfactants are gaining significant interest in medical applications, especially as drug carriers and in drug release systems.[94] These materials exhibit significant capacity to modulate their structural configuration, optical characteristics (including color and transparency), and physical properties in response to irradiation with light of a specific wavelength. Notably, these photoinduced transformations can be either reversible or irreversible, depending on the molecular design of the system and the irradiation parameters. This photoresponsive functionality enables highly controlled, site-specific, and temporally regulated drug delivery, thereby mitigating off-target effects and enhancing therapeutic efficacy.[95–97]

Recently, Wang and Chen *et al.*[11] demonstrated the potential of surfactants as drug carrier agents. They used a novel azobenzene-containing cationic surfactant that formed self-assemblies with the drug to be delivered. The drugs used for the studies were beta-cyclodextrin and the antitumor agent doxorubicin hydrochloride (DOX). Based on the photostimulated behavior of the surfactant, they reported that it can serve as a viable drug delivery agent due to its rapid photoresponsive drug release.

The use of photoresponsive nanocarriers (NCs) for targeted drug delivery, *via* either passive or active mechanisms, represents a promising approach in precision medicine.[98,99] A key concept in nanomedicine is the enhanced permeability and retention (EPR) effect, which takes advantage of the distinctive features of tumor physiology, such as leaky blood vessels. These structural abnormalities facilitate the preferential passage of larger entities such as NCs into tumor tissue. Additionally, tumors often have compromised lymphatic drainage, resulting in the retention and buildup of macromolecules and NCs within the tumor microenvironment. The EPR effect plays a pivotal role in the rational design and effective application of self-assembling NCs for cancer treatment. A micellar system can be designed to encapsulate an anti-tumor drug, which can be released upon light irradiation (Fig. 6).

Azobenzene and its derivatives are among the most extensively explored light-responsive chromophores, owing to their reversible photoisomerization behavior. Upon exposure to UV light, these molecules undergo a *trans–cis* isomerization around the nitrogen double bond, a process that can be reversed to the *trans* form using visible light. In contrast, other chromophores such as 2-diazo-1,2-naphthoquinone (DNQ) exhibit irreversible photoreactivity, where light exposure leads to bond cleavage, transforming hydrophobic compounds into hydrophilic ones.[100] Based on these mechanisms, light-responsive polymeric micelles (PMs) are typically classified into two categories: those that undergo reversible conformational changes and those that rely on irreversible photocleavage for structural destabilization.

Poelma *et al.*[101] reported polymeric micelles (PMs) that respond to one-photon visible light *via* a reversible structural transformation approach for the controlled, on-demand delivery of small molecules. They employed donor–acceptor Stenhouse adducts (DASAs), a class of photochromic compounds capable of undergoing a light-induced polarity shift from hydrophobic to hydrophilic when exposed to visible light in the 530–570 nm range. Compared to two-photon systems, one-photon responsive materials offer advantages such as more suitable activation wavelengths and higher efficiency, with DASA-based isomerization occurring at low light intensities ($\sim$1 mW cm^{-2}). In their work, DASA units were incorporated into PM structures, enabling visible light-triggered micellar disassembly and subsequent release of encapsulated small molecules into the cells.

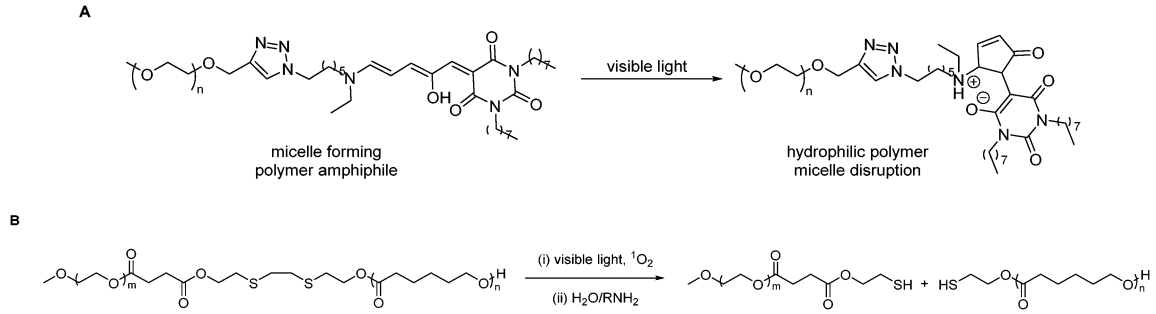


Fig. 5 (A) Photoswitching of micelle-forming polymer amphiphile. (B) Light-triggered bond cleavage of $^1O_2$-PEG-*b*-PCL.

