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Photoresponsive microgels and their applications: a review

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Photoresponsive microgels have gained significant importance in modern-day scientific research due to their fascinating properties and potential applications in various fields. This review presents the reported schemes for their synthesis and design, mechanism of photoresponsiveness, characterization and current applications. These microgels are intelligent polymeric materials that show a swift response (swelling/deswelling) upon irradiation with light due to the specific photoresponsive moieties incorporated in their network. These photoresponsive microgels and their hybrids are biocompatible, functional, less toxic and can be characterized using a number of characterization techniques. The intelligent behavior of these microgels makes them suitable candidates for use in drug delivery and release, tissue engineering, actuators and sensors, adaptive surfaces and smart coatings, catalysis, environmental science, photonic devices and many other fields. This overview highlights current developments in this specific area and potential directions for future study.

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1 Introduction

Smart microgels are adaptable soft polymeric colloidal materials known for their unique structures and responses to stimuli. They consist of cross-linked polymer particles with diameters ranging from 0.1 μm to a few hundred μm. The term "microgel" first appeared in scientific literature in around 1963.[1] These particles can expand in response to environmental changes, like temperature, pH, radiation, and ionic strength. This key feature sets them apart from other colloidal systems.[2] Microgels have a three-dimensional network of cross-linked polymer chains swollen with a solvent. The swelling behavior depends on the interactions between the solvent and the polymer as well as the structure of the polymer. The ability to swell and shrink gives microgels adjustable properties, making them useful for many purposes, including drug delivery,[3] sensors,[4] and stabilizers for nanomaterials.[5] Several factors shape the microgel properties, including crosslinking density,[6] responsive behaviour,[7] mechanical properties,[8] and colloidal stability.[9] Due to their unique properties, microgels have a wide range of

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applications in drug delivery,[10] sensors,[11] actuators,[12] enhanced oil recovery[13] and biomedical fields.[14]

"Smart" or "intelligent" polymers that can sense changes in their environment have grabbed the attention of scientists over the past few decades.[7] These materials can change their physical and chemical properties or shape in response to certain stimuli. These materials might change their color, become transparent, start conducting electricity, show water permeability, or change shape (shape memory polymers). These changes can happen because of chemical (like pH, redox potential or ionic strength),[15] physical[12] (temperature, light, ultrasound, mechanical stress, electrical or magnetic field) or biological[16] (enzymes, antigens, glucose, ligands or other biochemical agents) stimuli. These responsive microgels have become a hot topic in the last ten years because they could be useful in many ways, like in delivering drugs,[10] making sensors and actuators[11,12] and helping with regenerative medicine.[17]

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Light-sensitive microgels are colloidal particles that alter their properties when exposed to radiation of a suitable wavelength.[18] These microgels are fabricated by adding light-sensitive parts, like spiropyrans, azobenzenes, or coumarins, to the polymer network. When light of a specific wavelength shines on them, the microgels can swell or shrink, making them useful for many applications. Light has many perks as a trigger compared to other stimuli. It is clean, cheap, and can be an effective stimulus. Unlike other triggers, you can control light in space and time and switch it on from outside the system.[19] By tuning various irradiation parameters, such as light intensity, exposure time and wavelength, the degree of light-induced bending deformations can be well-controlled. Photoresponsive microgels have also drawn considerable attention in recent years because of their ability to sensitively control the responses spatiotemporally.[20] As a result, more and more people are becoming interested in photoresponsive polymeric microgels. Various new kinds of smart materials that react to light have popped up in fields such as nanotechnology, electronics, diagnostics, and medicine. These materials can change properties such as their shape, phase, solubility, permeability, and wettability, and how well they transform when light hits them.[21] Light-sensitive microgels have many uses in delivering drugs,[22] medicine and speeding up chemical reactions,[23] as well as in bioimaging and sensors[24] and as actuators. Over the past few decades, many researchers have investigated the synthesis, properties, and applications of photoresponsive microgels.[25,26] They have done a lot of work to make them, figure out how they are built, how they respond to light, how to characterize them, and how to use them in different areas. However, there is an urgent need to develop a greater scope for the application of photoresponsive microgels. Besides applications in drug delivery, catalysis, actuators, and environmental remediation, applications in the areas of microsensor arrays, micro-

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optomechanical systems, and membrane technology should also be focused on and explored. But we need to find more ways to use photoresponsive microgels in various fields. This review summarizes recent advancements in the synthesis, properties and applications of photoresponsive microgels.

2 Synthesis and design of photoresponsive microgels

2.1 Types of light-sensitive components

Photochromism can be simply described as a light-triggered reversible change of a chemical compound between two states with different light absorption patterns. When light-absorbing molecules are exposed to light with a specific wavelength, they can undergo a reversible or permanent shift between isomeric forms or ring opening/closing. The commonly used light-sensitive substances fall into two main groups: those that are light-switchable and those that are light-breakable. A substance shows photochromism when it can reversibly change between two states, triggered in one or both directions by different wavelengths of radiation.[27] Common light-switching groups include azobenzene, spiropyran, diarylethene, fulgide, salicylideneaniline, and coumarin.[20] Less frequently used are malachite green, leuconitriles, and diketone derivatives. Light-breakable groups differ from light-switchable ones mainly in their lack of reversibility. Pyrenylalkyl and *o*-nitrobenzyl esters are typical examples in this category. A third type of photoresponsive compound relies on the photothermal effect of plasmonic metal nanoparticles, such as gold.[28]

2.2 Methods of synthesis

Photoresponsive microgels can be designed in various ways. To synthesize photoresponsive microgels and hybrid microgels, a light-responsive functionality or component is incorporated into the microgel structure. The light-sensitive moieties are incorporated in various ways. Some methods of incorporation of photoactive components into the polymeric structure are discussed below.

2.2.1 By copolymerization of a photoactive component with monomers. By utilizing different polymerization techniques, the photoactive moiety is copolymerized with other comonomers and crosslinkers. The photoactive units are part of the polymeric system. These photoresponsive moieties undergo a change in their structure through different photoresponsive mechanisms, which cause the swelling and shrinkage of microgels or hybrid microgels under light stimulus. A poly(VCL-bis-ABSA) based microgel system was synthesized by chemically crosslinking and copolymerizing a photoresponsive moiety, 4-[(4-methacryloyloxy)phenylazo]benzenesulfonic acid (ABSA), and a thermoresponsive moiety, *N*-vinylcaprolactam (VCL), *via* surfactant-free radical precipitation polymerization of ABSA and VCL using *N*,*N*′-methylenebisacrylamide (BIS) as a crosslinker.[29] In addition to the monomer, a photoresponsive component may also be used as a crosslinker in the synthesis of a photoresponsive microgel system. For example, poly(*N*-isopropylacrylamide) [P(NIPAM)] based photoresponsive microgels were developed by using 4,4′-di(acrylamido)azobenzene (DAAB) as a crosslinker.[30] DAAB was selected as a crosslinker in the synthesis of the microgel system because it translates the photoisomerization process to a change in the microgel structure. As a result, a microgel response to the light stimulus takes place. Moreover, different polymerization techniques, such as reversible addition-fragment chain transfer (RAFT) polymerization, have been used to synthesize polymeric systems, which are then post-modified with light-sensitive functionality *via* coupling.[31]

2.2.2 By host–guest interaction. Photoresponsive components can also be incorporated into microgel systems *via* noncovalent functionalization. Host–guest interactions, electrostatic complexation, physical cross-linking with photoactive components, or photo-dimerization of pendant groups are included in noncovalent methods to design photoresponsive microgels. For example, a novel dual responsive (temperature and light responsive) microgel system based on poly(*N*-isopropylacrylamide) [P(NIPAM)] has been synthesized by Liang *et al.*[32] The host–guest interactions between β-cyclodextrin (β-CD) and *trans*-azobenzene (*trans*Azo) are the source of crosslinking within the microgel network. The same type of host–guest interactions was employed to functionalize poly(2-methyl-2-oxazoline) (PMOXA) chains. PMOXA chains are then grafted with polyacrylamide (AAm) to make a photoresponsive microgel network.[33] A photoresponsive microgel system consisting of poly(*N*-isopropylacrylamide-*co*-acrylic acid) [P(NIPAM-AA)] was made by Sharma *et al.*[34] by covalently complexing the anionic microgel system with cationic spiropyran surfactant. The resultant photoresponsive microgels showed reversible changes in size and gave a volume phase transition (VPT) in response to changes in the light intensity and wavelength of the incident light.

Photoactive groups are attached, or polymeric chains are crosslinked, by using physical interactions in noncovalent methods to fabricate photoresponsive microgels instead of using chemical interactions. Without changing the chemistry of the polymeric network, dynamic, reversible modulation of the microgel characteristics in response to light is offered by this method.

