NanoScience Technology, Volume 10 (2023), pp: 1-32

NanoScience Technology

Journal homepage : https://jnanoscitec.com

Micelles-based systems and their versatile application in different industries

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Abstract

Polymeric Micelles with their special chemical conformations which pose both hydrophobic and hydrophilic structures, provide valuable properties that enable them to be used in various applications either academic sciences or industrial aspects. They can offer useful features, such as increasing the solubility, stability, bioavailability, and compatibility for a wide category of materials. In this review, the important types of micelles including normal, reverse, and unimolecular micelles were first mentioned, and then their popular applications in medicine, cosmetics, detergents, agriculture, environment, food, district heating/cooling fluids, and oil and petroleum were discussed. Among different applications, micellar systems are appropriate systems to be served as significant vehicles in the delivery of various ingredients and biomolecules. Their applications are more common in the medical, food, cosmetic, and detergent industries. However, by improving their physicochemical properties with novel methods there is a hope to introduce new generations of smart micellar systems with more applications.

Keywords: *Micelles, Nanotechnology, Nanocarrier, Polymeric Micelles*

© Article info: Accepted by: 1 June 2023, Published by: 16 June 2023.

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1. Introduction

Nanotechnology, a widespread science, can be defined as a technique that uses materials that are nanometer-sized (scale of 1–100 nm, or occasionally 1 nm). Engineering, material manipulation, measurement, and organization on a nano-scale are the foundations of this technology [1]. Some experts predict that nanotechnology will usher in the following industrial uprising (by 2024, it is expected that the burgeoning global nanotechnology market would have exceeded US$ 125 billion) and be useful in a variety of fields. Products with nanotechnology capabilities are used in various industries which include consumer and home goods, energy, materials, technology, agriculture, medicine, and environmental science. Nanomaterials (including nanosystems), nanodevices, and nanotools are different forms of nano-based conformations [2].

Nowadays, for the creation of multifunctional nanosystems, such as nano drug/gene delivery in nanomedicine scope, nanocatalysts, nanocarriers for imaging probes, and nano-scale devices for data processing and storage, supramolecular structures created through the self-assembly of polymers with numerous functionality units, have recently received increasing interest [3-6]. Polymeric micelles (PMs), which are often constructed by block copolymers self-assembly, are representative of these kinds of nanosystems [7]. Historically, since the 1990s and even the late 1980s, PMs(PMs) have been extensively explored for the creation of self-assembled nanocarrier systems [8]. Generally, micelles are constructed from unimers that contain a hydrophilic head and a hydrophobic tail. These unimers are self-assembled in an aqueous media and form micelles with amphiphilic features [9].The exceptional amphiphilic copolymer-formed nanoscale core-shell structures have distinguished themselves as regulated, tumor-targeted nanocarriers with strong translational potential [10]. Surfactant structures with self-assembled features, those found in PMs, can be utilized to elevate particular rheological features in the solution, like viscoelasticity and shear-thinning behavior. These characteristics are crucial for a wide range of applications in different industries, such as medicine, detergent, and heating and cooling industries [11]. It is of note to mention that the use of micellar nanocarriers to reduce the toxicity of COVID-19 therapeutic drugs and their transfer was investigated in a study conducted by Manuja et al., by which they suggested the use of this type of nanocarriers could be useful for COVID-19 treatment [12]. In addition to carrying drugs, micellar nanocarriers are widely used to deliver biomolecules such as genes [13] and proteins, as well as evaluation of enzyme activity [14]. The application of nano-micelles is not only limited to the pharmaceutical industry but is also in the oil industry and environmental applications, such as neutralizing waste materials dumped in nature and wastewater treatment.

Despite micelle's pros (including size plasticity, which can be managed by selecting the length of the polymer, and self-assembly), their instability in response to environmental changes must be considered [15]. However, with strategies like covalent and noncovalent crosslinking micelles stability can be elevated in harsh environments [16]. For the special characteristics of micellar systems and their applications in different fields, this paper will discuss their different conformational structures with a focus on discussing their applications in industries.

2. Fundamentals

2.1 Types of micelles

Micelles are amphiphilic molecules with a size of 5 to 100 nm. The amphiphilic property of micelles is due to having unimers with hydrophilic heads and hydrophobic tails. When these unimers are placed in the water environment, they show a specific orientation, so that the hydrophilic heads move toward the water and the hydrophobic tails move away from the water's surface. In a certain amount of micelles' concentration, called critical micelle concentration (CMC), unimers start to join together [17]. In other words, CMC is the lowest amount of amphiphilic polymers in a solution at which the amphiphiles start to aggregate [18]. On the other hand, when the concentration of polymers is higher than the concentration of CMC in the solution self-assembly will start. Since the aggregation of micelles is kinetically favorable this phenomenon is also known as self-assembly**.** This aggregation eventually leads to the final formation of micelles, which consist of a hydrophobic core and a hydrophilic corona [17,19]. The self-assembly feature of micelles is desirable from an industrial point of view because it reduces production costs in comparison to using some cosurfactants or synthetic chemicals needed for micelles' conformation [20]. Table 1 shows more common types of compounds comprising the core and corona (outer layer) of micelles.

There are some types of classifications for micelles. For example, in a common type, micelles are categorized into two groups based on their size:

I. Star-like or hairy micelles or core-shell micelles. (In this kind of micelles, the soluble block is larger than the insoluble block)

II. Crew-cut micelles. (Here, the insoluble block is larger than the soluble block)

In hairy micelles, the core is smaller than the corona, whereas in the crew–cut micelles, the cores are thicker than the corona (Figure 1). Moreover, star-like micelles exist only in the spherical form, however, crew–cut micelles have been seen in spherical, cylindrical, and vesicle forms [21].

Table 1. List of organic substances that make up micelles reviewed by Kumar et al [22].

Material	Abbreviation
Hydrophilic shell-forming organic materials	
poly(ethylene glycol)	PEG
poly(vinyl alcohol)	PVA
poly(N-vinyl-2-pyrrolidone)	PVP
poly(acrylic acid)	PAA
poly(acrylamide)	PAAm
poly(glycerol)	PG
poly(amino acid)	PAA
Hydrophobic core-forming organic materials	
Polyesters	
poly(glycolic acid)	PGA
poly(ε-caprolactone)	PCL
poly(D,L-lactic acid)	PDLAA
Polyethers	
poly(ethylene oxide)	PEO
poly(propylene oxide)	PPO

On the other hand, according to the direction of the unimers after aggregation and the number of copolymers in the micelle composition, micelles are divided into 3 types including normal micelles (direct micelles), reverse micelles, and unimolecular micelles [23] which are described as blew:

- **Normal micelle (direct micelle)**

Normal micelles are formed in polar solvents and used in the delivery of hydrophobic or insoluble drugs. In this group of micelles, the hydrophilic heads are

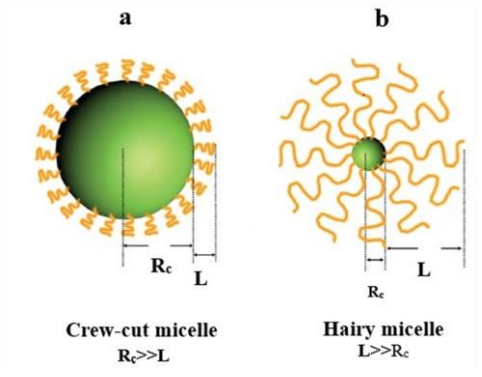

Figure 1. a) Crew-cut micelle b) Hairy micelle
L represents the coronas' thickness and Rc represents the core radius [21].

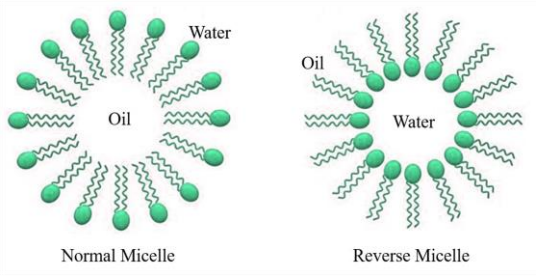

Figure 2. Schematics of normal and reverse micelles.

drugs, delivering food to patients who are in a coma, as well as protein transfer applications [23,24], NMR spectroscopy of proteins and nucleic acids [30-32], infrared spectroscopy of proteins [33], photocatalytic activity to remove pollutants [34] , and catalytic investigation of different enzymes such as lignin peroxidase [35]. Figure 2 illustrates schematics of normal and reverse micelles [36].

- **Unimolecular micelle**

Unimolecular micelles are called “Unimolecular” due to having a single block copolymer containing several hydrophilic and hydrophobic regions on a single molecule (polymers such as polyethylene glycol (PEG)). Figure 3 shows the structure of this type of micelles [23]. These single molecules, like other micelles-forming unimers, have the property of self-assembly, but their CMC is not obvious [23,24]. Since unimolecular micelles have a high capacity to load medicine such as hydrophobic and hydrophilic drugs [37], unimolecular micelles are used as drug carriers for cancer treatment and diagnosis [38-40]. Simple and more complex (star-shaped) forms of these micelles are applicable in different bioimaging methods [37,41] and used as nanotheranostics in treatment and diagnosis [37].

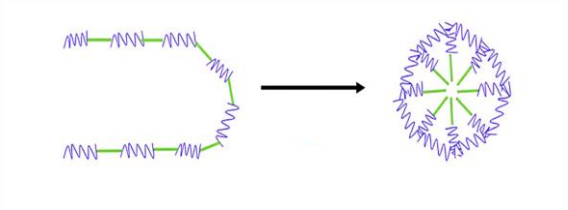

Figure 3. Schematic illustration of unimolecular micelles[23].