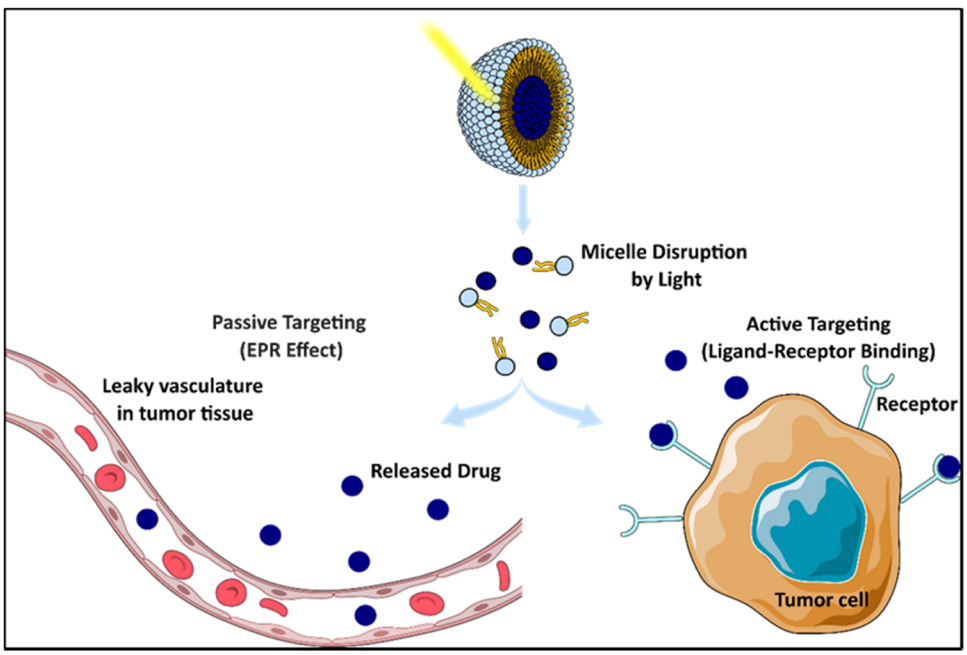


Fig. 6 Photoresponsive nanocarriers enable controlled drug release through passive and active targeting mechanisms.

Beyond reversible photoisomerization, light-triggered bond cleavage is another widely applied strategy in designing light-responsive polymeric micelles (PMs)[102] Saravanakumar *et al.*[103] developed a biocompatible amphiphilic block copolymer, $^1O_2$-PEG-*b*-PCL, which self-assembles into PMs featuring a singlet oxygen ($^1O_2$)-sensitive vinyldithioether linker at the core–shell interface. Upon exposure to visible light, this linker undergoes $^1O_2$-mediated photocleavage, enabling the controlled release of both singlet oxygen and anticancer agents, thereby enhancing the effectiveness of photodynamic therapy. The mechanism involves the double bond of the vinyldithioether moiety reacting with $^1O_2$ to form an unstable dioxetane intermediate, which rapidly breaks down. Unlike traditional photoresponsive micelles, which require direct UV activation, these PMs rely on $^1O_2$ generated under visible light during photodynamic therapy to trigger disassembly and release their therapeutic cargo within tumor cells (Fig. 5).