2.2.3 By loading inorganic material into the polymeric systems. Photoresponsivity is also introduced in the microgel system when nanoparticles of certain metals are incorporated into the microgel network to form hybrid microgels; however, here, our discussion is limited to microgels loaded with plasmonic metal nanoparticles. Ag[35–37] and Au[38–40] nanoparticles are well-known plasmonic metal nanoparticles that have been extensively incorporated into smart microgel systems to obtain photoresponsive microgels.[41,42] Stimuli-responsive microgels with incorporated nanoparticles, such as silver nanoparticles (AgNPs) or gold nanoparticles (AuNPs), exhibit photothermal effects. For example, a composite microgel system was synthesized by Zhang *et al.*[43] by combining photothermal plasmonic gold nanorods (AuNRs) and a photoresponsive polymeric network consisting of P(NIPAM). The AuNRs absorb light at two particular wavelengths, known as the transverse and longitudinal surface plasmon resonance wavelengths (λ_{SPR}). When light with wavelength λ_{SPR} falls on the gold nanorods, the

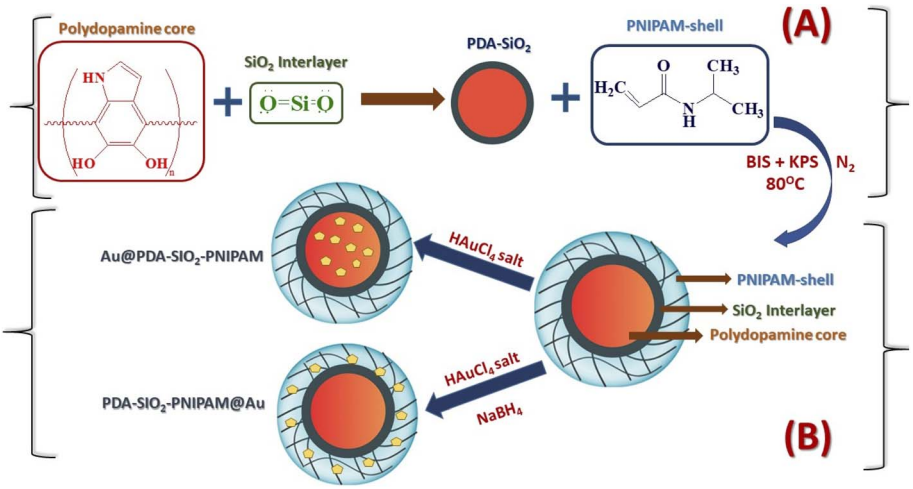


Fig. 1 Schematic of the synthesis of the polydopamine (PDA) core encapsulated in a SiO_2 interlayer (A) and construction of the PNIPAM shell and the formation of hybrid microgels with AuNPs in the core (Au@PDA-SiO_2-PNIPAM) and AuNPs in the shell (PDA-SiO_2-PNIPAM@Au) (B).

amplitude of oscillations of the surface electronic cloud of the gold nanorods is increased. When this excited state returns to the normal state, energy is released in the form of heat. This heat energy causes a rise in temperature, resulting in a fast thermal and photoresponse of the hybrid microgel system, which acts as a smart switch. Similarly, Chang *et al.*[44] reported the synthesis of hybrid microgels for the catalytic reduction of *p*-nitrophenol (*p*-NP). Two kinds of hybrid microgels were synthesized through electrostatic interaction and synergistic bonding between the AuNPs and the microgel networks. The electrostatic self-assembly approach between mercapto-modified polyethylene glycol (mPEG) and polyallylamine (PAH) was utilized to incorporate gold nanorods (AuNRs) to make a photoresponsive drug-delivery system consisting of AuNRs@poly(*N*-isopropylacrylamide-*co*-acrylic acid) [AuNRs@P(NIPAM-*co*-AA)] microgels.[45] Photothermally responsive nanoreactors with a yolk–shell structure have been synthesized by Xu *et al.*,[46] allowing control of catalyst placement. For this purpose, the polydopamine (PDA) core was encapsulated in a SiO_2 shell, and then a second shell of PNIPAM was constructed around the PDA-SiO_2 core shell system to obtain PDA-SiO_2-PNIPAM using potassium persulphate (KPS) as an initiator and *N*,*N*-methylenebisacrylamide (BIS) as a cross-linker under a N_2 atmosphere at 80 °C, as shown in Fig. 1(A). Interestingly, Au nanoparticles were fabricated in the PDA core without using any additional reducing agent, because $AuCl_4^-$ ions adsorbed on PDA were reduced to Au nanoparticles by the catechol groups of the PDA to generate the Au@PDA-SiO_2-PNIPAM system, while loading of Au nanoparticles in the PNIPAM shell of the PDA-SiO_2-PNIPAM system, in the form of PDA-SiO_2-PNIPAM@Au, was achieved by reducing $AuCl_4^-$ ions complexed with the nitrogen atom of PNIPAM using sodium borohydride as an external reducing agent, as shown in Fig. 1(B).

Table 1 highlights the various photoresponsive microgel systems and polymerization methods, photoactive components, functionalization approaches, and possible applications.

3 Mechanism of photoresponsivity in microgels

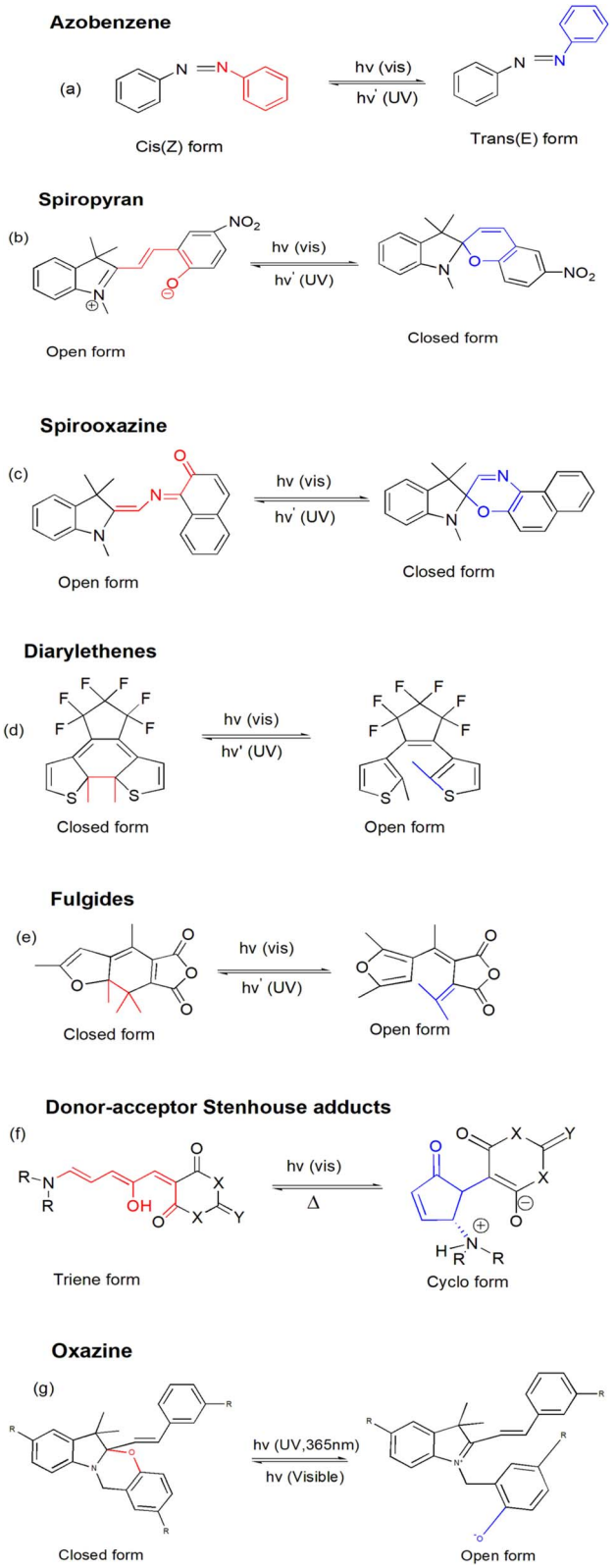
Photoresponsiveness in microgels mainly results from adding light-sensitive parts to the microgel's structure, allowing swelling or shrinking when exposed to different light wavelengths. Different photoresponsive mechanisms cause changes in the structure of the photoactive moiety in response to light. This leads to changes in hydrophilicity/hydrophobicity or crosslinking density of the microgel network, which, in turn, change the functional behaviour or swelling property of microgels. Some key mechanisms are described here.

3.1 Photoisomerization

This involves light-induced isomerization, which can be reversible or irreversible, depending on the photoswitchable compounds. The reversibility or irreversibility of the light-responsive behaviour of microgels is dependent on the chromophore type.[57] Diarylethene,[58] azobenzene,[57] spiropyran,[19] spirooxazine,[59] and DASA (Donor Acceptor Stenhouse Adducts)[60] are examples of reversible photoresponsive moieties that can undergo cyclic transition between the isomers. The reversible photoresponsive moieties follow two types of isomerisation mechanisms. The first mechanism is *cis–trans* isomerization, which results in no bond breakage. Azobenzene is a common example of a reversible photoswitchable moiety that undergoes *cis–trans* isomerization. This type of isomerization has little effect on the electric dipole moment but alters the molecular structure significantly.[61] Bond cleavage is the other type of isomerization process, with spiropyran as a prominent example. When exposed to ultraviolet light, this reversible isomerization can cause a significant change in polarity in addition to a color shift.[61] Photoresponsive microgel systems undergo a physicochemical transformation due to the reversible isomerization of photoresponsive components under the stimulus of light. Near-infrared light exposure changes the photochromic part from hydrophobic to hydrophilic, causing the

Table 1 Photoresponsive microgel systems synthesized using different synthesis approaches and their possible applications

