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Core-Shell Nanoparticles with Hydrophobic Shells

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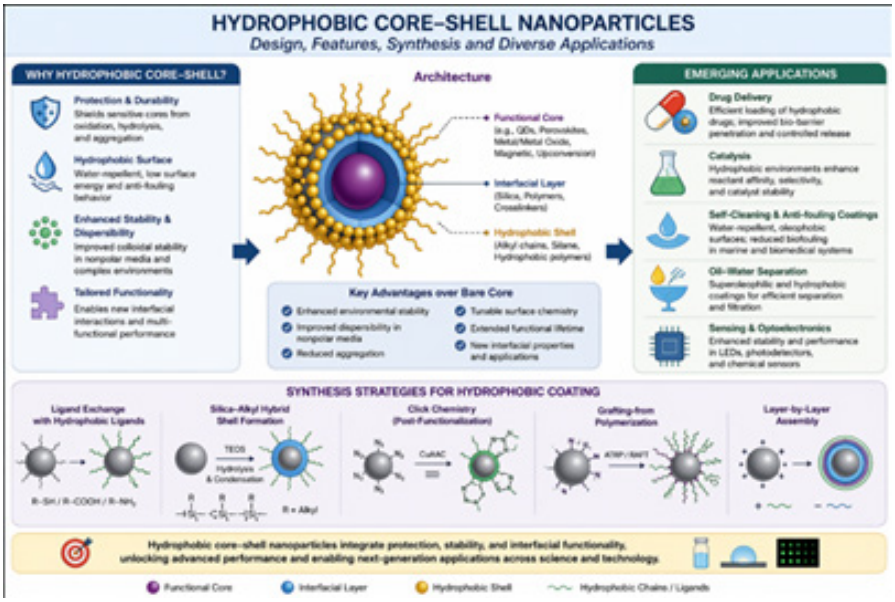
ABSTRACT

Hydrophobic core-shell nanoparticles have emerged as a versatile class of nanomaterials in which a functional core is combined with a hydrophobic outer shell to achieve enhanced performance beyond that of uncoated nanoparticles. The hydrophobic shell not only protects sensitive cores from oxidation, hydrolysis and aggregation, but also tailors interfacial behavior, colloidal stability and compatibility with nonpolar environments. These features enable advanced applications in drug delivery, catalysis, self-cleaning and anti-fouling coatings, oil–water separation, sensing and optoelectronics. This review summarizes the fundamental advantages of hydrophobic core–shell architectures, compares their properties with bare core nanoparticles and discusses exclusive functionalities arising from surface hydrophobization. Particular attention is given to nanoscale fabrication strategies, including silica-alkyl hybrid shell formation, ligand engineering, click chemistry and grafting-from polymerization, which allow precise control of shell thickness, chemistry and durability. Overall, hydrophobic core-shell nanoparticles represent an important platform for next-generation multifunctional materials.

Keywords: Superhydrophobicity; Nanoparticles; Coreshell particles



Introduction

Core–shell nanoparticles can be defined as versatile class of nanomaterials in which a functional core is encapsulated within a protective outer layer. These particles display a special blend of surface-dominated water-repelling behaviour and core-driven functionalities when the shell is hydrophobic. With few nanometres thick the hydrophobic coating at nanoscale level has potential to influence colloidal stability, interfacial activity and environmental robustness of the system. Such design offers opportunity of the integrating multiple properties into a single nanoscale platform, making these materials highly attractive for applications across catalysis, environmental remediation, drug delivery and energy harvesting.

Metals, metal oxides, semiconductors or carbon-based nanostructures can make up the core of these nanoparticles and each of these materials can play a unique functional role. For example, iron oxide and cobalt ferrite nanocrystals are highly sought after for their superparamagnetic behaviour in biomedical imaging and targeted therapies, while gold, silver and platinum nanoparticles offer catalytic and plasmonic activity. Semiconductor cores such as TiO_2, ZnO and quantum dots exhibit photocatalytic and optoelectronic characteristics, whereas carbon nanostructures, including graphene quantum dots and nanodiamonds, impart electrical conductivity along with chemical resilience. Therefore, the desired use guides the choice of core material, while the hydrophobic shell extends functionality and durability in challenging situations.