Unksov and Kasyanenko[104] explored the conformational changes in DNA upon interaction with a light-responsive cationic surfactant, azobenzene trimethylammonium bromide (AzoTAB), which undergoes reversible *trans–cis* isomerization under UV irradiation. Binding of AzoTAB to DNA induced compaction of the DNA strands, with the extent of compaction strongly influenced by both the isomeric state of the surfactant and the ionic strength of the surrounding medium. At lower NaCl concentrations (0.003 M and 0.005 M), the *trans* isomer promoted more pronounced DNA compaction and significant reductions in macromolecular volume and solution viscosity. In contrast, the *cis* isomer also induced DNA condensation but required higher surfactant concentrations to induce comparable phase separation, and the associated changes in volume and viscosity were comparatively less pronounced. The photoisomerization of AzoTAB enabled reversible modulation of DNA conformation, as UV light facilitated the transition between the *trans* and *cis* forms, thereby offering a dynamic and controllable method of influencing DNA structure. These findings provide key insights into the manipulation of DNA–surfactant complexes, with potential applications in gene therapy,

nanobiotechnology, and the development of light-controlled biomolecular systems.

Photosensitive surfactants have also been used to increase the efficiency of enzyme action. Seidel *et al.*[105] investigated the use of AzoTAB to enhance the activity of β-glucosidase from *Aspergillus niger*, an enzyme crucial in converting cellulose to glucose in bioethanol production. AzoTAB shifts the enzyme from dimers to monomers, increasing activity by 60%, unlike traditional surfactants, which deactivate it. Small-angle neutron scattering (SANS) data revealed selective unfolding at the dimer interface, reducing substrate inhibition and improving catalytic efficiency. This approach highlights a novel strategy for creating glucose-tolerant β-glucosidases to boost biomass conversion.

In their recent work, Wang *et al.*[106] introduced azobenzene-bridged ionizable amphiphilic Janus glycosides (IAJGs). These conjugates possess charged amine heads and long alkyl chains attached to the glucose rings, imparting amphiphilic character. These IAJGs were shown to independently form stable nanocomplexes with plasmid DNA. Photoisomerization can be used to spatiotemporally control the size, surface charge, and internal organization of the nanocomplexes. This allowed them to achieve distinct cellular transfection results, characterized by programmable, organ-modulable pDNA delivery. This work paves the way for precision gene therapy and targeted nucleic acid delivery.

6.3. Environmental pollution detection and remediation

Environmental contamination poses significant challenges that threaten both ecosystems and human health. Addressing these challenges has prompted researchers to explore innovative approaches, such as using photoresponsive surfactants for environmental remediation and pollutant detection. Photoresponsive surfactants are unique amphiphilic molecules featuring a hydrophilic head and a hydrophobic tail that self-assemble into micelles. These micelles can encapsulate various hydrophobic pollutants, and upon exposure to specific wavelengths of light, their photochromic groups undergo structural changes. This transformation can trigger the disassembly of the micelles and the subsequent release or activation of the encapsulated agents, making them potent tools for both remediation and detection.

In environmental remediation, one promising application involves the degradation of persistent organic pollutants such as polycyclic aromatic hydrocarbons (PAHs) in contaminated groundwater. For instance, in 2023, Dai and Li *et al.*[8] developed a system that combines a photoresponsive surfactant with peroxymonosulfate (PMS) to remediate PAHs. In their study, the application of a directed light beam activated the surfactant, which in turn accelerated the generation of reactive species that broke down PAHs efficiently. This approach not only enhanced degradation efficiency but also minimized adverse effects on the surrounding ecological systems by leveraging a controlled light-triggered activation mechanism. Beyond remediation, photoresponsive surfactants also offer significant promise in pollutant detection. When integrated into sensor platforms, these surfactants can provide real-time detection signals through changes in optical properties such as shifts in fluorescence or absorbance triggered by micellar assembly or disassembly. The dynamic behavior of the micelles allows for the encapsulation of pollutants followed by light-induced release, which can serve as a measurable signal for pollutant presence. This dual function, capturing and then signaling, improves the sensitivity and accuracy of environmental monitoring systems. These capabilities were further emphasized in a comprehensive review by Saxena *et al.*,[107] which discussed the versatility of surfactant-based approaches in removing contaminants ranging from heavy metals and dyes to pesticides through enhanced photo-induced micellar solubilization.

However, while the benefits are clear, the potential toxicity and environmental impact of surfactants must also be addressed. Badmus *et al.*[108] highlighted the importance of evaluating the fate of both surfactants and their degradation products to avoid secondary contamination. Their analysis underscores the need for developing biodegradable and eco-friendly surfactants that maintain high remediation efficiency without adverse environmental footprints (Fig. 8).