Photoresponsive/plasmonic microgel system	Polymerization approach	Photoactive component	Functionalization method	Application	Ref.
Au layer@P(NIPAM-AA)-azobenzene	Free-radical polymerization	Azobenzene and gold layer	Electrostatic complexation and adsorption on a gold layer	Spatio-temporal tuning based on the reversible expansion and contraction of a microgel network	47
Poly(NIPAM-*co*-AA)-azo microgels	Microfluidic technique	Cationic trimethylammonium bromide containing azobenzene	Electrostatic interactions	Potential applications in drug delivery, catalysis, sensors and functional coatings	48
Poly(*N*-isopropylacrylamide-*co*-aminoethyl methacrylate-Rose Bengal)	Single-step emulsion polymerization	Rose Bengal	Covalent interactions	Green synthesis of 5-hydroxy-2(5*H*)-furanone and photocatalytic degradation of diclofenac	49
Poly(*N*-isopropylacrylamide-*co*-nitrobenzyl methacrylate)	Precipitation polymerization	Nitrobenzyl methacrylate	Covalent interactions	Drug delivery	50
AuNPs@poly(*N*-isopropylacrylamide-*co*-acrylic acid) microgels	Surfactant-free and free radical precipitation polymerization	AuNPs	Adsorption (host–guest interactions)	Etalons fabrication with potential application in drug delivery	51
AuNPs@P(NIPAM)	Precipitation polymerization	Hollow AuNPs	Hybrid synthesis (electrostatic coupling *via* polyelectrolyte linker)	Vectors in drug-delivery systems	52
AuNPs@poly(*N*-isopropylacrylamide-*co*-allylamine)	Precipitation polymerization	AuNPs	Non-covalent functionalization (electrostatic interactions)	Light-based manipulation of pickering emulsion droplets	38
AuNPs@poly(methylmethacrylate-4-vinylpyridine) hybrid microgels	Free radical polymerization	AuNPs	Hybrid synthesis (*in situ* reduction)	Catalytic applications	42
Ag/poly(*N*-isopropylacrylamide-*co*-acrylic acid) microgels	Free radical precipitation polymerization	AgNPs	Electrostatic interactions	Sensors to efficiently track the motion of human joints	37
Ag–Au@poly(*N*-isopropylacrylamide-*co*-3-methacryloxypropyltrimethoxysilane) hybrid microgels	Seed emulsion polymerization followed by galvanic replacement reaction	AgNPs and AuNPs	Electrostatic interactions	Optical and catalytic applications	53
AuNPs@poly(*N*-isopropylacrylamide-hydroxyethyl methylacrylate) microgels and AuNPs@MG-SH (thiolated microgels)	Surfactant-free emulsion polymerization	AuNPs	Electrostatic interactions and synergistic bonding	Temperature-based controlled catalysis	44
AuNR@(AgNPs/PNIPAM) hybrid microgels	Seed precipitation polymerization	AuNR and AgNPs	Electrostatic interactions (adsorption)	Photoresponsive catalytic reduction of 4-nitrophenol	54
Au/Ag NPs@poly(*N*-isopropylacrylamide-*co*-acrylic acid-*co*-acrylamide) microgels	Free radical precipitation copolymerization	AgNPs and AuNPs	Coordination interactions and electrostatic interactions	Enhanced optical characteristics with potential application in biomedicine	55
AgNPs@[poly(*N*-isopropylacrylamide)-poly(acrylic acid)] interpenetrating polymer network microgels	*In situ* polymerization	AgNPs	Coordination interactions and electrostatic interactions	SERS (surface-enhanced Raman scattering) microsensors	56



Scheme 1 Photoisomerization of reversible photoswitchable compounds (a)–(g) used as photoresponsive organic components of microgels.

carrier to change shape and release its contents. Scheme 1 shows how reversible photoswitchable parts change when exposed to light of a specific wavelength.

3.2 Photocleavage

NIR light causes the polymer to break down through photoinduced cleavage, allowing for better cargo release and easier removal of the resulting small molecules. Coumarinyl ester is more readily split by two-photon NIR light compared to *ortho*-nitrobenzene (ONB).[62,63]

3.3 Photodimerization

Photodimerization is a direct-light reaction where each polymer build-up step is started by an absorbed light particle/photon. The coumarin group serves as a reversible linking point. Lu *et al.*[63] used coumarin group photodimerization to create photoresponsive elements in microgels, forming triply responsive microgels.

3.4 Photothermal effect

The photothermal effect is another mechanism by which photoresponsive microgels respond to light stimulus. Nanoparticles of certain metals, such as gold, exhibit a photothermal effect. These nanoparticles are often incorporated into the microgel network by using different methods to render them light-responsive.[64] These nanoparticles show strong absorption bands in the visible and near-infrared (NIR) regions, depending upon the shape and size of the Au nanoparticles. As a result, this allows for a very effective conversion of light energy into heat energy. When this heat energy is released into the environment, it causes an increase in temperature. This, in turn, causes a change in the size of the microgel network; *i.e.*, it causes photothermal deswelling or swelling of the microgels.[65]

4 Characterization

Key techniques for studying photoresponsive microgels include scanning electron microscopy (SEM), transmission electron microscopy (TEM), scanning transmission electron microscopy (STEM), nuclear magnetic resonance (NMR) spectroscopy, Raman spectroscopy (RS), dynamic light scattering (DLS), UV/visible spectroscopy (UV-vis) and Fourier-transformed infrared spectroscopy (FTIR). Computer-based methods, like molecular dynamics simulations and finite element analysis, can model light-induced shape changes in light-sensitive parts and assess their effect on overall microgel swelling.[66] Table 2 provides a brief overview of some techniques used for the characterization of photoresponsive microgels.

5 Applications of photoresponsive microgels

Light-sensitive microgels are smart materials that change when exposed to light. These unique microgels and hybrid microgels have many potential applications. Here are some key ways in which light-sensitive microgels can be used.

Table 2 Analytical techniques used for the characterization of photoresponsive microgels

Photoresponsive microgel system	Characterization technique	Purpose of the technique	Ref.
Dual responsive spiropyran-modified poly(*N*-vinylcaprolactam) microgels	^{1}H-NMR, FTIR, GPC, UV-vis and DLS	^{1}H-NMR and GPC confirmed the microgel formation. FTIR identified the functional groups. UV-vis provided the qualitative and quantitative analysis. DLS measured the hydrodynamic radius of the particles	31
Dual responsive donor acceptor Stenhouse adduct-modified microgels	^{1}H-NMR, FTIR, and Raman spectroscopy, TEM, UV-vis spectroscopy, and DLS	^{1}H-NMR verified the microgel formation. FTIR identified the functional groups. Raman spectroscopy quantified the GMA (glycidyl methacrylate) content. TEM determined the shape and size distribution. UV-vis quantified the *p*-anisidine content and photoswitching behavior. DLS measured the hydrodynamic radii	60
Azobenzene (mAzo)-modified poly(*N*-vinylcaprolactam-*co*-acrylic acid) microgels	Raman spectroscopy, STEM, UV-vis spectroscopy, DLS, and EM (electrophoretic mobility)	Raman spectroscopy quantified the AAc in microgels. STEM showed the morphology and distribution. UV-vis confirmed the light responsiveness. DLS measured the hydrodynamic radii. EM determined the electrophoretic mobility at different pH values	67
Spiropyran (SP)-modified poly(*N*-isopropylacrylamide-acrylic acid) microgels	DLS, TEM, SEM, RS (reflectance spectroscopy), UV-vis, AAS (atomic absorption spectroscopy), FM (fluorescence microscopy), and TEM	DLS and TEM measured the particle size. SEM showed the surface morphology. RS checked the thermoresponsiveness. UV-vis monitored the light and pH responsivity. AAS determined the Cu^{2+} ion content. FM investigated the NIR response	24
Poly(*N*-isopropylacrylamide-*co*-nitrobenzyl methacrylate) microgels	TEM, DLS, UV, and LC-MS	TEM and DLS determined the morphology and hydrodynamic radius. UV-vis measured the drug loading. LC-MS quantified the drug release over UV exposure time	68
Malachite green (MG) (photochromic dye)-integrated poly(*N*-isopropylacrylamide) hybrid microgels	FTIR, SEM, TGA, UV-vis, and RS	FTIR identified the functional groups. SEM calculated the particle size distribution. TGA performed the thermal analysis. UV-vis characterized the polymer synthesis and dye blending. RS characterized the microgel etalons with different metal layers	69
Gold nanoparticle core coated with poly(*N*-isopropylacrylamide) shell	TEM, UV-vis, and DLS	TEM calculated the AuNP size in the hybrid microgels. UV-vis determined the λ_{SPR} of the gold nanoparticles. PCS (photon correlation spectroscopy) characterized the thermoresponsiveness of the particles	70

5.1 Medical applications

Combinations of photoresponsive moieties and crosslinked polymeric systems have been widely reported for a variety of biomedical applications, including the development of dressing materials with wound healing and antibacterial properties,[71] injectable antitumor agents,[72] drug delivery[73] and tissue engineering. A few of them are described in the following subsections.