2.2 Mechanisms of micelle formation

Self-assembly is a type of relaxed arrangement that is seen in biological systems. Many biological micro- and nano-structures are also formed by this model of the assembly such as micelles [42,43]. Depending on the processing types, this phenomenon is done kinetically or thermodynamically [42]. Even though scientists have good information about the factors that stimulate the initiation of self-assembly, the mechanism of micelles formation has not been fully clarified [42]; what is known is that micelles are not a fixed system and are constantly falling and forming, and there is a dynamic balance between the micelles monomers and the micelles themselves. The formation mechanism of micelles is investigated with two different models including the phase separation and the mass action model [44].

- **Phase separation model**

In this model, the CMC is considered equal to the concentration of the monomer concentration in the saturated state, and it is assumed that the activity of the surfactant is more than the CMC. In this form, three parameters C_m, C_M, and C_T, representing the concentration of monomers forming micelles, the concentration of micelles, and the total concentration, respectively, are examined and their relationships are considered as follows:

If the total concentration is smaller than the CMC, then the concentration of the monomers forming the micelles will be equal to the total concentration.

If the total concentration is greater than the CMC, then the concentration of the monomers forming the micelles will be equal to the CMC and also the concentration of the micelles will be equal to the difference between the concentration of the monomers forming the micelles and the total concentration.

- **Mass action model**

In the mass action model, the following equation is used:

$$K = C_M a_M/(C_m a_m)^n$$

where "a" donates the activity coefficient related to the concentration and K is the equilibrium constant. This

model is very complex and can be treated analogously to a variety of homopolymerization processes. Nevertheless, there is an unanticipated disadvantage: the number of changeable parameters that must be fitted or inferred from experience increases with model complexity. This has made it impossible to consider the mass action model superior to the phase separation model. There are different opinions about the use of these two models in different studies. However, the truth is that both models give similar results and there is not much difference in the results of their use [44].

In view of the fact that the mechanism of micelles' formation remains unknown, it can be investigated to some extent by the simplified method of placing parallel plates inside a solution [45]. In the first step, the plates are spaced apart enough for the polymer density in the mid, or at halfway between the plates, to be matched to the bulk density. Additionally, close to an individual surface, the polymer density will be lower than it is in bulk. Since conformal polymer molecules that would typically pass the depletion interface are not included, density depletion occurs. When the plates approach each other, free energy and the lower polymer density close to the plates are still unaltered. Nevertheless, with the continuation of the phenomenon of the two plates approaching each other, the polymer density of the parallel plates cannot increase to the bulk density, then the second stage of formation will begin. As the two plates approached each other the maximum density of the polymer is uniformly reduced. So, polymer molecules distribution is less uniform which increases the free energy. In the last step of forming micelles, the distance between the plates being shorter than the width of the polymer molecules which means that no polymer can be located between the plates and react and dissolve with the external solvent. Due to the fact that dilution has negative free energy, this makes the free energy decrease. Finally, the repulsion created in the second step along with the increase in polymer concentration increases the minimum free energy and CMC, which itself will eventually cause aggregation [45].

2.3 Morphology of micelles

One of the important factors of nanoparticles' features that affects their efficiency is their shape. The shape of nanoparticles influences circulation half-life, biodistribution, and cellular uptake. These cases have been reviewed in more detail by Truong et al [46]. CMC, temperature, pH, the composition used and ionic strength are the factors that are effective in disrupting the balance of micelles and forming a new shape [47].

During the self-assembly phenomenon, the unimers of micelles can take different forms such as spherical, cylindrical, and vesical (flat bilayer or polymersomes). Micelles' self-assembling depends on a parameter called the critical packing parameter (CPP), which is calculated through the following formula:

$$CPP = v/a_s l_c$$

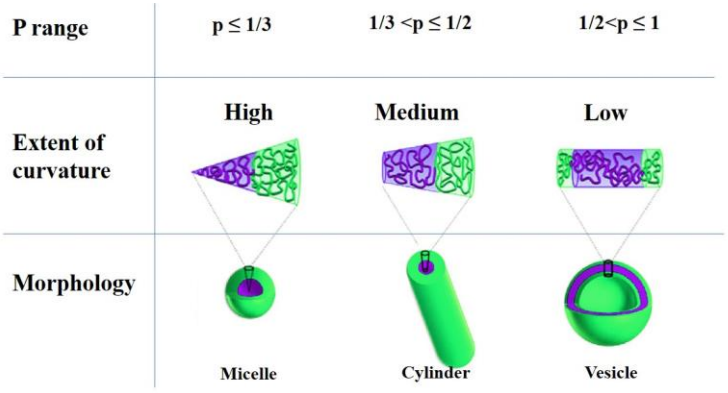

Figure 4. Self-assembly of different block copolymer as a function of critical packing parameter (p) [51].

in which v is the volume of the tail, a_s is the area per particle at the micelle head, and l_c is the basic length of the micelle's tail (Figure 4)[48,49] .

I. Spherical micelles

Spherical micelles are formed when the CPP number is smaller than 1.3 [48]. This morphology of micelles is the most abundant form of micelles. If spherical micelles are composed of two different subunits A and B (block A is soluble and block B is insoluble), they are called spherical micelles of diblock copolymer AB type. This type of spherical micelles forms star-like hairy micelles or core-shell micelles if block A is larger than block B, whereas if block B is larger than block A, crew-cut micelles are formed [50].

This type of micelles can also contain three blocks that are aggregated in ABA and ABC types. Micelles that are aggregated by ABA are known as triblock copolymers and are seen in two forms: star-like and flower-like micelles. Star-like is seen when micellization is done in the selected solution for external block A. On the other hand, if micelle formation occurs in the selected solution for block B, with the low molecular weight of block A and the low concentration of the copolymer, it forms flower-like micelles [50].

In the ABC type, the desired micelle consists of a hydrophobic part (B) and two hydrophilic parts (C and A). Like ABA, this type of model can be seen in different ways. Core-shell-corona micelles are among the forms that are aggregated in ABC type, which are constructed when a selected solvent is provided for block C or A. Block A forms an ionic core, block B causes region A to be formed, and finally block C will make the corona formation. These types of micelles are also known as three-layer or onion-like micelles. Another type of spherical micelles is Janus micelles and they are created when two solvent blocks are incompatible with each other. In this case, the two incompatible blocks form a corona (compartmentalized corona) and the last block will form the core (Figure 5) [50,51]. ABC-type micelles can be linear, branched (star-shaped and mictoarm), and finally, formed into a graft copolymer (A-graft-B/C and A-graft-B-b-C) if the two blocks B and C are hydrophobic and together are incompatible while block A is found to be hydrophilic [51,52].

II. Cylindrical micelles

Cylindrical micelles can be found in several different forms. In general, the CPP of cylindrical micelles is between 1.3 and 1.2. Among the different forms, Wormlike micelles (WLMs) are used more often, which is created when the concentration of surfactants is high and the cylindrical micelles are elongated. The CPP of WLMs is approximately 1.2 (Figure 6) [50]. WLMs are found to be 101 – 104 nm in length and 100 – 101 nm in width (38). Moreover, they can be provoked by/ or constructed from factors, including ionic surfactants (e.g., CTAB, CTAT, CTACI, CPBr), the addition of simple salts (e.g., NaCl, KCL), hydrotropes, co-surfactant (e.g., small chain alcohols), non-ionic surfactants (e.g., polyoxyethylene ethers), cationic and anionic mixtures, ionic and non-ionic surfactants, zwitterionic surfactants, Gemini surfactants, lipids and biological surfactants (e.g., lecithin), and block copolymers [53]. Due to the unique rheological behavior of WLMs (giving viscoelastic properties to aqueous solutions), this form of micelles has attracted the great attention of scientists and has been evaluated in many studies [54-57]. Thus, nowadays WLMs (smart WLMs) that react to various stimuli such as temperature, pH, CO_2, redox, solvent, light, and hydrocarbon, as well as multi–stimuli and responsive reverse micelles have been developed for applications in various industries such as oil and petroleum industry, biomedicine, self-care and home-care products, electrorheological fluids, photo-

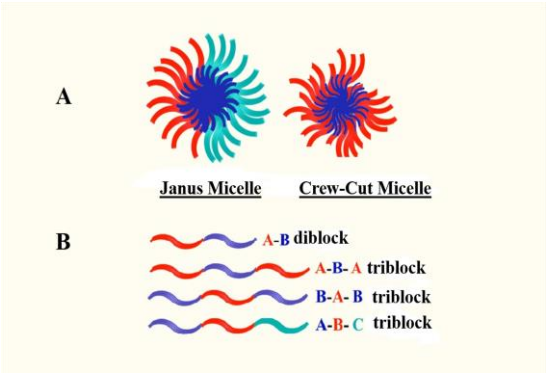

Figure 5. A) Two forms of PMs from different block copolymers B) A schematic illustration of some block copolymers [51].

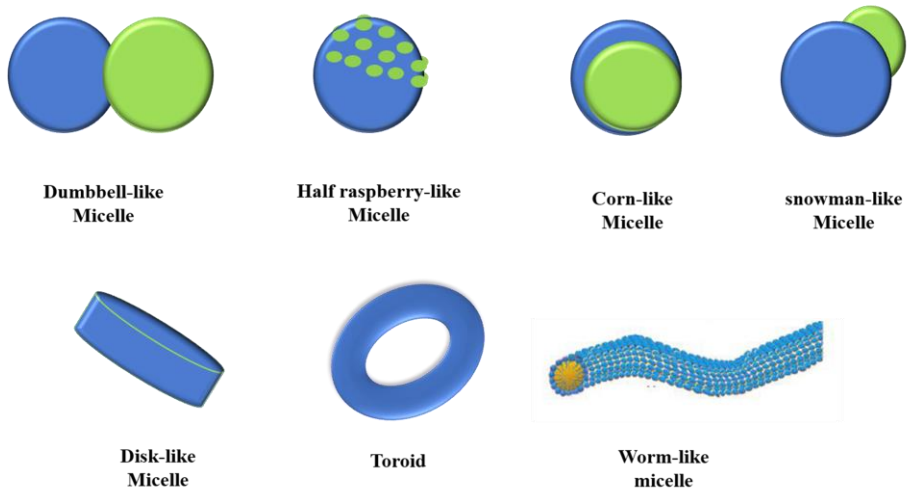

Figure 6. A schematic illustration of micelles with different morphology.

rheological fluids, drag reduction, and templating purposes [58,59].