The unique structures and different chemical moieties in these molecules endow them with a few very intriguing properties in terms of binding with other molecules *via* non-covalent interactions. Dai and Duan *et al.*[38] synthesized a conventional surfactant, 4-[4-[(4-butylphenyl)azo]phenoxy]-butyldimethylethylammoniumbromide (AzoPB) and a gemini photoresponsive surfactant, N^1,N^2-bis[4-[4-[(4-butylphenyl)-azo]phenoxy]butyl]-N^1,N^2-tetramethyl ethane-1,2-diammonium-bromide (AzoPBT). These two surfactants were studied in relation to phenanthrene and acenaphthylene, two common polyaromatic hydrocarbons that pollute groundwater. The surfactants were studied for their ability to solubilize these hydrocarbons *via* self-assembly and to dissipate them upon subsequent disrupion of these assemblies, a process facilitated by the photoresponsive azobenzene moiety. They found that the gemini surfactant was significantly better than the conventional surfactant in terms of surface activity, recyclability, reducing the hydrophobicity of hydrocarbons and subsequently solubilizing them, and release of solubilized hydrocarbons. Accordingly, they concluded that the gemini surfactant was superior to conventional surfactants in these applications.

Surfactants are also known for their metal detection activities, as shown by Xie and Wu *et al.*[13] They synthesized a surfactant capable of selectively detecting Hg(II) ions in UV spectroscopy, in the form of a characteristic UV-vis signal change in the band at around 240 nm, and this change was visible over a wide pH range. Additionally, the surfactant was water soluble owing to its cationic head, making it an attractive option for mercury detection in water. Photoresponsive surfactants integrate the capabilities of advanced pollutant detection and targeted remediation, paving the way for next-generation environmental cleanup technologies.

6.4. Motion manipulation

The precise manipulation of colloidal particles within microfluidic environments is a central challenge in soft matter and biomedical applications. Recent advances have demonstrated the integration of optically tunable flow mechanisms using photoresponsive surfactants to achieve fine control over fluid motion. One such approach utilizes a gold surface in aqueous media, coated with a photosensitive surfactant and irradiated by a focused UV laser. This setup induces two distinct interfacial flow regimes arising from diffusion-osmosis and thermo-osmosis.

At lower laser intensities, diffusion-osmotic flow dominates due to localized photoisomerization of the surfactant molecules. This optofluidic strategy thus offers a highly controllable platform for the dynamic regulation of microscale flow fields, presenting significant potential for targeted particle transport, on-demand assembly, and programmable microfluidic operations in next-generation lab-on-a-chip technologies.[109]

Recent advances demonstrate versatile light-controlled manipulation of microscopic droplets, including wetting, splitting, merging, and transport, using photoswitchable azobenzene-based surfactant films. These adaptive surfaces, formed by ionic assembly of azobenzene surfactants with oppositely charged polymers, undergo reversible *trans–cis* isomerization under light irradiation. This induces dynamic changes in surface energy, orientation of surfactant tails, and local temperature, enabling precise, reversible control over droplet behavior. Notable outcomes include a fivefold increase in droplet basal area, directional wetting, and controlled droplet locomotion at velocities up to 150 μm s^{-1}. This approach presents a promising platform for programmable, contactless microfluidic operations.[110]

A very interesting study was recently published in 2024 by Luzzatto-Fegiz, Alaniz and Zhu *et al.*,[37] in which they discussed the dynamic manipulation of water droplets on surfaces. The group used photoresponsive surfactants for their surface-acting properties and photoswitchability to show 2D motion of water droplets on a liquid-infused surface as well as rectilinear motion in capillary tubes. The designed surfactants were based on spiropyran and merocyanine, which were interestingly capable of surface tension changes of up to 5.5 mN m^{-1} on very short timescales of about 1.7 s. They were able to move a millimeter-sized droplet at a speed of up to 5.5 mm s^{-1} on liquid and 0.25 mm s^{-1} on a liquid-infused surface (Fig. 7).