5.1.1 Loading and releasing drugs. Light-sensitive microgels can carry drugs that are released when irradiated. This allows for controlled and targeted drug delivery. Controlled release is crucial for making drugs work better and reducing

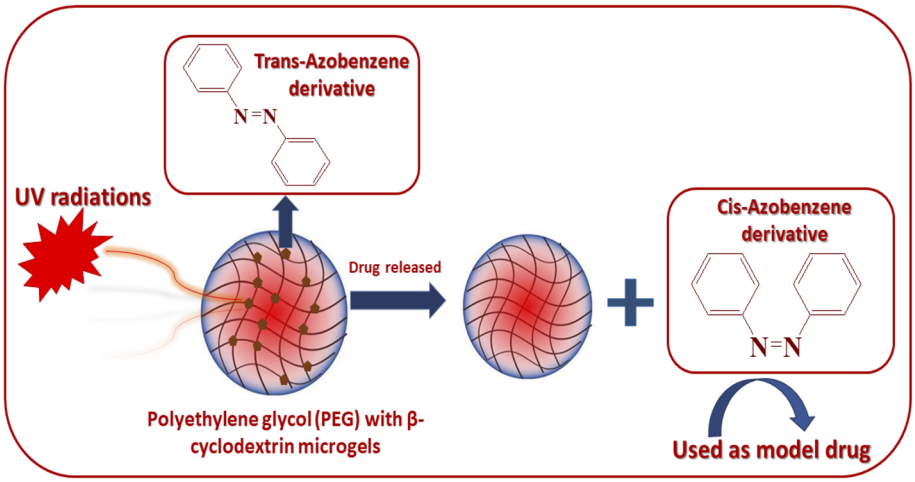


Fig. 2 Schematic illustration of UV radiation triggering drug release from a microgel network due to the isomerization of the *trans* form of the azobenzene derivative into its *cis* form, upon irradiation with 365 nm light, resulting in a decrease in host–guest interactions.

side effects by precisely controlling where and when drugs are released in the body. Light-sensitive microgels allow drugs to be released at specific times and places. Different pharmaceutical drugs can be trapped in 3-dimensional microgel networks and released when light triggers the transition. Functional groups that are photochromic or photolabile, *i.e.*, which change structurally or form new bonds when exposed to specific light wavelengths (UV, visible, or NIR), are used in photoresponsive microgels to achieve controlled drug release. The microgel network is modulated by these chemical alterations, enabling controlled drug release. The drug release in photoresponsive microgels can be controlled through various light-induced mechanisms. One of these mechanisms is the isomerization of azobenzene or related functional groups within the microgel polymeric network, which changes the conformational structure, polarity, or hydrophilicity of the microgel network. As a result, the microgel system can either swell or deswell, depending on the light intensity. The drug release rate is regulated in this way.[74] When exposed to light, the polymeric network can be degraded or de-crosslinked due to the breakage of the bonds of the photoresponsive moieties, such as coumarin or *ortho*-nitrobenzyl groups, present in the microgel network. This mechanism enables the encapsulated medicine to diffuse out of the microgel network by increasing its mesh size or the density of the pores.[75] In some photoresponsive microgels, a localized heating effect due to plasmonic nanoparticles present in the microgel network causes swelling or shrinkage of the microgel upon exposure to light radiation, allowing easy diffusion and release of the drugs.[76] In photoresponsive microgels, drugs are loaded either during the synthesis of microgels or post-synthesis loading of medicine can also be done. According to kinetic models such as Korsmeyer–Peppas, when light-sensitive microgels are exposed to light, mechanisms such as isomerization, photocleavage, or heating cause a change in the structure of the microgel, releasing the medication mostly *via* Fickian diffusion.[77] A dual (pH and thermo) responsive hybrid microgel system was reported by Xiao *et al.*[65] These microgels were loaded with fluorescent dyes and AuNPs. The AuNPs inside the polymeric network aggregated due to the shrinkage of microgels in response to pH or temperature. The aggregation causes the red shift in the NIR absorption of the AuNPs. This hybrid microgel behaviour made them useful for applications in photodynamic therapy, photoacoustic imaging, and photoactivated hyperthermia. Near-infrared (NIR) responsive microgels are especially promising for medical treatment because they are safe and can penetrate tissue deeper than other gels. Multi-stimuli-responsive microgels based on poly(*N*-isopropylacrylamide-*co*-nitrobenzyl methacrylate) [P(NIPAM-*co*-NBMA)] have been developed for the delivery and release of hydrophobic drugs when exposed to ultraviolet light.[50] Nehls *et al.*[78] showed how drugs could be released using light to control host–guest interactions between a gel and a drug molecule. They made microgels from the combination of polyethylene glycol (PEG) and β-cyclodextrin and used azobenzene functionalized peptide as a model drug. When this drug-loaded microgel system was exposed to 365 nm light, the drug was released (Fig. 2). The β-cyclodextrin component of the microgel system has a hydrophobic cavity that can accommodate the azobenzene derivative in its *trans*-form (planar form) through hydrophobic interaction. However, upon irradiation with light of 365 nm wavelength, the *trans* form of the azobenzene derivative isomerizes into its *cis* form [similar to the *trans* to *cis* conversion of azobenzene as shown in Scheme 1(a)], leading to the disruption of the complex of β-CD and the drug (azobenzene derivative) to increase the rate of release of the drug.

For the first time, Hu *et al.*[60] created new microgels that respond to both visible light and temperature. They added donor acceptor Stenhouse adduct (DASA) molecules to poly(*N*-vinylcaprolactam) microgels using a simple method. The DASA molecules were attached to the microgel structure through epoxy groups. These microgels were used to carry and release a fluorescent dye when exposed to visible light (Fig. 3). In the dark, the microgel exists in the colored, hydrophobic de-swollen state with the DASA-triene form, while, upon irradiation with

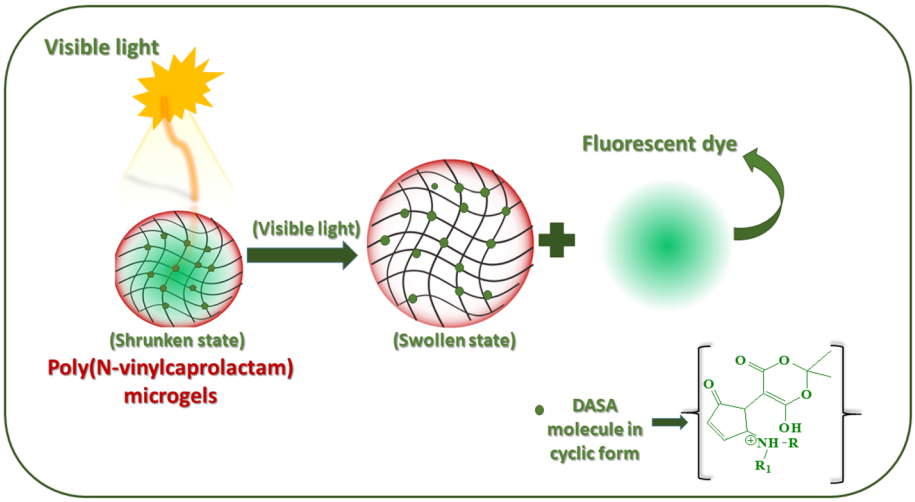


Fig. 3 Schematic illustration of the light-induced switching of the poly(*N*-vinylcaprolactam)@DASA microgels between their shrunken and swollen states and the release of a fluorescent dye.

visible light, the microgel system is converted into the colorless, hydrophobic swollen form due to the formation of the DASA-cyclic form, with release of the drug. The conversion of DASA-triene form into DASA-cyclic form upon irradiation with visible light is shown in Scheme 1(f). This suggests their potential applicability for the delivery of hydrophobic drugs using light as a trigger.

Protein hydrogels show promise for biomedical use due to their precise control over protein properties. However, research on photoresponsive protein hydrogels lags behind synthetic polymers. Wang *et al.*[79] developed B12-dependent light-sensitive protein hydrogels to control protein and stem cell release. These gels use CarHC (photoreceptor C-terminal adenosylcobalamin binding domain) that form tetramers with adenosylcobalamin (AdoB12) in darkness but break apart under green (522 nm) or white light. This dramatic structural change causes a quick gel-to-liquid transition, effectively releasing proteins and live cells when exposed to white light. The CarHC domains tetramerize when they bind to adenosylcobalamin (AdoB12) under dark conditions. The protein's structure is drastically changed when it is exposed to green (522 nm) or white light because they readily split into monomers. The hydrogel underwent a quick gel–sol transition due to light-induced CarHC disassembly, which was facilitated by the AdoB12-dependent CarHC tetramerization. When subjected to white light, this approach has been shown to efficiently encapsulate and release protein molecules together with living cells. Liu *et al.*[80] used emulsion polymerisation to generate multiresponsive folic acid-coupled poly(NIPAM-*co*-functional palygorskite-Au-*co*-acrylic acid) [(FA-PNFA)] hybrids for the efficient release of doxorubicin hydrochloride (DOX), for the treatment of breast cancer. These hybrid microgels, which have the ability to respond to many stimuli, including light, pH, and temperature, successfully decreased the proliferation of cancer cells, hence expanding their potential applications in medicine. Near-infrared (NIR) light is preferred for *in vivo* use due to its deeper tissue penetration and lower health risks. Chen *et al.*[81] developed an NIR-controlled drug-delivery system using PEGylated gold nanorods loaded on thermoresponsive microgels to release DOX. This hybrid microgel system releases the drug *via* photothermal effect of the encapsulated AuNPs. Other researchers have also created various photoresponsive hybrid microgel systems for site-specific drug release,[50,82,83] increased drug loading efficiency,[84] and wound healing through the photothermal effect.[85]

Photoresponsive microgel systems loaded with plasmonic nanoparticles and azobenzene/spiropyran based microgel systems have their own advantages and disadvantages with respect to their biomedical applications. In case of photoresponsive microgels containing azobenzene or spiropyran as the photoresponsive moiety, UV or visible light is needed to cause the transition from one conformation to another, while NIR radiation is needed to cause volume phase transition in microgels loaded with plasmonic metal nanoparticles. Volume changes in azobenzene/spiropyran based microgels are more significant upon light irradiation in comparison to plasmonic metal nanoparticle based photoresponsive hybrid microgels. However, UV/visible radiation is absorbed by the human body and may be toxic or dangerous, making UV/visible photoresponsive microgels unsuitable for biomedical applications (*i.e.*, drug delivery); in contrast, Au nanorod based hybrid microgels absorb NIR radiation to cause volume phase transition in the polymeric network, making the hybrid system suitable for drug-delivery applications.