Primarily ligand-based surface modifications, hydrophobic polymers or silane coupling agents can be used to engineer the hydrophobic shell. Silane-derived coatings, such as octadecyltrichlorosilane or fluoroalkylsilanes, provide strong chemical resistance and long-term stability. Polymeric shells including polydimethylsiloxane (PDMS), polystyrene and fluoropolymers allow for tunable shell thickness and flexibility. In metallic nanoparticle systems, self-assembled monolayers of alkyl-thiols has ability to impart hydrophobicity while preserving the core's catalytic or optical properties. Hybrid approaches, such alkyl-modified silica shells, improve mechanical robustness and immunity to water degradation by combining stiffness and hydrophobic surface chemistry.

The existence of a hydrophobic shell at the nanoscale results in special physicochemical behaviour. These nanoparticles exhibit strong interfacial activity, often confining to oil–water interfaces enacting as stabilizers or destabilizers of emulsions in Pickering-type systems. Additionally, the hydrophobic coating prolongs the operational lifetimes of sensitive cores like quantum dots or perovskite nanocrystals by protecting them from hydrolytic destruction. In some cases, hydrophobic shells can enact stimuli-responsive allow for controlled release or exposure of the core's functionality, adding a dynamic dimension to the material's performance. Such control is particularly valuable in biomedical and catalytic contexts, where selective activation is desired.

The combination of hydrophobic shells and functional cores has produced a wide range of applications. In environmental engineering, these nanoparticles are employed for oil–water separation and wastewater purification, as their hydrophobic surfaces reject water while the core selectively adsorbs or catalyzes contaminants. In the biomedical field, hydrophobic coatings improve the stability of nanoparticles in physiological fluids and enable the loading of hydrophobic drugs, facilitating targeted delivery systems. In energy research, hydrophobic core–shell nanoparticles have been incorporated into triboelectric nanogenerators, considering humid situations, the shell improves charge retention. Similarly, in catalytic reactions conducted in biphasic systems, hydrophobic shells allow the active cores to remain accessible in the organic phase while preventing deactivation in aqueous environments.

Synthesis of hydrophobic core–shell nanoparticles can be achieved through a variety of techniques tailored to the material system. Functional nanocrystals are frequently encapsulated into hydrophobic polymers using miniemulsion and seeded emulsion polymerisation techniques. Sol–gel methods provide a route to fabricate silica-based shells, which can subsequently be modified with alkyl groups to introduce water repellency. Ligand exchange processes enable the replacement of hydrophilic stabilizers with hydrophobic counterparts, particularly in metallic and semiconductor nanoparticles. Furthermore, "grafting-from" or surface-initiated polymerisation techniques enable the direct development of hydrophobic polymer brushes from the nanoparticle surface, providing exact control over surface chemistry and shell thickness.

Taken together, core–shell nanoparticles with hydrophobic shells constitute an emerging platform for multifunctional materials at the nanoscale. Their design brings together the intrinsic properties of the functional core with the protective and interfacial advantages of a hydrophobic coating. As research advances, these systems are expected to play an increasingly important role in bridging nanotechnology with applications in catalysis, biomedicine, environmental science and energy technologies.

In this review, we focus on the existing class of hydrophobic core–shell nanoparticles, examining their diverse applications where the hydrophobic shell plays a decisive role. Particular interest is paid to how the addition of a hydrophobic shell confers functions that are different from those of bare core nanoparticles, enabling unique and advanced applications in fields like environmental remediation, drug delivery, catalysis and self-cleaning coatings. We further highlight the unique features of these core–shell systems, discuss their ability to carve out new technological niches and outline the synthesis techniques by which nanoscale cores are coated with hydrophobic entities. This review's overall objective is to give readers a comprehensive understanding of the design approaches, application advantages, along with potential uses of hydrophobic core-shell nanoparticles.

Metallic nanoparticles (Au, Ag, Pt, Pd) for catalysis, plasmonics, sensing applications

Hydrophobic metallic nanoparticles in catalysis: Metallic nanoparticles such as gold (Au)[1], silver (Ag)[2], platinum (Pt)[3] and palladium (Pd)[4] are widely recognized for their superior catalytic[5] activity surfacing from their high surface-to-volume ratio[6] and tunable electronic properties[7,8]. At the nanoscale, the surface atoms of these metals act as highly active catalytic centers, capable of driving chemical transformations ranging from hydrogenation[9] and oxidation[10] to carbon-carbon coupling[11] reactions. However, the catalytic efficiency of these nanoparticles is not solely determined by the intrinsic properties of the metallic core; the surface chemistry[12] particularly

adsorption[13] based catalysis can be manipulated[14]. Here comes the role of hydrophobicity[15], plays a crucial role in controlling access of reactants[16], product desorption[17] and stability[18] under reaction conditions.