Various forms of cylindrical micelles differ in terms of length and diameter. For instance, rod-like micelles (which are used synonymously with cylindrical micelles in different sources) are cylindrical micelles with a shorter length but thicker than other forms of cylindrical micelles [50]. In other words, rod-like micelles are cylindrical micelles where the micelle length (L) is smaller than or equal to its perseverance length (l_P). Besides, WLMs and rod-like micelles differ in their aggregation number, such that the aggregation number of WLMs is larger than the latter. Another form of this type of micelles is thread-like micelles, which have a cylindrical shape with a longer length and less thickness [50].

III. Vesicle micelles (polymersomes)

If the CPP value is between 1.2 and 1, micelles are constructed in the form of vesicles (polymersomes) [50]. Vesicles can be formed with 3 different types of block copolymers:

1. Diblock copolymers (AB copolymers)

2. Triblock copolymers (ABA, BAB & ABC copolymers)

3. Multiblock copolymers (ABCA & ABABA copolymers)

The unique feature of these micelles is their aggregation in complex forms. Various forms of complexes such as onion, tube containing vesicles, vesicles with uniform small size, entangled vesicles, empty concentric vesicles, and expansive polydisperse vesicles are formed by changing the length of blocks, the concentration of copolymers, solvent compounds, temperature, and adding ions to the system [50].

IV. Other micelles' morphologies

Disk-like micelles are formed in CPP between 1.2 and 1.3 at the rim and CPP ~1 at the surface [60]. Ribbon-like micelles with a CPP of approximately 1 are elongated forms of disk-like micelles [48]. Bicontinuous micelles and toroids (with a CPP of 0.44) were first reported and reviewed by Holder et al [61]. Moreover, corn-like, raspberry-like, dumbbell-like, and snowman-like micelles are among the structures that are less used than the previous forms (Figure 6) [62].

3. Application of micelles in industries

3.1 Medical applications

3.1.1 Drug delivery

As studies suggest, PMsprovide several great advantages to drugs, namely improving medication stability, prolonging in vivo circulation time, boosting the solubility of hydrophobic medicines, and lowering

systemic toxicity [16]. Since micelles can take different shapes during self-assembly, and in the case of proper design and choosing the right formation method, these nanocarriers can carry a variety of pharmaceutical agents such as hydrophilic and hydrophobic drugs as well as biomolecules such as siRNA, mRNA, proteins, pDNA and small molecules like photosensitizers [63,64].

In various studies, micelles have been also used as nanocarriers to carry anticancer drugs such as Paclitaxel, Ursolic acid, Doxorubicin, and Docetaxel [65-67]. In addition to cancer, the efficacy of drugs and agents encapsulated in micelles have been evaluated for the treatment of various diseases such as diabetes [68], tuberculosis [69], asthma [70], eczema [71], CNS diseases, and neurological disorders [72,73], malaria [74], mitochondrial disorders [75], dermal diseases [76], respiratory system diseases [77].

Maintenance of the medicine in the nanocarriers is one of the major difficulties in the clinical translation of PMs(PMs) [78]. The efficacy of PMs in drug delivery is largely dependent on their integrity, which includes their capacity to withstand drug dropping and immature disassembly. When PMs are administered orally, various factors such as medium volume, bile salts, secreted enzymes, pH, and ionic strength can affect PM integrity until the nanocarrier reaches the intestine and colon for absorption. To deal with this problem, it is very important to choose the right composition for micelles formation. CMC is another important factor to deal with drug loss. In general, the lower the CMC, the higher the resistance of micelles to disassembly. Camptothecin packed in Pluronic® - Poly(acrylic acid) PM system nanocarrier, Efavirenz in F127:T904 mixed micelles and Meso-tetraphenyl porphine (mTPP) in F127- b-poly(ethylene glycol)-b-distearoyl phosphatidylethanolamine are among the drugs that are considered promising oral administration platforms [79]. PMs can also be administered parenterally. In various studies, drugs packaged in injectable PMs have been investigated. Laquinimod with D-α-tocopherol PEG 1000 succinate (TPGS) as a micellar nanocarrier based on polyethylene glycol, which is used to treat liver fibrosis whose administration by injection has been investigated. In addition, nanocarriers can be delivered through topical (thymoquinone encapsulated in soya lecithin & chitosan-based micellar nanocarrier), Transdermal (curcumin encapsulated in hyaluronan-based nanocarrier), Periodontal (baicalin encapsulated in TPGS based PM) and Intranasal (rotigotine encapsulated in Polyethylene Glycol (PEG)-Poly (Lactic-Co-Glycolic Acid) (PLGA)-based Nanoparticles) [78]. Micellar nanoparticles are also used to treat ocular diseases. Generally, to treat these kinds of diseases, drugs are either systemically entered into the eye or directly entered into the eye in three ways: topical, intraocular injection, and periocular injection. Depending on the pathway of drugs to reach the target in the eye (corneal and non-corneal pathways), there are four different barriers (tear-film barrier and the corneal barriers for the corneal pathway, blood-aqueous and blood-retinal barriers for the non-corneal pathway) to prevent foreign substances from entering the eye, and nanoparticles are used to cross these barriers. In vivo studies on the delivery of drugs through the eyes of rabbits showed that the application of micellar nanoparticles is a promising method for the delivery of drugs for eyes. In all these studies, the topical method was used for ocular drug delivery; also, the efficiency of various micellar copolymers including polyvinyl caprolactam-polyvinyl acetate-polyethylene glycol, 1, 2-Distearoyl-sn-glycero-3-phosphoethanolamine-Poly(ethylene glycol)/solutol HS, P123/ d-α-tocopheryl polyethylene glycol in delivering Curcumin, Genistein, and Resveratrol into the eye site was evaluated [80].

- **Smart micellar nanoparticles**

The development of stimuli-responsive micelles (so-called smart micelles) has led to the creation of systems with tunable or switchable catalytic activity and solubility. There are two signal models for drug release from nanocarriers. These signals include exogenous signals, which have signals emitted outside the body, and endogenous signals, which are known as signals originating from within the body. The arrival of these signals to the smart nanocarriers will release the drug from the nanocarrier [81,82].

I. Endogenous signals responsive smart micelles:

❖ **pH-responsive smart micelles:** The drug release trigger from this type of micelles is the pH

difference between normal cells and tumors. With the increase in glycolysis in tumors and the rising production of lactic acid, the pH around cancer cells is around 6.5 [82,83]. This pH difference between normal cells (7.4) and cancerous cells can affect the conformation of the nanocarrier and/or its behavior. On the other hand, this difference can cause the components of the nanocarrier to dissolve and collapse, resulting in the release of the drug at the tumor site [82].

- **Redox-responsive smart micelles:** The reduction of substances in the body such as vitamin E, vitamin C, and glutathione, and their effects on the bonds in micelles' construction is a method that can be used to release the drug from nanocarriers [82]. Research showed that the amount of glutathione around tumors is much higher than the glutathione found inside and outside normal cells. Bonds such as disulfide, tetrasulfide, diselenide bonds, platinum conjugation, and thioether succinimide linkages are used in smart nanocarriers because glutathione can reduce them and as a result, the drug is released at the tumor site [84].

- **Enzyme-responsive smart micelles:** Some polymeric nanocarriers are designed to respond to enzymes [82]. These systems are kind of engineered nanocarriers in which hydrolases (including proteases, phospholipases, and glycosidases) and oxidoreductases can cause collapse and release the drug at the tumor site [85].

II. Exogenous signals responsive smart micelles:

- **Temperature-responsive smart micelles:** PMs are among the substances that respond to temperature. Therefore, a method based on cooling and heating called hyperthermia has been designed to affect PMs [83,86]. In this method, the body temperature is increased to 41-43 degrees so that the body temperature reaches above the polymer's critical solution temperature. In this case, as a result of losing the balance between the hydrophilic and hydrophobic interaction forces of the polymer, the nanocarrier collapses and the drug is released [82,83].

- **Light-responsive smart micelles/photo-responsive smart micelles:** If the appropriate wavelength is chosen, light causes the drug to be released from the nanocarrier [82,87]. This type of drug release is very reassuring due to the adjustable location and time of light irradiation. Moreover, visible light, UV light, and NIR light can be used as light stimuli [83]. This type of drug release consists of five different mechanisms including Photo-isomerization, Photo-cleavage, Surface plasmon resonance (SPR) absorption, Hydrophobicity change, De-crosslinking [88].

- **Magnetic-responsive smart micelles:** A magnetic part is used in the composition of these smart micelles. When the nanocarrier reaches the desired location, a magnetic field can make the nanocarrier collapse and release the drug [82,89].

- **Ultrasound-responsive smart micelles:** High-pressure waves that have a wavelength of greater than 20 kHz are known as ultrasound [90]. These waves cause the release of the drug by introducing thermal and mechanical effects on the drug-containing PMs [82].