While the manipulation of colloidal droplets or synthetic microbeads is certainly impressive, Umlandt *et al.*[111] took it to the next level by demonstrating the manipulation of living microorganisms using a simple azo-based surfactant. This suggests that the technique is non-invasive and gentle enough to sustain biological entities. They used light to trigger the characteristic isomerization of azobenzene, which generated an osmotic pressure gradient that pulled the liquid and the bacteria within it toward the light, demonstrating light-driven diffusion-osmotic (LDDO) flow. This has implications for programmable micro-bioreactors or light-controlled environmental remediation, where cyanobacteria blooms can be removed from water samples using light-responsive smart surfactants.

6.5. Catalysis

Owing to their stabilization properties and micelle formation, surfactants are increasingly applied in catalysis. Many of these properties arise from their mode of action being similar to that of phase-transfer catalysts due to the stabilization of a mixture of phases by surfactants and the tunable stabilization-destabilization by photoresponsive surfactants without using any chemical additives. Micelles function as heterogeneous nanoreactors, with a hydrophobic interior and a hydrophilic exterior, all suspended in a seemingly uniform aqueous environment.[112] Their structure enables the solubilization of both organic reactants and catalysts, primarily through interactions such as hydrophobic forces[113] and ion pairing.[114]

Blayo *et al.*[115] studied the effect of micellar size and shape on micellar catalysis of a model Claisen–Schmidt aldol condensation reaction by cationic azobenzene trimethylammonium bromide (AzoTAB) in water. They concluded that the reaction efficiency depended on the ratio of micelle volume to micelle–water interfacial area (the Stern layer), with the compact, spherical micelles formed by *cis*-AzoTABs exhibiting enhanced reaction yields. *Trans*-AzoTAB forms ellipsoidal micelles, which showed lower reaction yields. They also concluded that the reaction occurred at much lower temperatures of 35 °C and milder conditions than conventional setups (Fig. 9).

Li *et al.*[116] developed oil-in-water Pickering emulsions for catalysis. A light-responsive Pickering emulsion was constructed using Pd-loaded silica nanoparticles, an azobenzene-based ionic liquid surfactant, *n*-octane, and water. UV and visible light caused this emulsion to undergo reversible emulsification and demulsification, which was attributed to the azobenzene surfactant. The system enabled catalytic hydrogenation under ambient conditions, while also allowing product separation and reuse of both the emulsifier and catalyst. Catalysis using photoresponsive surfactants is still a very new field of work, and consistent efforts are being made by many groups to find new information every day.

Similarly, many photoresponsive surfactants have been widely used in a variety of different fields, making it infeasible to mention them all. These applications span diverse areas, including engines, drug delivery systems, biological systems, water purification, lithography, and sensors. In all these areas, surfactants, particularly photoresponsive surfactants, are actively employed in industrial and research settings.

6.6 Harnessing solar energy

Krishna *et al.*[117] introduced the first class of visible-light responsive solar thermal fuels (STFs) based on liquid crystals, utilizing a series of azobenzene- and cholesterol-based surfactants that exhibit enantiotropic chiral nematic mesophases. This LC-based approach helped them overcome the drawbacks of solid-state STFs, such as light scattering and nonuniform