Hydrophobic modification of metallic nanoparticles introduces a selective interfacial environment that enhances catalysis, especially in biphasic[19] systems where immiscibility between aqueous[20] and organic phases[21] poses a challenge. For instance, Au and Pd nanoparticles coated with hydrophobic ligands[22] or shells preferentially delimiting at the oil-water interface[23], acting as catalytic nanoreactors[24] that efficiently shuttle reactants from the organic phase into proximity with catalytic sites. This not only improves mass transfer but also prevents deactivation of the metallic surface by water or polar impurities. In many oxidation and reduction reactions[25], the hydrophobic surface further aids in stabilizing nonpolar reactants[26], thus increasing the reaction rate and selectivity[27].

Moreover, hydrophobicity contributes significantly to catalyst stability. Bare metallic nanoparticles often undergo aggregation, oxidation or leaching in aqueous conditions[28], leading to rapid deactivation[29]. A hydrophobic coating, such as alkylsilane-modified shells or fluorinated polymers[30], protects the metallic core from direct exposure to water and corrosive agents, thereby extending catalyst lifetime[31]. In addition, in reactions involving volatile organic compounds or hydrophobic substrates, the hydrophobic shell provides an affinity-driven adsorption effect, concentrating reactants near the catalytic surface[32]. This hydrophobic enrichment effect[33] has been reported to dramatically improve turnover frequency[34] in Pd-catalysed cross-coupling and Pt-catalysed hydrogenation reactions35. Overview of hydrophobisation of metallic nanoparticles are shown in **(Figure 1)**.

Plasmonic metallic nanoparticles and hydrophobicity: Metallic nanoparticles such as Au, silver (Ag)[36], platinum (Pt) and palladium (Pd)[37] are well known for their plasmonic properties, which arise from the collective oscillation of conduction electrons when excited by light at specific frequencies[38]. This localized surface plasmon resonance (LSPR)[39] effect leads to strong light absorption and scattering, making these nanoparticles highly useful in sensing[40], imaging[41], catalysis and photothermal[42] applications. The optical response is highly sensitive to the surrounding medium, particle size, shape and surface chemistry, which makes careful surface engineering crucial.

Introducing a hydrophobic shell around plasmonic nanoparticles provides several important advantages. First, hydrophobic coatings prevent direct contact of the metallic core with water or aqueous electrolytes, which otherwise lead to oxidation, corrosion or aggregation, particularly in the case of Ag and Pd nanoparticles. The hydrophobic layer therefore ensures long-term stability and preserves the plasmonic response[43] in harsh environments. Second, hydrophobic shells tune the dielectric environment around the nanoparticle[44], shifting the LSPR band and allowing better control over light-matter interactions[45]. This is especially useful in applications such as surface-enhanced Raman scattering (SERS) and biosensing, where signal intensity depends strongly on local refractive index changes[46].

Moreover, hydrophobic plasmonic nanoparticles display unique interfacial activity. Due to their non-wetting shells, they preferentially localize at oil-water or air-water interfaces, which enhances light–interface interactions and makes them valuable in plasmon-enhanced photocatalysis, emulsion stabilization and interfacial sensing[47]. In addition, the hydrophobic encapsulation allows their integration into organic solvents, polymers or hydrophobic matrices, expanding their applicability in flexible electronics, optoelectronic coatings and photonic devices. In biomedical contexts[48], hydrophobic surface modification can also serve as a platform for further ligand exchange or amphiphilic encapsulation, improving nanoparticle transport across lipid membranes and enabling targeted plasmonic therapies[49].

Thus, while the metallic core provides the fundamental plasmonic activity, the hydrophobic shell plays a decisive role in ensuring stability, environmental resistance and enhanced functionality. This synergy between plasmonic metallic cores and hydrophobic shells underpins the development of next-generation plasmonic nanomaterials for sensing, catalysis and energy conversion.