Among various micellar systems investigated in the literature, there are some types that were in clinical trials such as paclitaxel PM nanoparticle (Cynviloq IG-001 (Sorrento)) against breast cancer; paclitaxel PM nanoparticle (Genexol-PM (Samyang Biopharmaceuticals) against head and neck or breast cancer; polyamino acid, PEG, and cisplatin derivative micellar nanoparticle (NC-6004 Nanoplatin (Nanocarrier)) against advanced solid tumors, lung, biliary, bladder, or pancreatic cancers; polyamino acid, PEG, and oxaliplatin micellar nanoparticle (NC-4016 DACH-Platin micelle (Nanocarrier)) against advanced solid tumors or lymphomas; paclitaxel micelle NK105 (Nippon Kayaku) against Breast cancer; Docetaxel micelle (Docetaxel-PM DOPNP201 (Samyang Biopharmaceuticals)) against head and neck cancer and advanced solid tumors; Docetaxel micelles (CriPec (Cristal Therapeutics)) against solid tumors, ovarian cancer; PEG, iron, and amifostine micelle (RadProtect (Original BioMedicals)) for dose escalation and safety for acute radiation syndrome; micelle encapsulated epirubicin (NC-6300 (NanoCarrier)) for advanced solid tumors or soft tissue

sarcoma; micelle formulation of SN-38 (IT-141 (Intezyne Technologies)) against advanced cancer; micelle covalently conjugated to indocyanine green (ONM-100 (OncoNano Medicine) for Intraoperative detection of cancer [91].

3.1.2 Biological cell monitoring

As also mentioned before micelles have a variety of useful applications, but one of the most essential ones includes the process of monitoring biological cells. During the last decade, several researchers' attention has been brought to this topic. Mouffouk et al. developed a simple and sensitive bioassay method for the detection of Escherichia coli using self-assembled pH-responsive PMsbioconjugated to anti-(E. coli). Poly(ethylene glycol— trimethylsilyl methacrylate) containing silicon moieties was used to remotely activate the dye release via pH [92]. Since anthrax spores are dangerous to humans and animals and are potential biological warfare agents, rapid, sensitive, and selective quantitative detection of pyridine dicarboxylic acid (DPA) is needed as a biomarker. Luan et al. designed Tb3+ ions in lanthanide-functionalized micelles that may be sensitized to generate intrinsic luminescence when DPA chromophore coordinates with them owing to energy transfer. The terbium functionalized micelle sensitively detected DPA with a linear relation in the range of 0 μM to 7.0 μM in an aqueous solution and exhibited outstanding selectivity to DPA over other aromatic ligands [93]. Uthaman et al. studied Probe indocyanine green hydrophobically modified and encapsulated into CD 44-specific HA micelles for Near-Infrared Fluorescence imaging. Photoacoustic imaging showed tumor-selective micelle accumulation in vivo. These findings were proposed using HA micelles in theranostics [94]. In a study by Cho and colleagues, PEG-b-PCL micelle was loaded with 1,1-dioctadecyl-3,3,3,3-tetramethylindotri-carbocyanine iodide (DiR) via nano-precipitation to achieve optical imaging in mice. Mice optical imaging of DiR showed considerable tumor tissue delineation. Thus, optical imaging using DiR-loaded PEG-b-PCL micelles in the animal model may guide solid tumor removal surgery intraoperatively [95]. Shi et al. created two fluorophores, Dy-488 and Dy-676, core-linked cystamine-based, reduction-sensitive PMs (CCPM). Micelles accumulated well in CT26 tumors in mice utilizing optical imaging [96]. Xiao et al. self-assembled a pH-responsive micelle from biodegradable poly(amino acid) block copolymer mPEG-PAsp(DIP)-co-PLLeu to load both chemotherapeutic paclitaxel (PTX) and MRI contrast agent superparamagnetic iron oxide nanoparticles (SPIONs). Hydrodynamic particle size was 134.5 ± 11.2 nm for the SPIONs/PTX-coloaded PDPL micelle. The theranostic SPIONs/PTX-PDPL delivered PTX and SPION to Bel-7402 cancer cells and promptly released PTX in acidic lysosomes to trigger cell death. After co-incubation with SPIONs/PTX-PDPL micelle, in vitro MR imaging identified liver cancer cells sensitively. The SPIONs/PTX-PDPL micelle may be a theranostic nanosystem for simultaneous treatment and MR imaging of hepatocellular cancer [97]. Yi et al. created a self-guiding PM (TB@PMPT) with two AIE photosensitizers and a reduction-sensitive paclitaxel prodrug (PTX-SS-N3) for improved chemo-photodynamic treatment using dual-stage light irradiation [98]. Piñol et al. offered a novel technique for intracellular temperature mapping under a fluorescence microscope (uncertainty of 0.2 K) utilizing rationally designed luminous Ln3+-bearing polymeric micellar probes incubated in breast cancer MDA-MB468 cells, showing the organelles' thermogenic activity and the tool's ability to research intracellular processes [99]. In a research work of Sun and colleagues, using confocal laser scanning microscopy based on aggregation-induced emission, mPEGss-Tripp was produced and self-assembled into redox-sensitive PMswith a diameter of 105 nm to create a smart drug delivery system for simultaneous imaging and cancer treatment. A new redox-sensitive drug delivery system with AIE property was created for simultaneous cellular imaging and intelligent drug administration and release [100].

3.1.3 Detection of pharmaceuticals

Micellar-mediated extraction might eliminate the usage of poisonous, combustible, and generally hazardous organic solvents. It can purify plant-derived chemicals and other substances. In low concentrations, extraction solutions of surface-active chemicals are safe, non-toxic, non-flammable, and waste-friendly. This method's extracts may be used in the food, cosmetic, and pharmaceutical sectors, thus their effects on living organisms must be assessed. To extract

physiologically active plant components like polyphenols, such extraction methods have been used numerous times [101,102]. Zhang and colleagues used mixed micellar liquid chromatography and isolated nine isoflavones present in traditional Chinese medicine in 30 min. This research adjusted chromatographic settings such as pH, surfactant composition and concentration, organic solvent type and ratio, and column temperature. This strategy saves time, money, and effort [103]. Zhao et al. developed a sensitive flow injection chemiluminescence (CL) technique for lornoxicam testing using micelle synergism. The interaction of Ce(IV) with lornoxicam in an acidic solution produced a modest CL signal, although SDBS increased CL strength. The approach effectively assayed lornoxicam in pharmaceuticals with good recovery [104]. Liu et al. suggested a capillary electrophoresis separation technique using mixed micelles and electrochemiluminescence detection to simultaneously determine procaine, lidocaine, ropivacaine, and bupivacaine. SDS-Tween 20 micelles considerably increased separation [105]. Al-arfaj and El-Tohamy devised a simple and sensitive spectrofluorimetric approach to determine ribavirin in pharmaceutical formulations using its fluorescence spectrum characteristics in an SDS micellar system. Ribavirin fluorescence was observed at 396 nm following excitation at 270 nm [106]. Salim et al. devised a sensitive spectrofluorimetric approach for sitagliptin measurement in pharmaceutical formulations. The approach was based on an SDS micellar system study of sitagliptin's fluorescence spectrum characteristics [107]. Ibrahim et al. investigated the separation performance of six active pharmaceutical components without organic solvents using sodium dodecyl sulfate and polyoxyethylene-23-lauryl ether (Brij-35). The projected optimal conditions were confirmed using a mobile phase of 93.6 mM SDS, 32.0 mM Brij-35, and 10.0 mM sodium dihydrogen phosphate, adjusted at pH 5.2, column temperature 35° C, and detection at 215 nm wavelength. The approach effectively determined 12 alternative combination formulas of the analytes in their pharmaceutical products and was appraised for greenness on two unique criteria [108].

3.1.4 Detection of biomolecules and their activity

Both basic research and clinical diagnostics place a significant emphasis on the detection of biomolecules, as well as the quantitative assessment of their quantities and activities. He et al. studied DNA's conformation modification when it interacts with cationic Gemini surfactant 12-4-12. A hydrophobic pyrene probe was utilized to study microenvironment changes, and Gemini surfactant 12-4-12's CMC was estimated [109]. The deprotonation kinetics of the N-heterocyclic DNA intercalator acridine in SDS micelles were monitored by Sarangi et al. to study DNA's dynamic protonation [110]. Bandyopadhyay et al. examined model membrane contact and DNA binding of bee venom-derived lasioglossin II. The N-terminal tryptophan residue of lasioglossin II interacted with the hydrophobic core of a dodecyl phosphocholine micelle in fluorescence tests, and agarose gel retardation showed its DNA-binding capacity [111]. Yan and colleagues invented Micelle-Tagging Electrophoresis (MTE), a gel-free DNA separation method. By transiently attaching non-ionic micelles to DNA as drag-tags by end-alkylation, MTE separates DNA in free solution quickly and accurately [112]. Vafaei and colleagues produced micellar nanostructures that self-assembled into a Förster resonance energy transfer (FRET) nano platform for DNA detection. FRET pairs' fluorescence responses are produced by lipid oligonucleotide FRET probes in micellar scaffolds fusing and fissioning DNA-mixed micelles. The micellar multiplex FRET technology boosted detection limit and FRET efficiency by 20- and 3-fold, respectively, compared to traditional FRET substrates with a single donor and acceptor. FRET-based diagnostic testing of nucleic acid and non-DNA targets may benefit from this supramolecular signal amplification approach [113]. According to Abel et al., Four peptides (mTM10, mTM16, TM17, and KTM17) and micelles of dodecylphosphocholine and dodecyl-β-D-maltoside were simulated using molecular dynamics. These peptides are three transmembrane segments of membrane-spanning domains of hMRP1 [114]. Vicente et al. used fluorescence to study the interactions between a membrane mimic and a cationic micelle. All peptides interacted with the micelles. Many antimicrobial peptides interact with and damage the bacterial cell membrane, which increases their effectiveness and selectivity. Antimicrobial peptide membrane disruption in anionic micelle SDS has been studied using fluorescence measurements [115,116].