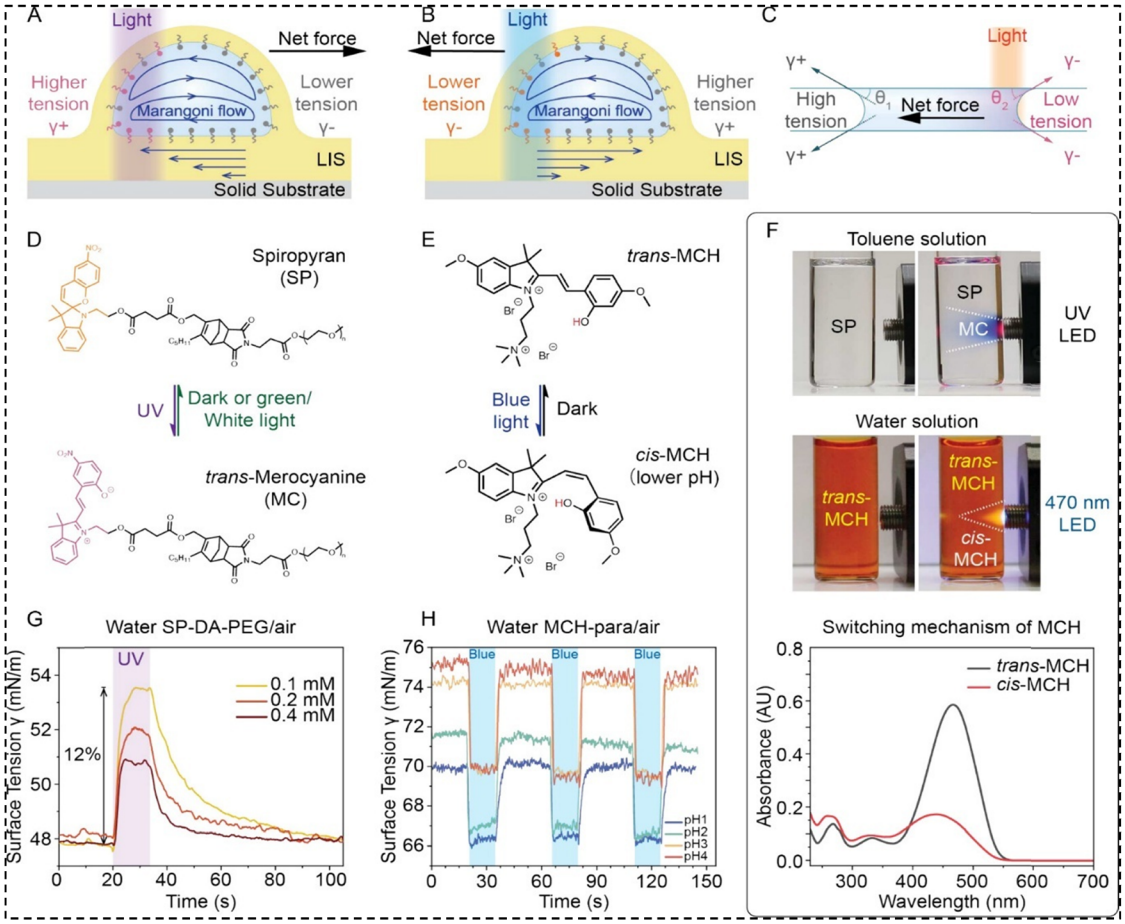


Fig. 7 Mechanisms of liquid movement driven by photoresponsive surfactants, molecular structures of the photoresponsive surfactants, and surface tension response. (A) Schematic of a lubricant-cloaked droplet on a liquid-infused surface (LIS). The interfacial tension increases under illumination, causing a Marangoni flow from the unilluminated region to the illuminated region, and a net shear force away from the light. (B) Schematic of a lubricant-cloaked droplet on LIS. The interfacial tension decreases under illumination, causing a Marangoni flow from the illuminated region to the unilluminated region, and a net shear force toward the light. (C) Schematic of liquid in a microchannel or capillary tube. The surface tension changes under illumination, which results in an unbalanced total surface tension force on the liquid column and the subsequent liquid movement. Molecular structures and photoswitching of (D) SP-DA-PEG and (E) MCH-*para*. (F) Change in the color of SP-DA-PEG in toluene under UV illumination and MCH-*para* in an aqueous solution (phosphate buffer, pH 3) under 470 nm illumination. UV-Vis spectrum demonstrating the switching mechanism of MCH-*para* in an aqueous solution (pH 1). (G) Surface tension response of SP-DA-PEG in water under UV (365 nm) illumination with an optical intensity of 37.1 mW cm^2. (H) Surface tension response of MCH-*para* in water at various pH levels under blue light (470 nm) illumination with an optical intensity of 31.8 mW cm^2.[37]