5.1.2 Tissue engineering. Microgels and microgel-based composites have emerged as promising candidates for tissue regeneration. Their versatile chemical properties, easy synthesis and handling, biocompatibility, and cost-effectiveness make them attractive options for tissue engineering.[86] Photoresponsive microgels can create scaffolds mimicking the dynamic behavior of biological tissues. When exposed to light, these scaffolds can release growth factors and other bioactive

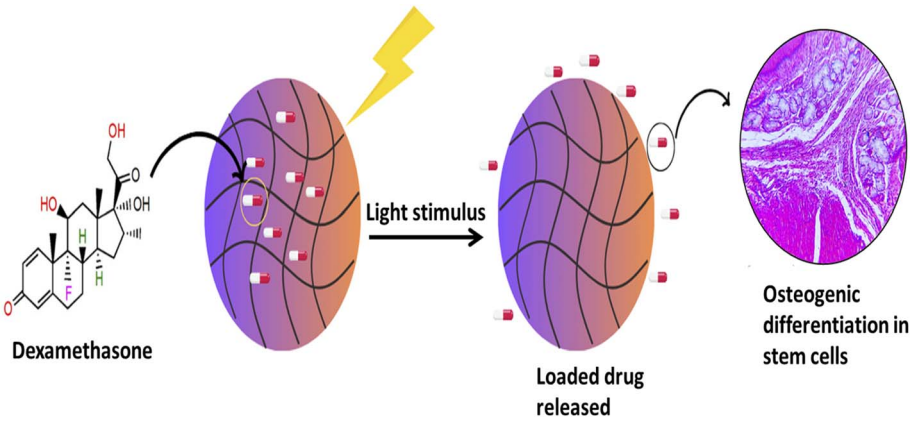


Fig. 4 Schematic illustration of the DEX release from the p(NIPAM-*co*-NBMA) microgels used to trigger stem cell bone formation.

substances in a controlled manner, promoting tissue regeneration and healing.

With the help of free radical precipitation polymerisation, Zhang *et al.*[68] created photoresponsive poly(*N*-isopropylacrylamide-*co*-nitrobenzyl methacrylate) [P(NIPAM-*co*-NBMA)] microgels. For this purpose, dexamethasone (DEX), which is a synthetic glucocorticoid osteogenic inducer, was then incorporated into the microgels through its putative hydrophobic interaction with the NBMA (nitrobenzyl methacrylate) group. LC-MS and UV-vis spectroscopy were employed to evaluate the light-triggered discharge of DEX. The possibility that the released DEX would induce osteogenic differentiation in hMSCs (human mesenchymal stem cells) was assessed. *In vitro* tests showed that the DEX released from the light-sensitive microgels may boost bone cell growth. By changing the UV light strength (*i.e.*, in on-off UV exposure tests), researchers could control stem cell transformation in bone cells. The results indicated DEX-loaded photoresponsive microgels can guide stem cell bone growth in response to light. Fig. 4 shows a schematic of the release of dexamethasone from the microgel network upon light stimulus. The released drug (DEX) was used to induce osteogenic differentiation in stem cells.

The development of responsive DNA (deoxyribonucleic acid) microstructures that can mimic living tissues is challenging due to difficulties in regulating the dimensions of DNA microstructures and integrating them with a quick and significant structural reaction. Merindol *et al.*[87] tackled this using a light-sensitive, azobenzene-containing cationic surfactant to drive light-controlled coil-globule transition in the DNA microgel network. The affinity of the azobenzene moiety for DNA was decreased by photoinduced *trans–cis* isomerization of azobenzene [Scheme 1(a)], leading to rapid and large-amplitude microgel swelling.

5.2 Soft actuators and sensors

Living things stand out because they can move on their own and control their movements. Our muscles act like motors, letting us move freely. A motor works by turning energy into motion. For tasks that need remote control, light-activated motors are useful in fields such as mechatronics, soft robotics, and microfluidic valves. However, making soft motors or tiny devices that react quickly to light is still very difficult because the functional materials need to be able to fully contract and expand reversibly.

Chen *et al.*[88] showed how to make a tiny motor using light-induced isomerization of an alkene resembling a chiral helix to change the shape of a twisted molecule. As illustrated in Fig. 5, UV light (365 nm) turns the stable form of an alkene molecule ("Stable 1") into an unstable one ("Unstable 1"). This unstable form then goes through a full cycle, releasing tension as it flips its shape when heated (50 °C). In water, these twisted molecules join up to form tiny fibers. These can be precipitated into a gel with calcium to make strings with an aligned nanofiber configuration. When lit up, these gel strings bend at 90° at a rate of 1.5° per second. This system acts as a tiny molecular motor. However, it takes about 2.7 hours for the strings to go back to their original shape, which is much slower than muscles. Furthermore, the inability of this process to repeat was ascribed to the supramolecular string's instability around 50 °C.

Significant improvements in photoresponsive behaviour were obtained by Iwaso *et al.*[89] through additional work on α-cyclodextrin (α-CD) based systems. These systems have a unique network structure where α-CD moieties are covalently bonded to amino-capped azobenzene moieties, which dimerise to form daisy-chain-type interlocked molecules. They were subsequently crosslinked to form photoactive actuators (hydrogel and xerogel systems) using a polycondensation process and succinimidyl ester capped four-arm poly(ethylene glycol). These actuators bend towards the direction of the light source in response to UV light. However, hydrogels respond at the rate of 7 degree per every three hours whereas xerogels showed a faster response rate of 7 degree per second. When the isomer of azobenzene changes from the *trans* to the *cis* form, UV irradiation (365 nm) causes the azobenzene moieties to be removed from the α-CD cavities. Light-responsive microgel actuators can be used for on-site drug delivery and release. Biocompatible and multi-stimuli-responsive functional microgels were developed and optimized by Agnihotri *et al.*[74] These multi-stimuli-responsive microgels exhibit photoactuation under different stimuli and were used

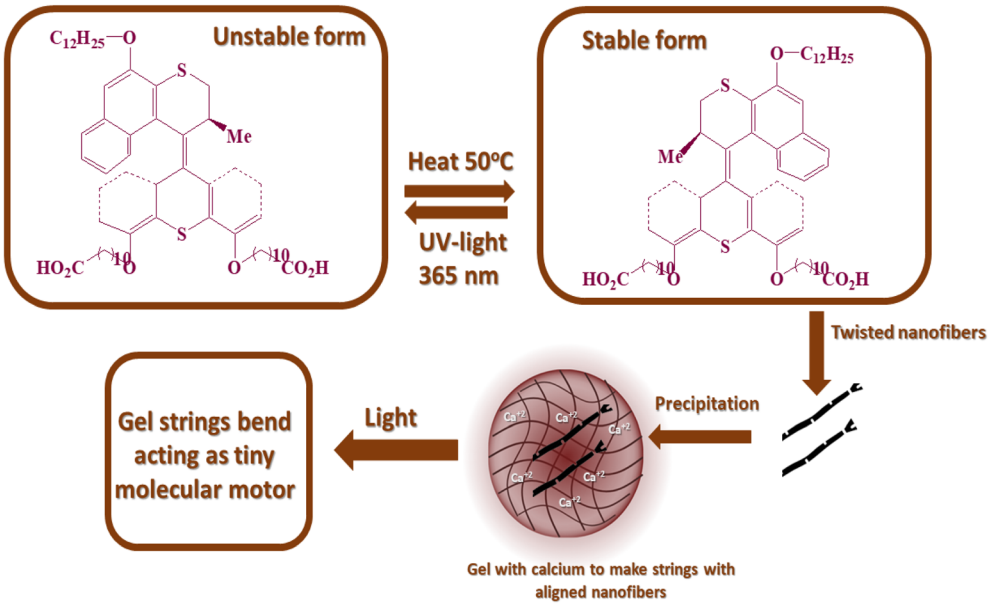


Fig. 5 Photoresponsive hydrogel actuator. The chemical equation shows the light-triggered change of the Stable 1 to Unstable 1 forms, which then reverts to Stable 1 with heat (50 °C), completing a full molecular rotation cycle. Stable 1 forms aligned nanofibers at the cm scale, allowing the molecular motion to cause macroscopic bending when exposed to UV light.

for on-site release of the antibacterial drug, ciprofloxacin. A light-sensitive hydrogel walker,[90] a thin-film sensor[69] and etalon sensors[51] have been developed thanks to the application of photothermally responsive microgels.

5.3 Catalysis

Photoresponsive hybrid microgels are attracting interest in catalysis research due to their excellent control over catalytic activity by adjusting the intensity of light of a specific wavelength. Li *et al.*[54] developed a three-component composite microgel system containing a gold nanorod core and *N*-isopropylacrylamide shell filled with silver nanoparticles [AuNR@(AgNPs/PNIPAM)]. The AuNR@AgNPs/P(NIPAM) system acted as a smart microreactor to reduce 4-nitrophenol into 4-aminophenol using $NaBH_4$ (sodium borohydride) as a reductant in aqueous medium. The reaction speed could be adjusted by changing the near-infrared (NIR) light intensity, showing the light-controlled catalytic activity of these composite microgels (Fig. 6). AuNR present in the centre of the hybrid microgel particles absorbs NIR light and converts it into heat energy by a radiation-less relaxation process, which increases the local temperature. As a result of the rise in temperature, the polymeric network becomes shrunken and the diffusion of reactant molecules (4-NP and $NaBH_4$) towards the silver nanoparticles (catalyst) through the polymeric network becomes difficult. So, the rate of reaction is decreased. However, the rate of catalytic reduction of 4-NP may be increased by decreasing the light intensity. Thus, the catalytic activity of the hybrid system is light-controllable.