Amiri et al. investigated how Gemini surfactants interact with two model proteins, ribonuclease A and hen egg white lysozyme. Fluorescence spectroscopy indicated that these cationic Gemini surfactants do not interact substantially with or destabilize these well-folded proteins under physiological settings, making them excellent membrane mimetics for researching membrane component-positively charged protein interactions [117]. Chandrababu et al. studied amelogenin's structural adaptability to SDS. Fluorescence spectroscopy was used to investigate the physiological implications of amelogenin's flexibility and sequence-specific helical tendency, which may allow it to structurally adapt to charged targets such as cell surfaces, minerals, and proteins during enamel biomineralization [118]. Pinto et al. investigated β-galactosidase activity in mixed micelles of SDS and an imidazolium ionic liquid. The research assessed mixed micelles as reaction media for glycol-oligosaccharide commercial synthesis [119]. A fluorescence approach employing pyrene as a probe was utilized to explore the mechanism of distinct stimulatory effects of the biosurfactant rhamnolipid and the commercial surfactant Tween 80 on lignocellulose enzymatic hydrolysis. The findings showed that surfactants boosted enzyme stability and activity, improving enzymatic hydrolysis efficiency [120]. Mixed micelles and fluorescence detection were used by Ross et al. to identify amino acids in the human colipase enzyme [121]. Methylmercury caused severe neurotoxicity, nephrotoxicity, and pulmonary toxicity. Oh and colleagues created the fluorescent probe in SDS micelles which enhanced red fluorescence at 575 nm to detect Hg^{2+} ions and methylmercury among 16 metal ions in an aqueous solution in 1 min. Methylmercury was detected exceedingly sensitively and selectively in SDS micelles with EDTA [122]. Tanaka et al. used an asymmetric poly(styrene-b-acrylic acid-b-ethylene glycol) triblock copolymer to soft-template mesoporous iron oxide. In aqueous liquids, this polymer produces a soft-template micelle with a PS core, PAA shell, and PEG corona. The combined detection platform for H_2O_2 and glucose will be used to produce biosensors for personalized medicine, food safety, environmental pollution control, and agro-biotechnology [123]. Wang et al. created a photosensitizer Ce6-loaded fluorinated PM (Ce6-PFOC-PEI-M) by self-assembling an amphiphilic polymer made from perfluorooctanoic acid and branching polyethyleneimine (10 kDa). Perfluoroalkyl groups in the PM Ce6-PFOC-PEI-M preserved the oxygen-carrying ability of perfluorocarbon, raised oxygen levels, and overcame hypoxia in C6 glioma cells under oxygen-deficient conditions [124]. Excessive Al^{3+} in the body may harm the central nervous system and cause Alzheimer's, Parkinson's, and amyotrophic lateral sclerosis. Wang et al. encapsulated hydrophobic salicylidene schiff bases probe (Dye) in DSPE-PEG nano micelles to detect Al^{3+} in aqueous systems and image Al^{3+} in live cells [125]. Zhuang et al. created a multifunctional PM system based on pH and redox dual responsive mPEG-P (TPE-co-AEMA) copolymer for stimuli-triggered drug release and aggregation-induced emission (AIE) active imaging. These mPEG-P (TPE-co-AEMA) micelles have good biocompatibility and emission properties, making them suitable for cellular imaging [126]. Tyrosinase (TYR), a copper-containing enzyme, biomarks melanoma cancer cells and is essential for melanin generation in melanocytes and malignant melanoma cells. Wang et al. created a FRET-based self-assembling ratiometric fluorescent micelle nanoprobe (NanoDPA-NMP-tyr) to react to B16 cell TYR activity. NanoDPA-NMP-tyr responds well to TYR in B16 cells using fluorescence imaging [127].

Due to its biological importance, zinc ion (Zn^{2+}) is an important cationic element in living systems. Many enzymes involved in cellular metabolism, gene expression, cell death, metalloenzyme control, and brain signal transmission use Zn^{2+} as a catalytic center and structural cofactor. Pyrophosphate ($P_2O_7^{4-}$, PPi) is an essential biological anion because of its functions in ATP hydrolysis, DNA polymerization, and other metabolic activities. A micellar probe for Zn2+ and pyrophosphate (PPi) ions was reported by Chang et al. In the presence of sodium dodecyl sulfate, a micelle was generated using tetraphenylethene (TPE) as an AIEgen and naphthalimide-dipicolylamine (DPA) scaffold as a Zn^{2+}-chelating fluorescent reporter. Aggregation-induced emission (AIE) of TPE moiety caused the micellar probe to fluoresce blue at 482 nm [128].

Halder et al. created FeFlu, an anionic micelle (SDS)−hematoporphyrin (Hp) complex sensitized by Fe(III) chloride to detect fluoride in water. The

micelle−hematoporphyrin (SDS−Hp) combination is basically nonfluorescent, but the prototype (FeFlu device) based on it exhibits high turn-on fluorescence upon F− identification in water [129].

3.2 Cosmetic industry application

According to the Food and Drug Administration (FDA), cosmetics are attributed to particles intended to be applied onto human bodies or any part thereof for cleansing, beautifying, promoting attractiveness, or altering appearance [130]. Based on this definition, a list of cosmetic segments has been characterized, including skin moisturizer, anti-aging, facial makeup, shampoo, toothpaste, deodorant, hair color, and other products used for appearance enhancement [131]. The cosmetics market is one of the largest global markets which is projected to reach $463.5 billion by 2027 [132]. Various large beauty companies such as Bioderma, L'Oréal, Avenue, Laroche-posay, and Garnier are applying micellar nanoparticles technology in their skin cleanser product segment, and due to its successful performance, micellar nanotechnology is very likely to be used in wider cosmetic products segment. For instance, L'Oreal, a leading cosmetics manufacturer, has patented its micellar-based cosmetic formulations through nanoemulsion systems showing various beneficial applications on the skin, mucous membranes, hair, scalp, and eyes. Furthermore, Naturel Kiss, a Malaysian cosmetic brand, has launched "Micellar Series" products consisting of micellar facial cleanser and mist containing various herbal ingredients incorporated with micellar nanoparticles in nanoemulsion systems [133,134]. Skin cleansers are mostly oils that absorb dirty oil and sebum from the skin, removing dirt and debris from the skin and allowing unwanted oil and dirt to be washed away. However, the shiny and grassy appearance of an oil-cleansed face is unfavorable. On the other hand, cleaning the skin with water and soaps, and or other detergents cause dryness of the skin since old pipes and water supply systems increase the water's hardness. Thanks to micellar waters, and dispersed tiny particles of oil in an aqueous solution without a traditional emulsion, these issues are effectively solved. They have also known as oil-in-water (O/W) emulsion systems and are largely investigated as the most suitable system to be implemented in cosmetics formulation.

The surfactants in a micellar solution allow for normally insoluble compounds to be suspended in a liquid [135]. To improve the solubility of poorly soluble compounds, surfactants are commonly used in cosmetics, but due to safety issues, only their minimal amounts are allowed to be added. PMs with relatively low CMC [136,137] in cosmetic formulations reduces the required amounts of surfactant. CMC is the minimum concentration of polymers in solution leading to the micelles' self-assembling. In an aqueous solution, amphiphilic molecules act individually as surfactants, but by the increase of copolymers' concentration, copolymers begin to aggregate and reach CMC due to the saturation of the bulk solution. Therefore, at concentrations above CMC, micelles are highly stable, however, if the system experience dilution to values under CMC they are going to be disassembled [47,138]. The stability of PMs is controlled by chemical principles of thermodynamics and kinetics. The thermodynamic stability of micelles is measured by the CMC of polymers [139]. Polymers with lower CMC values form more thermodynamically stable micelles that can be maintained even when diluted [140]. The kinetic stability and strong thermodynamic stability of tiny PMs provide great stability for nanoemulsion-based cosmetics against sedimentation, flocculation, and Ostwald ripening. Moreover, the micellar formulation provides an efficient feature for nanoemulsion-based cosmetics i.e., creating a transparent solution, which is rooted in processes of small micelles distribution throughout the aqueous solution. Their size is even smaller than the light wavelength, showing no light scattering which results in the development of a transparent solution [141].

PMs are also able to incorporate and thus solubilize a variety of lipophilic compounds used in cosmetics with different physicochemical characteristics and to efficiently deliver lipophilic constituents while protecting them from decomposition caused by biological reactions within the human body [142].

In addition, penetration of active compounds into the skin is of great importance in the high efficacy of cosmetics, and their incorporation with PMs is a promising way to enhance this matter. Although the

effectiveness of PMs' penetration depends on various factors such as their particle size, lipophilicity, and the degree of ionization, they show more desirable permeability compared to other nanocarriers such as liposomes [134,143]. Besides, the trans epidermal delivery of PMs can also be enhanced by using cell-penetrating peptides. The stratum corneum, which is the topmost layer of the epidermis and the target of transdermal drug delivery, functions as a skin barrier by forming a layered structure between the keratinocyte lipids and the keratin proteins [144]. So, it interferes with the penetration of the external substance and makes the percutaneous permeation of the active substance difficult. It has been established that the peptides with the ability of cell permeation can easily access the cell membrane as a result of their arginine-rich sequence with high positive charges [145]. Since both cell membrane and epidermal keratinocyte lipids have lipid bilayer structure, if the arginine oligomer, a key amino acid sequence of a cell-penetrating peptide, is applied to a cosmetic product along with an active substance, it could boost the skin absorption of the active substance and thus maximize its efficiency [146].

In addition to improving solubility, stability, and permeability, PMs also provide a high loading capacity for hydrophobic active ingredients despite their small particle size [147]. The drug loading capacity of PMs is critically dependent on the compatibility between the drug and the block copolymer in the micelle core. By optimizing the chemical structures of the inner core segment for stronger drug/carrier interaction, the loading capacity can be improved [148].

Studies conducted through these years on the application of PM in cosmetic products are mainly related to anti-aging compounds. In a study by An et al., PMs of Capryol 90 and poloxamer were developed to encapsulate oleanolic acid, a natural antiaging ingredient for cosmetic products alleviating skin wrinkles. PMs of oleanolic acid developed in this study not only showed improvement in oleanolic acid solubility with a minimum amount of surfactants but also enhanced the permeation of oleanolic acid through the skin. This formulation also showed excellent stability with no precipitation, phase separation, or degradation at 40 °C after 3 months. Figure 7 indicates the clinical trial outcomes of the formulation with fine efficacy in alleviating human wrinkles [143].