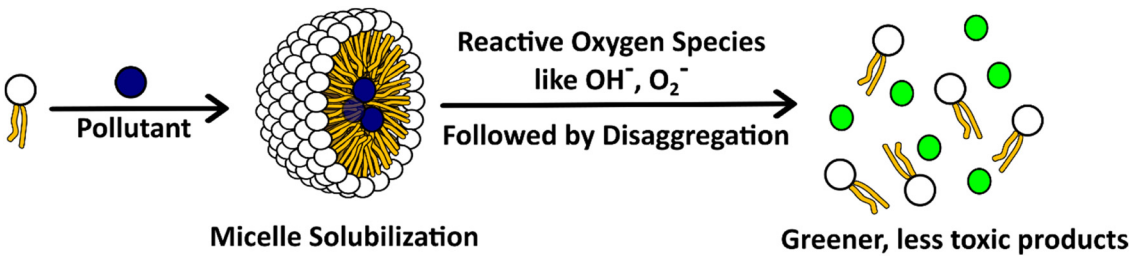


Fig. 8 Mechanism of pollutant conversion into green products through ROS generation by photoresponsive surfactants.

heat release, by introducing self-organized and self-healing thin films. They achieved a maximum solar charging efficiency of 70% and a record thermal half-life of 110 days for *ortho*-fluoro-substituted derivatives. Additionally, the transition through multiple phase states during discharging yielded a macroscopic heat release of 5.4 °C, a significantly larger amount than previous attempts.

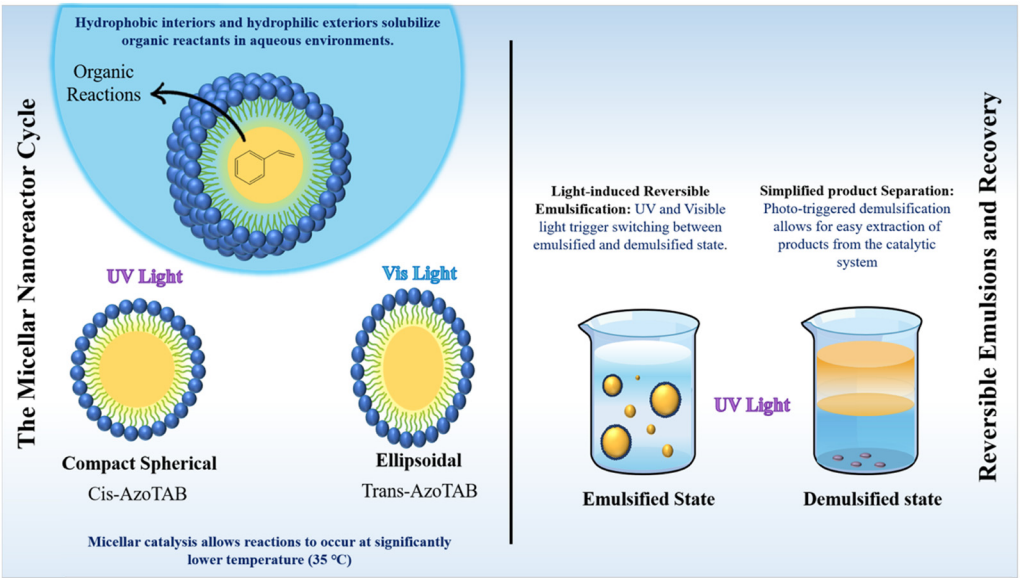


Fig. 9 Light-controlled micellar nanoreactors enable reversible emulsification and efficient catalytic reactions with easy product recovery.

In a recent study, Jones *et al.*[118] were the first to use lyotropic liquid crystals (LLCs) for molecular solar thermal (MOST) energy storage. The authors drove the self-assembly of azobenzene photosurfactants into highly ordered liquid crystalline phases through the sparing addition of solvent. This helped reduce the steric hindrance that usually stops photoisomerization in rigid solid-state systems. They used small-angle X-ray scattering (SAXS) and *in situ* differential scanning calorimetry (DSC) to show that these light-responsive LLCs allowed isomerization without considerable hindrances. This work demonstrated a record energy storage density of 123 J g^{-1}. This hybrid method works well because it combines the high isomerization efficiency of liquids with the structural benefits of solids. This makes it a very interesting framework for the next generation of solar fuels.

Chen *et al.*[119] recently addressed the environmental concerns associated with lipophilic STFs, which use toxic organic solvents to function, by developing a family of water-soluble azobenzene derivatives (WASTFs) through a one-step quaternization strategy. WASTFs use the intra- and intermolecular cation–π interactions to stabilize the *cis* isomer. This resulted in a thermal half-life exceeding 30 days and a maximum energy density of 143.6 J g^{-1}. The authors reported a 2.5-fold increase in storage enthalpy compared to neutral analogues.