In the above example, the light-absorbing species (AuNR) is not a catalyst or photocatalyst. AgNPs present in the shell are the true catalysts in the above hybrid system. Microgels may be loaded with light-absorbing material having photocatalytic activity. For example, Liu *et al.*[91] studied cadmium sulphide (CdS) quantum dots containing poly(*N*-isopropylacrylamide-*co*-acrylic acid) microgels for breaking down an organic dye present in water. The CdS quantum dots loaded in these microgels acted as a photocatalyst and could degrade Rhodamine B under UV light. This setup allowed for efficient RhB breakdown and reuse of the catalyst. The results showed that 95% of the Rhodamine B could be broken down in an hour, along with other organic compounds.

Xu *et al.*[46] developed light- and heat-responsive yolk–shell hybrid microgels with an AuNP-containing polydopamine core covered with a SiO_2 layer and a PNIPAM shell, and core–shell microgels with a polydopamine core covered with a SiO_2 layer and a PNIPAM shell containing AuNPs. The silica (SiO_2)

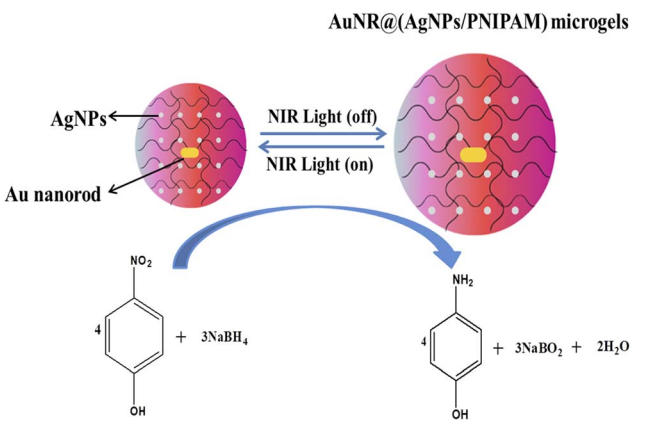


Fig. 6 Near-infrared (NIR) light adjustable catalytic activity of an AuNR@(AgNPs/PNIPAM) hybrid microgel system.

interlayer present between the core and the shell may restrict the passage of reactants to the AuNPs. To overcome these difficulties, the SiO_2 interlayer was removed at room temperature with ammonium bifluoride. The empty space between the yolk and shell facilitates diffusion of reactant molecules towards the catalytically active Au nanoparticles present in the yolk or shell. The catalytic activity of the yolk shell hybrid microgel system was tested using the catalytic reduction of 4-nitroaniline (4-NA) with $NaBH_4$ under irradiation with light (heating caused by the light) and electric heating (conventional heating). The reduction process of 4-nitroaniline to *p*-phenylenediamine under external heating was investigated in the presence and absence of light irradiation. The photothermal effect is the major driving force behind the reaction process. In this nanoreactor system, the increased reduction rate is due to PDA's outstanding NIR photothermal conversion characteristics, which caused heating of the local environment in the nanoreactor core. Under NIR irradiation, the unique AuNPs (active reaction sites) on the PDA cores directly use the local heat surrounding the PDA, which makes the surface temperature of the Au and PDA greater than that of the bulk solution. The rate of catalytic reduction of 4-NA in the presence of these hybrid systems could be increased by both light and conventional heating. Under NIR light, the nanoreactors with a yolk-loaded catalyst showed a higher reduction rate than under direct heating (Fig. 7). This enhanced activity is linked to the conversion of laser light into heat to increase the temperature in the central region where the reduction is taking place, while the temperature of the shell region remains low, and the diffusion of reactant molecules is not significantly affected by this rise in temperature. As a result, the catalytic reduction does not deviate from Arrhenius behavior. However, the catalytic reduction of 4-NA in the presence of the yolk shell hybrid microgel system, with AuNPs in the shell, deviates from Arrhenius behavior under conventional heating due to the shrinkage of the P(NIPAM) network at $T \geq 32$ °C. This work opens up new possibilities for designing smart nanoreactors and efficient light-assisted catalysis.

Another multi-responsive hybrid microgel system for light-controlled catalytic activity has also been developed.[92] In this system, the synthesis of poly(*N*-isopropylacrylamide-*co*-methacrylic acid) microgels functionalized with thiol-containing gold nanoparticles was reported. In reaction to environmental factors, such as light irradiation, temperature, and pH, the multi-stimuli-responsive hybrid microgels exhibited a clearly defined swelling/deswelling transition. Using external triggers such as temperature and pH to cause a volume phase change in hybrid microgels allows for modulation of their plasmonic characteristics. The hybrid microgel system demonstrated adjustable catalytic activity in 4-nitrophenol reduction, since the reaction rate fell sharply within a certain temperature range rather than rising monotonically with temperature. Catalytic reduction of various nitroarenes by plasmonic metal nanoparticles other than gold stabilized within the microgel systems has been widely reported by us[93–95] and others,[35] but the photoresponsive behavior of such hybrid systems and the effect of light intensity on their catalytic activity have not been described for most of the hybrid microgel systems. Therefore, discussions on catalysis by Ag nanoparticles loaded into microgel systems have not been presented in this section and may be found in reviews published in the last few years.[41,96,97]

5.4 Responsive interfaces and intelligent coatings

The boundary between a material and its surroundings is its surface. Surfaces are crucial when interfacial shape or interactions, rather than bulk properties, determine material characteristics, as seen in coatings, membranes, or heterogeneous catalysts. Beyond their chemical makeup, surfaces are defined by physical traits such as roughness, porosity, or topology. Photoresponsive surface topologies can be created using photoresponsive hydrogels. For example, Chen *et al.*[98] developed a method to covalently attach a light-sensitive spiropyran layer ($\approx$1.2 μm) to a hydrogel surface. This was done by quaternizing tertiary amino groups with iodide-functionalized spiropyran. The resulting surface was superhydrophobic due to both

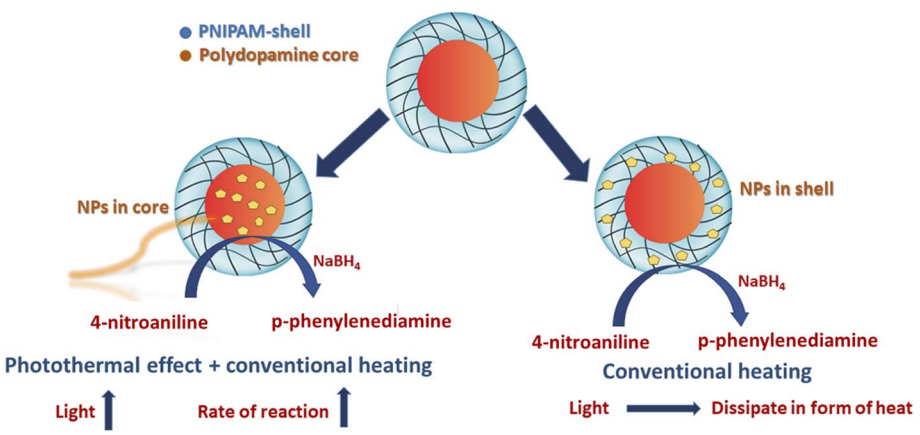


Fig. 7 Schematic illustration of the improved catalytic reduction of 4-nitroaniline into *p*-phenylenediamine by a nanoreactor with AuNPs in the core due to both conventional heating and photothermal effect compared with the nanoreactor with AuNPs in the shell due to the conventional heating.

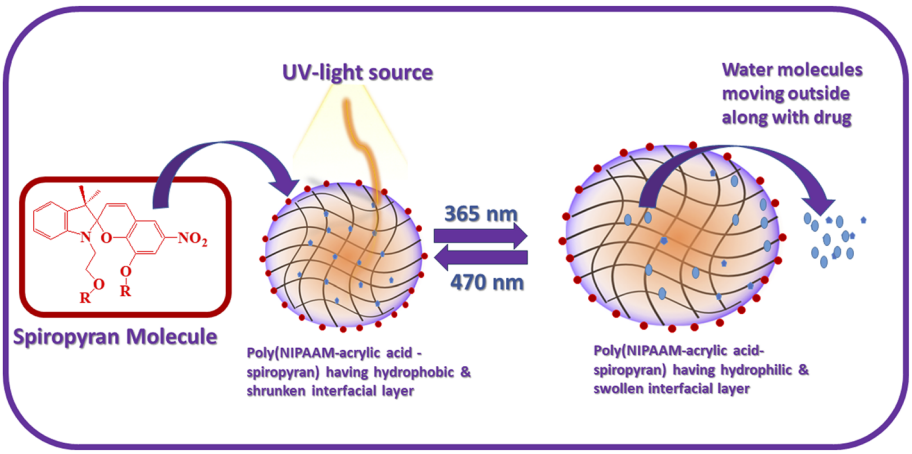


Fig. 8 Schematic illustration of light controlling the molecule movement through a hydrogel layer using the shape-changing ability of spiropyran.

spiropyran's hydrophobicity and the formed hierarchical micro/nanoscale surface texture. UV light exposure changed the spiropyran-coated layer from superhydrophobic to hydrophilic, enabling light-controlled diffusion of polar substances in and out of the hydrogel (Fig. 8). This was demonstrated by inhibiting fluorescein release from a hydrogel in water. A photoresponsive cotton fabric modified with poly(NIPAM-*co*-acrylic acid-*co*-spiropyran) hydrogel has also been developed by Schiphorst *et al.*[99]

Light can also trigger remote cell release from hydrogel surfaces through photoinduced surface-charge changes, as demonstrated by Ming *et al.*[100] Liu *et al.*[101] expanded on this concept, creating a hydrogel that not only kills bacteria with its cationic charge but also detaches them using light to prevent further adhesion. Exposing a 4,5-dimethoxy-2-nitrobenzyl group attached to a quaternary ammonium functionality to 365 nm light transforms the surface from cationic to zwitterionic. This change induces dead bacteria to detach from the hydrogel surface.