In another study, a fabricated micelle using hyaluronan (HA) was evaluated to deliver the different active ingredients into the stratum corneum and deeper layers in different model systems. Since both PMsand HA (in native form) are mostly considered skin penetration enhancers, HA-based micelles are expected to increase skin penetration of hydrophobic drugs. Smejkalová et al. showed that hydrophobized HA-based PMs could improve the transport of hydrophobic compounds across the stratum corneum barrier into deeper skin layers via the transcellular route, thereby increasing their cutaneous availability. Although it is believed that any molecule larger than 500 g/mol is generally impermeable through the skin, the HA delivery system developed by Smejkalová et al. could enhance the permeation of large hydrophobic molecules (MW 318

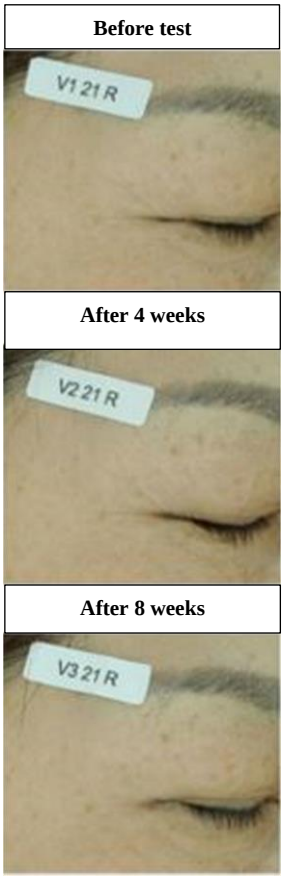

Figure 7. The effects of PMs of oleanolic acid in reducing human wrinkles [143].

and 863 g/mol). They also assessed the delivery of coenzyme Q10, a common cosmetic ingredient in anti-aging, by sodium oleyl hyaluronan (HAC18:1) PMs. To this end, they incorporated Q10-loaded PMs in o/w cream formulations. The stable formulation was applied once daily for one month by human volunteers. The results showed a clear skin hydration effect, whereas this was not the case for either free coenzyme Q10 or HA vehicle alone. Hydrated skin is less susceptible to wrinkles, which is a key aspect of today's cosmetic approaches [149].

Micelles, in addition to nano-emulsification of hydrophobic active ingredients, are also applied in the analytical monitoring of compounds making up cosmetics. Since cosmetic products are in direct contact with the skin, the presence of harmful substances in their structures can cause skin diseases, irritation, and allergic reactions, as well as absorption and storage in different organs, which will show toxicity in the short or long time. Hence, governmental entities have listed prohibited substances for use in cosmetics. One of the analytical separation techniques applied in this respect is called micellar electrokinetic capillary chromatography which has attracted considerable interest due to its efficiency, speed, reproducibility, very small volume of sample, and ease of contaminants cleaning up. This technique is mostly applied to analyze sunscreen ingredients in cosmetics [150-152].

Another analytical methodology based on micelles particles was developed by Wang et al., which is an FI-fluorimetric analysis (FIA) for the continuous and sequential determination of rhodamine B (RhB), a carcinogenic dye, in cosmetic products. The FIA spectrofluorimetric method for RhB determination in real samples of lipsticks has multiple advantages, including simplicity, precision, speed, low detection limit, and the need for cheap equipment. They added different surfactant solutions during the flow-injection analysis and studied RhB fluorescence signal changes, showing that the anionic surfactant of SDS (sodium lauryl sulfate) increase the sensitivity of the fluorescence from RhB until 2.5-fold. This method can be used to determine RhB present in other cosmetics products such as eye shadows, rouge, and other types of samples [153].

3.3 Detergent industry application

Home care products include hard surface cleaning (i.e., floors, tiles, toilet bowls, kitchen sinks, and the like), drain openings, and paints. In these products, two conspicuous features of the amphiphiles, namely elasticity and viscosity, are exploited to produce viscoelastic surfactants, the major components of detergents and personal care formulations [154,155]. Sink cleaners, for example, should have a high surfactant concentration in order to solubilize grease and resuspend particulate material. WLMs give a creamy feel, but they can suspend the abrasive particles only when the system is gelled by the addition of soluble polymers. The polymers also enhance the feel and texture of the product [156].

Another application for WLMs may be found in paints. For storage stability, paint at rest should behave like a solid (gel) to prevent settling (i.e., formation of hard sediments at the bottom of the container) of its pigment and binder particles [157].

The principles underlying personal care formulation based on WLMs are similar to those applied in home care products. By personal care products, we mean shampoos, shower gels, bath additives, and other grooming aids and products. For example, concerning hair bleaching and oxidative dyeing, the major thickening feature from the formation of WLMs (leading to an isotropic and viscous gel) or lamellar phases (leading to an anisotropic and viscous cream or gel) for hair application. For both cases, the thickening is suppressed in the original containers by adding more than 20 wt% of solvents to the lotion formulation. The lotion is viscose when it is mixed by the consumer with a predominantly aqueous developer container to trigger the thickening in situ immediately, before hair application. However, it has been observed that hair bleaches or colorants based on WLMs are more skin-irritating than those based on lamellar phases. This difference in behavior may arise from the higher viscosity of lamellar phases. Therefore, this drawback could, in principle, be eliminated by tuning the viscosity of the WLMs [158].

3.4 Agricultural application

Finding appropriate alternatives for synthetic pesticides with the main goal of decreasing severe chemical side effects is considered a global mission in agriculture science and technology. Furthermore,

scientists have been investigating novel strategies as delivery systems for various pesticides which will result in a controlled release and lowering residue challenges. Literature showed several types of research have been done based on the encapsulation of agrochemicals using polymeric and biomaterials. In this regard, chitosan-based nanosystems have been used as a useful vehicle for the delivery of pesticides for crop protection.

N-(octadecanol-1-glycidyl ether)-O-sulfate chitosan (NOSCS) as an amphiphilic compound was applied for carrying insecticide rotenone which is known as a water-insoluble botanical insecticide. It is constructed of octadecanol glycidyl ether as its hydrophobic agent, and sulfate as hydrophilic agent. PMsof NOSCS had a size of 167.7 - 214.0 nm and were formed self-assembly in an aqueous solution. In this system, rotenone was entrapped in the core of NOSCS micelles through the reverse micelle method. This technique helped release rotenone as its concentration was up to 26.0 mg/ml comparably higher than that in water (0.002 mg/ml) [159,160].

The synthesized carrier of the botanical pesticide azadirachtin (Aza) was an amphiphilic carboxymethyl chitosan with ricinoleic acid (R-CM-chitosan). The micelles were formed as nanospheres and they were able to protect Aza properly as the highest loading efficacy of the Aza/R-CM-chitosan was 56% when the ratio of R-CM-chitosan and Aza was 18. Therefore, a non-toxic micellar carrier was developed to be a potential green and safe botanical pesticide water dispersion formulation [161].

Najma Akhtar Shakila et al., developed micellar copolymers of polyethylene glycol and different dimethyl esters for encapsulation of carbofuran, [2,3-dihydro-2,2-dimethylbenzofuran-7-yl methylcarbamate] which is known as an insecticide-nematicide. These systems aimed to have a better controlled release. Their results indicated that it is important how the matrix of the polymer formed as it affects the carbofuran's applications for insect control [162].

There are some other types of micelle-based carrier systems for pesticides such as thiram [163], thiamethoxam [164], imidacloprid [165] , and carbendazim [166].

Among various research proposed for utilization of biopesticides as antifeedant against Crocidolomia pavonana, Melanie Melanie and coworkers prepared Lantana camara ethyl acetate fraction (EAF) micellar suspension. The highest larvae mortality (86.67%) and LC50 0.39% at 48 h were achieved by EAF (with surfactant organic-phase ratio (SOR) 11) [167].

3.5 Environmental application

Nowadays different types of chemical products are producing wastes leading to air, soil, water, and general environmental pollution which also threats human beings' and animals' lives. perfluoroalkyl substances (PFASs) are a group of synthetic surfactants that caused serious environmental problems. Particularly it made critical issues for human health. Recently much research has been done to decrease the side effects of PFASs mainly for their resistance. Zhanghao Chen et al. developed a composite of self-assembled micelle including indole acetic (IAA), cetyltrimethylammonium bromide, and PFAS. They used perfluorooctanoic acid (PFOA) which is known as one of the most widespread types of PFASs, as the contaminant and the experiment showed after 2.5 h UV irradiation, the concentration of PFOA decreased from 10 mg L^{-1} to 60 mg L^{-1}. In fact, they were able to introduce a novel method for mass transfer regulation related to the micelle which resulted in better degradation of PFASs [168].

Heavy metals are problematic issues for the environment as they make contaminations in soil and water and their removal is a difficult task. There have been many efforts for soil and water decontamination using various methodologies but those conventional methods did not have high efficacy and therefore the remained effluents caused serious challenges. Herein the application of micellar solutions of synthetic surfactants can be an intelligent approach. Utilization of biosurfactants that are biocompatible, environmentally friendly, and tolerant to a wide range of pH, temperature, and salt concentration was proposed. They were applied as decontamination agents for various pollutants. For example, in dye removal processes of the wastewater, through an adsorption method lipopeptide biosurfactant based on corn steep liquor (CST) was used in the formulation of a lignocellulosic. The experimental results indicated a

significant decrease in dye compound and sulfates to 10% and 62% respectively [169].

3.6 Food industry application

Micellar systems are also applied for delivery and/or pre-formulation in the food industry to increase their functional properties. Solubilization of curcumin using casein micelles and improving cellular uptake of carotenoids by phospholipid micellar carriers are examples of the wide range of micelles' applications [170].