7. Future outlook

The field of photo-responsive surfactants has evolved from a niche academic curiosity into a rapidly expanding area of materials science with significant potential. While substantial progress has been achieved, the next phase of development must address several unresolved scientific and technological challenges to enable the translation of laboratory-scale concepts into real-world applications. Despite the wide range of demonstrated applications, critical gaps remain in mechanistic understanding, molecular design, and system-level integration that must be systematically addressed to fully realize their practical potential.

An obvious path forward is the diversification of the photoresponsive core. Although azobenzene continues to dominate due to its synthetic accessibility and reliable switching behavior, its applicability, particularly in biological contexts remains limited. A key unresolved issue is the incomplete understanding of how *cis–trans* isomerization pathways are influenced by complex environments, such as micellar assemblies, interfaces, and biological media, where deviations from idealized behavior are frequently observed. Furthermore, systematic comparative studies between azobenzene and alternative photoswitches remain scarce, hindering the establishment of universal design principles. In response, increasing attention is being directed towards red-shifted photoswitches such as spiropyran, diarylethene, and hemithioindigo, which enable activation using visible or near-infrared light. While these systems offer clear advantages, challenges related to their long-term stability, fatigue resistance, and switching efficiency under practical conditions remain insufficiently explored.

The development of multi-stimuli-responsive surfactants represents another emerging direction, where light, pH, temperature, and other environmental triggers are combined to achieve enhanced control. However, the interplay among multiple stimuli is not yet fully understood, particularly in terms of synergistic *versus* competing effects in complex systems. In parallel, increasing attention must be paid to the environmental impact and lifecycle of these materials, especially for large-scale or industrial applications. The design of inherently biodegradable and environmentally benign photo-responsive surfactants remains a significant unresolved challenge.

Orthogonal photocontrol, achieved through the incorporation of multiple photoswitches within a single molecular framework, offers exciting opportunities for wavelength-selective and sequential control of processes. Nevertheless, precise control over cross reactivity and spectral overlap remains a key limitation that must be addressed.

In the context of biomedical applications, particularly drug delivery, photoresponsive surfactants have demonstrated considerable promise. However, their translation into *in vivo* systems is still limited by insufficient understanding of their biocompatibility, toxicity, and long-term stability under physiological conditions. The integration of therapeutic and diagnostic functionalities represents a logical progression, yet challenges remain in achieving reliable, real-time monitoring without compromising system performance. Beyond biomedical applications, photo-responsive surfactants hold potential in soft matter systems, including self-healing materials, hydrogels, and microfluidic devices. While these concepts are highly promising, their practical implementation is constrained by limited control over spatiotemporal responsiveness and system robustness under repeated cycling conditions. Recent advances in computational approaches, including density functional theory (DFT) and machine learning, offer powerful tools for molecular design. However, their predictive capabilities are currently restricted by limited experimental datasets and the absence of standardized structure property relationships, necessitating closer integration between computational and experimental efforts. A major bottleneck for industrial translation remains the reliance on multistep, time-consuming, and costly synthetic routes, which hinder scalability and widespread adoption. The development of efficient, cost effective, and scalable synthetic strategies, along with the incorporation of green chemistry principles, remains an urgent and unresolved priority.

Overall, the future of photoresponsive surfactants lies in addressing these fundamental challenges while advancing toward increasingly complex, functional, and application-oriented systems. The coming decade is expected to witness a transition from primarily curiosity-driven research to rationally designed, robust, and scalable systems capable of addressing real-world problems in medicine, materials science, and environmental technology.

Author contributions

Mittal Gunjan Lokesh and Shivank Sharma contributed equally to this work and share first authorship. Both authors were involved in literature survey, conceptualization, writing, and revision of the manuscript. Amit Kumar Tiwari: conceptualization, validation, resources, writing – review & editing, visualization, and supervision.

Conflicts of interest

There are no conflicts to declare.

Data availability

This review article does not report new experimental or computational data. All data discussed are derived from previously published literature, which is cited within the article.

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