The mechanical surface properties of hydrogels can also be used to alter their biological functionality, including biophysical and bioadhesive traits. Thus, Yang *et al.*[102] utilized a photodegradable hydrogel to control surface stiffness magnitude and spatial organization, thereby directing cell fate. By manipulating these properties, researchers can fine-tune hydrogel surfaces for specific applications in biomedical and materials science fields.

Photoresponsive microgels offer innovative applications in smart coatings that change their properties when exposed to light. These versatile coatings find use in antifouling surfaces and self-cleaning materials, where microgels adjust their surface features based on environmental factors. Chen *et al.*[103] developed a photothermal hybrid microgel system based on ferric oxide@poly(*N*-isopropylacrylamide-*co*-polyacrylic acid) [Fe_3O_4@P(NIPAM-PAA)], which they sprayed onto a poly(-ethyleneimine) (PEI)-modified substrate to create a lubricating microgel coating. At room temperature, this hydrophilic coating achieved good hydration and lubrication with low friction. The photothermal characteristics of the Fe_3O_4-containing hybrid microgels in an aqueous medium were examined in the study, which found that NIR light raised the suspension temperature. The capacity of the hybrid microgel coating to regulate local temperature was demonstrated by illuminating it in both dry and aqueous conditions using an 808 nm NIR laser. It was found that illumination of the hybrid microgels with NIR light regulated the local temperature of the hybrid microgels. When exposed to NIR light, this photothermal microgel coating rapidly converted light energy into heat, thereby increasing its temperature. The thermosensitive shell composed of P(NIPAM-PAA) caused the coating's wettability to shift to hydrophobic above the lower critical solution temperature (LCST), significantly increasing friction. Essentially, NIR light could reversibly modulate the coating's surface friction. This study presents functional microgels for intelligent lubricating coatings and expands the uses of microgels in aqueous lubrication. An effective technique for building intelligent actuator systems, regulated transmission, and interfacial sensing may be offered by this foundational study in friction control. Smart microgel films were developed by Nigro *et al.*[104] by utilizing lithium fluoride (LiF). These microgel films were then utilized as smart coatings for photoluminescent solid-state radiation detectors. Effective regulation of morphological and optical characteristics is possible by this method. Moreover, by employing P(NIPAM) microgel films, this method paved the way for the creation of hybrid and perhaps biocompatible radiation detectors.

5.5 Environmental applications

For the detection and remediation of water contamination, novel approaches are provided by light-responsive microgels, a relatively young but promising field in environmental research. Their photoresponsiveness makes them ideal for jobs requiring precision and adaptability in solving environmental problems. These microgels will probably become even more

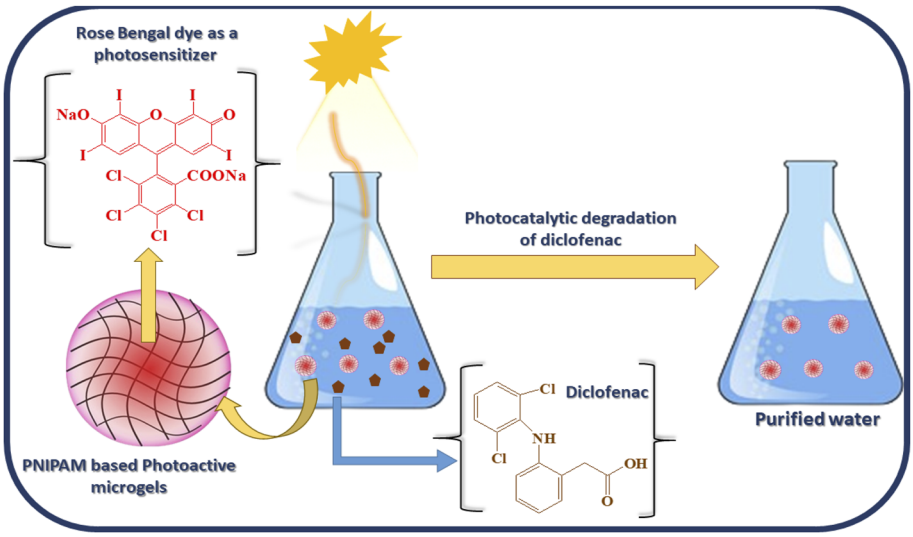


Fig. 9 Schematic illustration of the degradation of diclofenac from wastewater using Rose Bengal containing PNIPAM-based microgel as a photocatalyst.

beneficial for a variety of environmental tasks as further research is done on their fabrication and functionalization.

It is possible to design photoresponsive microgels with the ability to absorb and filter out contaminants from water. Their ability to successfully trap pollutants is enhanced by the fact that their swelling behaviour can be controlled utilizing light of certain wavelengths. For example, these microgels have the ability to create a process of capturing and releasing by expanding in the presence of light to trap pollutants and contracting in the absence of light to release them.[4] Gu *et al.*[105] made a smart enzyme machine (SEM) by putting tyrosinase (Tyr) into light-sensitive microgels using a polydopamine-assisted self-building method to clean phenol-contaminated water. Polydopamine (PDA) not only held Tyr to the gel to keep it from leaking out while still working well, but also absorbed light to make the PNIPAM gels switch between hydrophobic and hydrophilic, helping to get clean water back efficiently. Similarly, a recyclable and sustainable microgel system made of poly(*N*-isopropylacrylamide-*co*-aminoethyl methacrylate) and Rose Bengal [(P(NIPAM-*co*-AEMA)-RB)] was developed by Fabregat *et al.*,[49] which was used for the removal of emerging pollutants, such as diclofenac, from wastewater. Diclofenac is one of the commonly used pharmaceutical drugs that persist in the environment due to its low natural degradation rate and high toxicity. As illustrated in Fig. 9, the photosensitizer, Rose Bengal, was covalently linked to the pre-synthesized microgels *via* a carbodiimide coupling mechanism. The resulting microgel system, [P(NIPAM-*co*-AEMA)-RB], was used for the degradation of diclofenac to generate purified water. The microgel system fabrication based on the sustainability approach and green chemistry principles also enabled the synthesis of 5-hydroxy-2(5*H*)-furanone, an important intermediate, from photo-oxidation of furoic acid. Moreover, some photothermally responsive hybrid microgels can be used in advanced environmental cleanup plans where specific pollutant removal is needed.[106]

5.6 Microfluidic applications

Photoresponsive microgels can also serve as light-activated valves, pumps, or detectors. For example, for utilization in microfluidic devices, photoresponsive microvalves consisting of photoresponsive microgels were developed by Jadhav *et al.*[107] These microvalves are flexible and can be easily installed on lab-on-chip devices. When NIR radiation hit the valves made of microgels consisting of PNIPAM (thermosensitive polymer) and polypyrrole nanoparticles (photothermally active), the shrinking and swelling of the microgels in response to the NIR light caused the opening and closing of the valves, respectively. Depending on the intensity of the NIR light and the duration of exposure, the valves show a swift response, thus allowing precise control of fluid flow. Fast fluidic switching applications are now possible with this optically controlled hydrogel microvalve. Similarly, Sershen *et al.*[108] suggested light-irradiated microvalves based on the volume change of nanocomposite hydrogels made of thermoresponsive polymer PNIPAM gels with strongly optically absorbing Au nanoparticles. The gels were created within specific regions of microchannels using *in situ* photo-polymerization to create nanocomposite hydrogel microvalves. The microvalves were opened with laser light irradiation as powerful as 1600–2700 mW cm^{-2}. Au nanoparticles converted the high optical energy into heat, which caused the thermoresponsive P(NIPAM) gels to shrink and caused the microvalves to open. The approach involved irradiating the whole device with laser light of varying wavelengths to regulate the two microvalves, each of which had a distinct absorption spectrum. More details on the utilization of photoresponsive microgels as valves and mixers, and their use inside microfluidic devices, can be found in a review.[109]

5.7 Optical properties-based applications

Photoresponsive microgels are promising materials for various optical uses. These applications are wide-ranging and

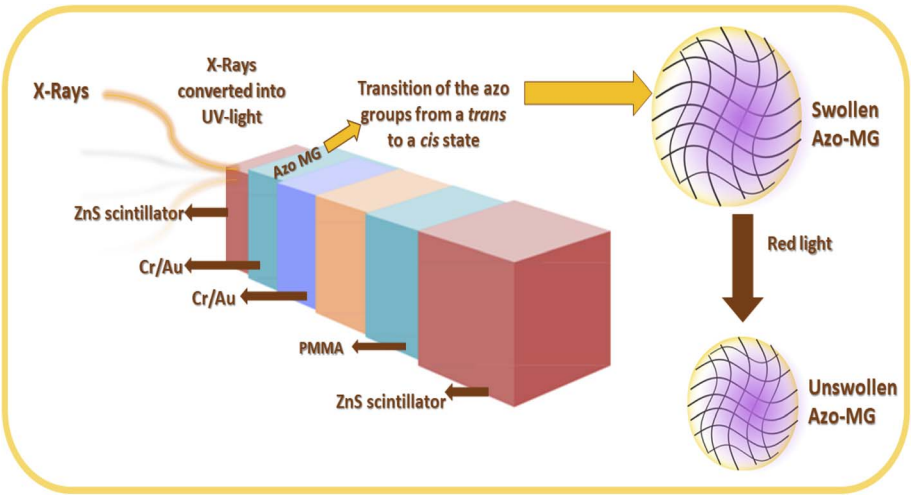


Fig. 10 Schematic of the conversion of X-rays into UV rays by a ZnS layer, causing the isomerization of azobenzene, which, in turn, causes swelling of the microgels and a red shift in the optical signal of the interferometer.