The ingredients that are insoluble in water can be functionalized using micelles which have both hydrophilic and hydrophobic properties because of their unique structures. They are named microemulsions which normally consist of oil, water, and an amphiphile or surfactant [171] and have wide applications in the food industry. One common application of these microemulsions is providing glycerides for food additives for instance [172] . In addition, microemulsions comprising non-ionic surfactants, polyols, limonene, and ethanol also were used for the solubization of lutein and lutein esters which can be useful in the beverage industry as they were optically transparent [173]. Other types of microemulsions were used to solubilize some types of hydrophobic phytochemicals such as resveratrol [174], phytosterols [175], and α-linoleic acid [176].

In meat processing, Yusop et al. figured that the physical and sensory features of nanoparticle paprika oleoresin (in chicken breast fillets) were enhanced with a carrier system based on micelles [177].

Micelle-based carriers especially biopolymer micelles used for the encapsulation of useful nutrients such as vitamins, minerals, essential oils, etc. For example, vitamins C and E and special fatty acids were encapsulated by micellar systems commercially by Aquanova company. Their innovative micellar-based products showed higher bioavailability of their active ingredients [178]. They claimed their micellar carriers are potential systems for delivering some groups of ingredients to be used in the food and beverages industries. In fact, those micelles were able to deliver vitamins A, C, E, and Q_{10} to food and beverages without changing substances [179]. Regarding self-emulsifying micellar systems in the food industry, NutraLease Ltd. proposed a micelle-based system for the solubilization of mixed lipophilic flavors/food ingredients like omega-3 fish oil, vitamins, and others [171].

The reverse micelle methodology which is based on a liquid-liquid extraction method for biomolecules separations and purifications [180] also been applied for various food industry applications such as protein extraction from peanut cake using bis(2-ethylhexyl) sodium sulfosuccinate (AOT) [181], improved extraction of walnut protein (WP) using sodium bis(2-ethylhexyl) sulfosuccinate (AOT) with the help of microwave backward extraction (MABE) [182], producing chitosan nanoparticles with high efficacy [183], magnetic lipase-immobilized nanoparticles fabrication with valuable enzymatic activity [184], solubilization of anthocyanins in apolar medium [185], and purification of bromelain [186-189].

Stevia (Stevia rebaudiana Bertoni), is known as a sweeter in the food industry as it has steviol glycosides which produce sweetness 100-300 times higher than sucrose. Its micellar structure in the aqueous phase enabled emulsifying features for the stabilization of oil-in-water emulsions as well as delivery systems with improved solubility and bioavailability of insoluble cargos [190].

Another important application of micellar systems in the food industry is related to using anionic micelles for stabilizing color. Phycoyanins are known as pigment-protein complexes that their color is dependent on pH. In a study by Mia Fiilsøe Falkeborg et al., sodium dodecyl sulfate (SDS) micelles were used for the stabilization of the color. The interaction of the non-protonated, circular helical (blue) structure of phycocyanin and the anionic SDS micelles was the main factor influencing the stabilization [191].

Casein is well-known as a natural vehicle for nutraceutical and bioactive ingredients. It is the most abundant protein in cow's milk and is able to form micellar nanoparticles with a size in a range of 50 and 20 nm naturally [192,193]. Table 2 illustrates important structural factors of casein micelles [194].

Generally, caseins consisting of four phosphopeptide subfractions (αs1-, αs2-, β-, and κ-caseins) bridged by calcium phosphate nanoclusters [195], were used as

Table 2. Average characteristics of casein micelles.

Factor	Value
Diameter	120 nm (range: 50-500 nm)
Surface area	8×10^{-10} cm^2
Volume	2.1×10^{-15} cm^3
Density (hydrated)	1.0632 g cm^3
Mass	2.2×10^{-15} g
Water content	63%
Hydration	3.7 g H_2O g^{-1} protein
Voluminosity	44 cm^3 g^{-1}
Molecular mass (hydrated)	1.3×10^9 Da
Molecular mass (dehydrated)	5×10^8 Da
Number of the peptide chain	5×10^3
Number of particles per ml milk	10^{14}-10^{16}
Surface of micelles per ml milk	5×10 cm^3
Mean free distance	240 nm

delivery systems for various food categories including vitamins, plant extract, phenolic compounds, and essential oils, probiotics, oils, and fatty acids [194]. Regarding integrating casein-based micellar systems, there are two different techniques for the incorporation of the target molecules into the casein carriers. In the first model which is named to-down techniques, the micelles are broken down into smaller particles to enhance the encapsulation's efficacy. On the other hand, the second method uses micelles' self-assembling features to gather target molecules in the micellar complex. The latter method is named the bottom-up technique [194].

Various studies used casein-based systems for different applications in the food industry. In a study by Nazanin Ghayour et al., quercetin and curcumin as two phenolic compounds were encapsulated in casein-based delivery systems to improve their low water solubility. As a result of this experimental work the chemical stability of the system was increased and the aqueous solubility of the targets was higher than in their naked situation [196].

To tackle the issue of deficiency of fat-soluble vitamin D_3 (VD_3), Levinson and coworkers investigated the effects of bioavailability of VD_3 from fat-free yogurt, in reassembled casein-micelles (rCMs) to assess physicochemical and sensory features with a comparison with polysorbate-80 as a common synthetic surfactant. Their results showed the rCMs could provide more protection as the carrier [197,198].

Moreover, it was shown that casein micelles can protect some targets like β-Carotene during key industrial processes such as sterilization, pasteurization, high hydrostatic pressure, and baking which is an important property on an industrial scale [199].

In another study, Esmaili et al. used β-Carotene as the nanocarrier to improve the solubility of curcumin in food industrial processes. They showed camel casein in this study was able to improve curcumin's solubility 2500 fold [200].

Kang Pan and coworkers used spray-dried ethanol solution with co-dissolved sodium caseinate (NaCas) for the enc capsulation of curcumin. The results indicated curcumin encapsulated with hydrophobic interactions and its bioavailability was increased [201].

Saponins which are a category of glycoside are applied as biosurfactants and have promising applications as food additives. For instance, quillaja saponins (QS) makes micelles including fat-soluble lutein esters to improve the solubility [202].

The improved stability of Chlorophyll α monomers encapsulated in the cremophor EL micellar system provided a new window to use such a self-assembly pigment-detergent nano micellar system that can be valuable in the food industry [203].

3.7 Oil and petroleum industry application

The widespread applications of micelles in various industry also been seen in the oil and petroleum section. In this regard, there are several types of research on oil recovery [204,205]. As before mentioned, WLMs are counted as one of the popular forms of micelles. Their aggregated structures are formed spontaneously using surfactant molecules in a solution. They could provide special properties since can break their structure and reconstruct dynamically

[206]. Although they have been studied for decades in various areas including biomedicine, the oil industry, tissue engineering, etc., the only application on large scale was in the oil and petroleum industry [207]. Their applications can be seen in oil well drilling, fracturing fluid cleaning, oil recovery, etc. [206]. There were researchers investigating the rheological behavior and various multi-responsive aspects of these worm-like micellar systems [206,208]. The use of environmentally-friendly fracturing fluids has been applied in the petroleum industry. By taking advantage of decreased environmental damages, improved operational efficacy, and higher fracture conductivity, the application of such fluids has attracted attention. Viscoelastic surfactants (VES) which are able to construct rod-like micellar systems have been utilized as fracturing and frac-packing fluids. Their experimental results approved faster fracture cleanup time, and improved productivity [209]. In fracturing fluid scope, some other studies are also done to improve the micellar systems' function [210,211]; for example, the construction of a worm-like micellar system was reformed with low molecular weight hydrophobically modified polyacrylamide (HMPAM) which showed an applicable temperature (30 ˚C higher than the WLMs) and improved shear resistance than WLMs [212].

Apart from this study, Zhe Li, Wanli Kang, et al. introduced an organic acid-enhanced viscoelastic surfactant consisting of WLMs by coupling an ultralong hydrophobic chain surfactant N-erucamidopropyl-N,Ndimethylamine(EA), with p-phthalic acid (p-PA). This reusable system showed low damage to core permeability and less steel corrosion in addition to appropriate proppant-carrying ability [213].

In a study by M.F. Attallah et al, mixed micelles solutions were applied for the treatment of Technologically Enhanced Radioactive Materials Found Naturally (TENORM) scale wastes which are producing in the oil and gas industry and contaminating by various radionuclides. Three different types of surfactants including polyoxyethylene sorbitanmonooleate (Tweeen-80), cetyltrimethylammonium bromide (CTAB), and sodium-dodecylsulfate (SDS) were used. The mixed micelles produced synergistic effects for remediation of the contamination which resulted in the removal of 80% of the most contamination factors (i.e. isotopes ^{226}R and ^{228}R) [214].

There are also some processes like micellar flooding for oil recovery which is based on emulsifying the crude oil. However, some important agents such as surface conditions are playing a key role in the appropriate function of micellar flooding. In this regard, a conventional surfactant SDS (88%) solution was tested by Rishiraj Goswami and coworkers. They investigated the effects of monovalent and divalent ionic strength of 0-4 wt% NaCl/$CaCl_2$. The results illustrated monovalent salt (0.5 and 1.0 wt%) in a temperature between 60 and 98˚C could produce over 60% cumulative oil [215].

Another important application of biodegradable SDS was introduced as a useful agent for the removal of filter cake of oil-based drilling fluid. The microemulsion system contained SDS, kerosene (oil phase), n-butanol (cosurfactant), and distilled water. Results showed that SDS could be offered as a good alternative to microemulsions and the cosurfactant plays a key role in the removal of filter-cake in these systems [216].