significant, covering areas from sensing technology to medical devices, with ongoing studies promising even more advanced uses in the future. Therefore, hybrid microgels with the tunable optical properties of plasmonic metal nanoparticles have been developed for a variety of applications. For example, Wu *et al.*[55] fabricated Ag nanoparticles and Ag–Au bimetallic nanoparticles within poly(*N*-isopropylacrylamide-*co*-acrylic acid-*co*-acrylamide) microgels and reported that the Ag photoluminescence intensity can be increased and the wavelength of the maximum emission can be shifted to a lower wavelength *via* pH-induced shrinkage of the polymeric network. The surface modification of Ag nanoparticles with Au nanoclusters can also enhance the intensity of the photoluminescence signal in the NIR region for potential use in the biomedical field. Similarly, the red shift in surface plasmon resonance wavelength (λ_{SPR}) of Ag–Au bimetallic nanoparticles from 446 nm to 452 nm by changing the temperature of the medium from 20 °C to 50 °C has been reported by Li *et al.*,[53] which may be attributed to a change in the refractive index of the medium around Ag–Au nanoparticles due to the shrinkage of the *N*-isopropylacrylamide segment of the microgel system. A similar trend in the λ_{SPR} value of Ag nanoparticles loaded into microgels made of an interpenetrating network of poly(acrylic acid) and crosslinked poly(*N*-isopropylacrylamide) with an increase in temperature has been noted by Liu *et al.*[56]

An etalon system based on poly(*N*-isopropylacrylamide) microgels was created by Gao *et al.*[110] that changed color when exposed to light. These systems were made by adding pH-sensitive microgel-based etalons to a solution of the photoacid *o*-nitrobenzaldehyde (*o*-NBA). When exposed to ultraviolet (UV) light, the photoacid released a proton, lowering the solution's pH. As the P(NIPAM) microgel-based etalon was pH-sensitive, it changed its optical properties and thus its visible color. These etalons only changed color in pH-responsive areas, creating patterns that shift color under UV light. The etalon's color was shown to be fully reversible and could be switched multiple times. These unique systems could be used for displays and controlled drug delivery. Zhang *et al.*[30] used 4,4′-di(acrylamido)-azobenzene as a crosslinker in the fabrication of poly(*N*-isopropylacrylamide)-based microgels to create microgel-based optical materials (etalons).

Wei *et al.*[70] developed Au nanoparticle (AuNP) cores coated with a poly(*N*-isopropylacrylamide) (PNIPAM) shell [(Au@pNIPAM)] using seed-mediated free radical precipitation polymerization. They then created a temperature-photoresponsive photonic device by placing Au@PNIPAM particles between two thin Au layers. This device showed visual color changes and unique multipeak reflectance spectra, with peak positions mainly determined by the distance between the Au layers. The dual responsiveness of the device comes from combining the AuNPs core's photothermal effect (localized surface plasmon resonance) and the PNIPAM shell's temperature sensitivity. These uniform Au@PNIPAM microgel-based devices respond quickly and show consistent spectra across the slide. Potential uses include drug delivery, active optics, and soft robotics.

P(NIPAM)-based microgels containing the azo group were synthesized by Wang *et al.*[111] These microgels were used to construct interferometers (ZnS/Au-azo microgels-Au multilayers). X-rays are converted into UV rays by a ZnS (scintillator) layer, which causes the *trans* to *cis* transition in the azo group, causing the microgels to swell, which, in turn, causes a red shift in the UV-visible signal of the interferometer due to the increase in distance between the two Au layers (Fig. 10). The designed interferometer has potential applications in the areas of radiobiology, actuation in space, and chemoradiotherapy.

Zhang *et al.*[112] synthesized new multiresponsive microgels based on poly(*N*-isopropylacrylamide) containing triphenylmethane leucohydroxide (TPL) to build etalons. This study highlights the adaptability of microgel-based etalon structures and demonstrates their potential for remote activation, color-tunable ics, sensing, and possible remotely triggered drug-delivery applications.

6 Conclusion and future directions

In this review, synthesis methods, mechanism of photoresponsiveness, characterization, and applications of photoresponsive microgels have been outlined using data published in recent literature. To sum up, photoresponsive microgels are promising smart materials with many potential uses in fields including medicine, sensing, catalysis, and light-based electronics. Being able to control changes in the microgel size with light opens doors for creating advanced materials. However, more research is needed to fully grasp how their structure relates to their properties and to find more ways to use these interesting materials.

As light-sensitive microgels improve, new paths open up. Making microgels with uneven shapes could boost their surface features. Studies to make microgels responsive towards multiple triggers, such as heat and pH, in addition to photosensitivity, may be interesting. Mixing these microgels with components such as nanoparticles or enzymes, building them into larger structures like crystal-like formations, and using them in medicine are exciting areas to explore. Researchers are showing increasing interest in creating safe, biodegradable microgels that respond to radiation for deep use in different applications. Complete biodegradable and biocompatible photoresponsive microgels with medically approved components and injectable formulations may be developed for biomedical applications in the future. Most of the work reported so far in this area is based on the azobenzene-based photoresponsive organic components of microgels. Photoresponsive organic species other than azobenzene and its derivatives may be explored for the fabrication of light-responsive microgels. Hybrid microgels with light-tunable catalytic activity are usually based on Au and Ag nanoparticles. The combination of plasmonic metal nanoparticles with non-plasmonic, inexpensive transition-metal nanoparticles stabilized in polymer microgels may be interesting for sustainable catalysis in the future.

Conflicts of interest

The authors declare no conflicts of interest.

List of abbreviations

AA	Acrylic acid
AAm	Acrylamide
AAS	Atomic absorption spectroscopy
ABSA	4-[(4-Methacryloyloxy)phenylazo] benzenesulfonic acid
AdoB 12	Adenosylcobalamin
AEMA	Aminoethyl methacrylate
AgNPs	Silver nanoparticles
ALA	Allylamine
α-CD	alpha-Cyclodextrin
AuNPs	Gold nanoparticles
AuNRs	Gold nanorods
Azo-MGs	Azobenzene microgels
BIS	*N*,*N*′-Methylenebisacrylamide
CarHC	C-terminal adenosylcobalamin binding domain
CdS	Cadmium sulfide
CD	Cyclodextrin
DAAB	4,4′-Di(acrylamide)-azobenzene
DASA	Donor acceptor Stenhouse adduct
DEX	Dexamethasone
DLS	Dynamic light scattering
DMNPB	3-(4,5-Dimethoxy-2-nitrophenyl)-2-butyl ester
DNA	Deoxyribonucleic acid
DNQ	2-Diazo-1,2-naphthoquinone
DOX	Doxorubicin hydrochloride
DTA	Differential thermal analysis
ECM	Extracellular matrix
EM	Electrophoretic mobility
FA	Folic acid
FA-PNFA	Poly(NIPAM-*co*-functional palygorskite-Au-*co*-acrylic acid)
Fe_3O_4	Ferric oxide
FM	Fluorescence microscopy
FTIR	Fourier-transformed infrared spectroscopy
GMA	Glycidyl methacrylate
GPC	Gel permeation chromatography
hMSCs	Human mesenchymal stem cells
LCMS	Liquid chromatography-mass spectrometry
LCST	Lower critical solution temperature
LiF	Lithium fluoride
MAPTMS	Methacryloxypro-pyltrimethoxysilane
MG	Malachite green
mPEG	Mercapto-modified polyethylene glycol
4-NA	4-Nitroaniline
$NaBH_4$	Sodium borohydride
NBMA	Nitrobenzyl methacrylate
NIPAM	*N*-Isopropylacrylamide
NIR	Near-infrared
NMR	Nuclear magnetic resonance
NPs	Nanoparticles
ONB	*ortho*-Nitobenzene
o-NBA	*o*-Nitrobenzaldehyde
PAH	Polyallylamine
PAA	Polyacrylic acid
(PNIPAM-*co*-NBMA)	Poly(*N*-isopropylacrylamide-*co*-nitrobenzyl methacrylate)
PCS	Photon correlation spectroscopy
PDA	Polydopamine
PEG	Polyethylene glycol
PEI	Poly(ethyleneimine)
Poly(MMA)	Poly(methyl methacrylate)
PMOXA	Poly(2-methyl-2-oxazoline)
PNFA	Poly(*N*-isopropylacrylamide-*co*-palygorskite-Au-*co*-acrylic acid)
P(NIPAM)	Poly(*N*-isopropylacrylamide)
p-NP	*para*-Nitrophenol
PS	Polystyrene
P(VCL)	Poly(vinyl caprolactam)
RAFT	Reversible addition fragment chain transfer
RhB	Rhodamine B
RB	Rose Bengal

RS	Raman spectroscopy
RS	Reflectance spectroscopy
SEM	Scanning electron microscopy
SER	Smart enzyme reactor
SFEP	Surfactant-free emulsion polymerization
SiO_2	Silicon dioxide
SP	Spiropyran
STEM	Scanning transmission electron microscopy
SWNT	Single-walled nanotubes
TEM	Transmission electron microscopy
TGA	Thermal gravimetric analysis
TPL	Triphenylmethane leucohydroxide
Tyr	Tyrosinase
UV	Ultraviolet
UV-Vis	Ultraviolet visible spectroscopy
VCL	Vinylcaprolactam
VPT	Volume phase transition
VPTT	Volume phase transition temperature
ZnS	Zinc sulphide

Data availability

No data was used in this article.

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