3.8 District heating/cooling fluids application

To discuss the application of micelles in heating and cooling systems, we first need to introduce the phenomenon of drag reduction. It is a physical phenomenon that reduces friction and increases fluid flow. Chemicals that reduce drag are called drag-reducing agents (DRA). In other words, DRA are able to decrease the turbulent flow of water and as a result the energy required to pump a fluid (Figure 8)[217]. District heating and cooling (DHC) systems are among the important applications of drag reducers where recirculation of fluid is required. They are widely used in eastern and northern Europe, the United States, Canada, Japan and Korea. DHC systems rely on centralized power generation or energy conversion systems which supply energy services to a number of buildings in a district. Energy is delivered as heated or chilled water via distribution channels to heat or to cool the surrounding buildings. The water is continuously recirculated (through a 10 to 20 km loop) to the central station for reheating or recooling.

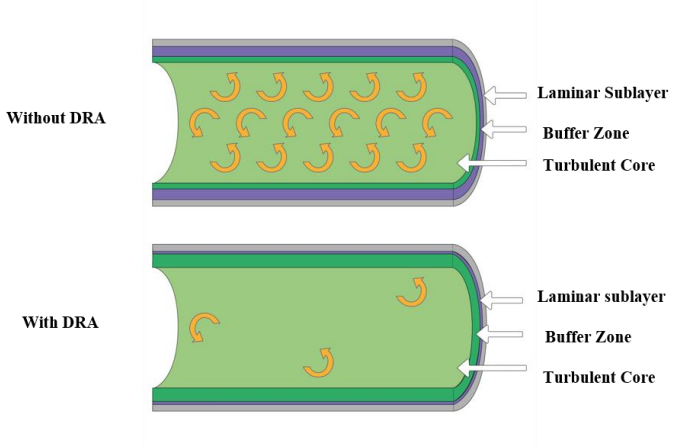

Figure 8. The effect of DRA in turbulence reduction in pipes [217].

The additives that reduce drag can be divided into five categories: polymers, surfactants, fibers, microbubbles, and compliant coating [218]. Surfactants and polymers are most commonly used in industrial application. The drag reduction ability of polymers depends on many factors such as molecular weight, aggregation, and chain flexibility [219]. A linear flexible polymer with a high molecular weight is generally considered to be the most effective drag-reducing polymer. Polymer DRA have been applied to reduce by more than 70% the pumping energy in the transportation of hydrocarbon fluids over the past 50 years [220]. As a common drag reducer, polyacrylamide (PAM) has often been used; in some cases, surfactants were also employed [221]. Traditional polymers are degraded and lose their efficiency by pumping circulation. However, since the WLMs have a dynamic association structure, they can break and form through self-association as a 'living polymer'[222].

Moreover, the effect of different molecular structure was studied. The narrow temperature range for effective drag reduction limits the usefulness of non-ionic surfactants. Precipitation caused by bivalent ion (Ca, Mg) in hard water limits the application of anionic soap surfactants. The cationic surfactants were extensively studied as it has a wide temperature range and no sensitivity to Ca and Mg ions [222]. For drag-[223] reducing cationic surfactant solutions, there is a minimum micelle length below which rodlike micelles are unable to induce drag reduction. Because of electrostatic interactions between the surfactant headgroups which limit the growth of micelles in the dilute region, cationic surfactants alone cannot form strong threadlike micelles necessary for drag reduction at low concentrations. Through the addition of counterions to the solution, electrostatic repulsion forces between cationic surfactant molecules in a micelle can be reduced, thus promoting micelle growth. Because of the greatly screened headgroup repulsion forces between surfactant molecules and the increased dimensional packing parameter of micelles, the length and flexibility of the micelles increase considerably. With strongly binding counterions, the resulting surfactant solution resembles a polymer solution with viscoelasticity and strong extensional resistance [224]. A typical drag-reducing cationic surfactant is cetyltrimethylammonium bromide (CTAB) and chloride (CTAC) in water. These surfactants generally form micelles when mixed with an additive, such as sodium salicylate (NaSal). Tamano *et al.* comprehensively investigated nonionic-type surfactants of alkylamine N-oxide, oleyl-N, N-dimethylamine N-oxide (ODMAO) and octadecyl-N, N-bis(2-hydroxyethyl)amine N-oxide (C18BAO), in ethylene glycol aqueous solution for turbulent pipe flow. Compared to the effect of the individual surfactant, the mixture of ODMAO and C18BAO in EG aqueous solution exhibited excellent drag reduction exceeding 60% in a wide range of solution temperature ranging between 20 and 60 ℃, perhaps due to a synergetic effect. However, the drag reduction

performance deteriorated below 0 ℃and beyond 60 ℃. To conclude, in addition to cationic and zwitterionic surfactants, nonionic-type surfactants are promising candidates as drag reducing additives in EG aqueous solution [225]. Moreover, it was found that an increase in the length of the alkyl chain increases the upper temperature limit for effective drag reduction. The drag reduction effect was reduced in the middle temperature range of 60-80 ℃. In addition to surfactant characteristics in alkyl chain length, saturated/unsaturated chain, odd/even numbers of carbons, the characteristics of counterion including size, polarity, etc. have effect on feature of DRA [224]. The most effective counterion is aromatic hydrophobic salts such as salicylate and 1-naphthol. The environmentally friendly drag-reducing properties of N-alkyl, N, N-dimethylglycinate combined with sodium alkylbenzene sulfonate at a 4:1 molar ratio was found useful for such applications. The wide use of drag reduction surfactants depends on development of more environmentally friendly surfactants and the modification of proper mechanical systems for such applications [226-228].

A photoresponsive micellar solution was developed by Shi et al. as a promising DRA for DHC systems. This solution is a smart fluid unbaling to be reversibly switched between a drag reduction mode and an efficient heat transfer (EHT) mode by external stimuli. The DR mode is beneficial during fluid transport, and the EHT mode is preferred when the fluid passes through heat exchangers. This fluid contains an aqueous solution of cationic surfactant oleyl bis(2-hydroxyethyl)methyl ammonium chloride (OHAC, 3.4 mM) and the sodium salt of 4-phenylazo benzoic acid (ACA, 2 mM). ACA is initially in a trans configuration and the OHAC/ACA solution is viscoelastic and exhibits 80% DR in comparison with pure water. This solution cannot be effective for heat transfer at the same time. Upon UV irradiation, the trans isomer of ACA changes to cis isomer, resulting in the solution being transformed into its EHT mode with no viscoelasticity and drag reduction ability, but this time, it acquires heat-transfer ability relative to that of water. The fluid can back to DR mode and retain its viscoelasticity through subsequent irradiation with visible light. In the DR mode, the OHAC/trans-ACA molecules assemble into long threadlike micelles that give the capability of viscoelasticity and drag reduction to the fluid. OHAC and cis-ACA, on the other hand, form much shorter cylindrical micelles in the EHT mode in which viscoelasticity is negligible and heat transfer is effective. The schematic illustration of this switched modes can be seen in Figure 9 [229].

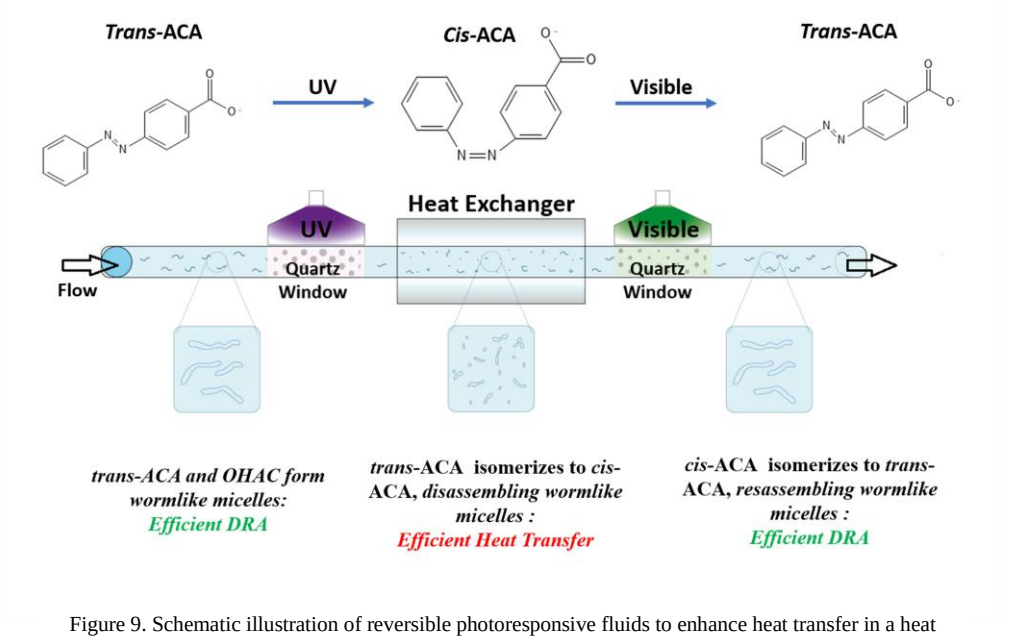

Figure 9. Schematic illustration of reversible photoresponsive fluids to enhance heat transfer in a heat exchanger temporarily [229].

In general, the phenomenon of drag reduction in turbulent flow, attracts many attentions not only in DHC section but also in other sections involving turbulent fluids namely oil production [230], firefighting [231], and sewage systems [232]. Therefore, micellar solutions are also widely applicable in these areas.

4. Conclusion

The special characteristics and activities of micelles, particularly in recent decades, promoted great attention from the scientific society which resulted in the introduction of different types of micellar systems and various applications. Nano micellar delivery systems and micellar-based techniques have been emerged to increase the solubility, stability, and biological properties of many compounds with various applications as well as improve the functional processes on an industrial scale. Micelles by providing hydrophobicity and hydrophilicity at the same time are good options to be used for promoting materials' properties, especially insoluble drugs. Therefore, it is of great significance to engineer micelles, and take advantage of their unique structural properties to produce more applicable and smart compatible micellar systems for various industries.

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