Metallic nanoparticles in sensing and the role of hydrophobicity: Metallic nanoparticles such as gold (Au), silver (Ag), platinum (Pt) and palladium (Pd) have emerged as highly versatile platforms for nanoscale sensing owing to their unique surface plasmon resonance (SPR), catalytic activity and electronic conductivity. Their nanoscale size provides a large surface-to-volume ratio, which makes them extremely sensitive to changes in the local chemical environment. For example, gold and silver nanoparticles exhibit strong SPR shifts in response to molecular adsorption on their surface, while Pt and Pd nanoparticles are well known for their high catalytic efficiency in redox reactions that can be transduced into detectable signals[50]. These properties allow metallic nanoparticles to function as optical, electrochemical or catalytic sensors for a wide variety of analytes, including biomolecules, toxic gases, heavy metals and environmental pollutants[51].

In this context, hydrophobic surface modification plays a critical role in expanding the functional scope of these metallic nanoparticles. Hydrophobic shells or ligands not only prevent aggregation in aqueous systems but also make the nanoparticles more selective toward hydrophobic analytes such as organic pollutants, oil-phase contaminants and nonpolar biomolecules. For instance, Au[52] or Ag[53] nanoparticles coated with long-chain alkyl thiols exhibit enhanced affinity toward hydrocarbons or aromatic compounds, allowing highly sensitive detection in biphasic or non-aqueous environments where conventional hydrophilic sensors fail[54]. Similarly, Pd[55] and Pt[56] nanoparticles with hydrophobic coatings can act as interfacial sensors at oil-water boundaries, where their catalytic or electrochemical response is triggered only when target molecules partition into the hydrophobic phase[57].

Another advantage of hydrophobicity in metallic nanoparticle-based sensing is the improvement of anti-fouling properties. In biosensing, hydrophobic shells can reduce nonspecific adsorption of proteins or biomolecules, thereby increasing sensor selectivity and stability under complex biological conditions[58]. Moreover, in environmental sensing, hydrophobic modifications help the nanoparticles remain stable in humid or aqueous environments by creating a barrier that minimizes oxidation, corrosion or unwanted adsorption of polar impurities[59]. In triboelectric[60] or electrochemical sensing[61]

platforms, hydrophobicity additionally assists in maintaining charge retention and stability under high-moisture conditions, which is often a major limitation of unmodified metallic nanoparticles.

Thus, by combining the intrinsic sensitivity of metallic nanoparticles with hydrophobic surface engineering, it becomes possible to design robust, selective and stable nano sensors capable of operating across aqueous organic and interfacial environments. This makes hydrophobic core-shell metallic nanoparticles particularly attractive for environmental monitoring, biomedical diagnostics and industrial process control, where analytes often exist in heterogeneous or moisture-rich conditions.

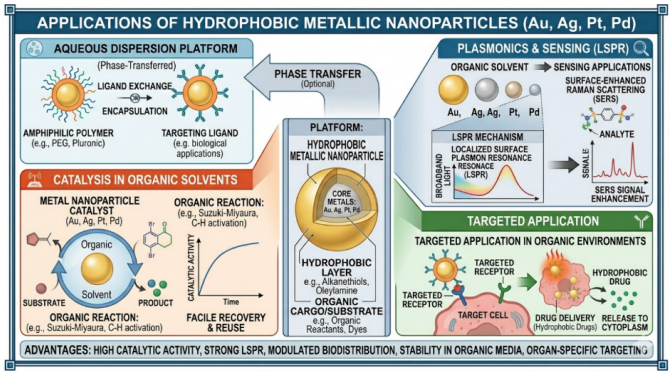


Figure 1: Overview of metallic nanoparticles being hydrophobized.

Magnetic nanoparticles (Fe_3O_4, $CoFe_2O_4$): MRI contrast, magnetic hyperthermia, targeted delivery

Magnetic nanoparticles for MRI contrast: Magnetic nanoparticles such as Fe_3O_4 (magnetite)[62] and $CoFe_2O_4$ (cobalt ferrite)[63] have gained significant attention as magnetic resonance imaging (MRI) contrast agents owing to their strong superparamagnetic properties at the nanoscale[63]. These particles are capable of shortening the transverse relaxation time (T_2)[64] of protons in water, thereby enhancing image contrast in MRI scans. Their small size (typically 5-20 nm) ensures high surface-to-volume ratios, which in turn provides strong magnetic responsiveness even under low external magnetic fields[65]. Compared to traditional gadolinium-based contrast agents, iron oxide nanoparticles are considered safer and more biocompatible, while cobalt ferrite offers higher magneto crystalline anisotropy and thermal stability, making it suitable for high-performance biomedical imaging applications[66].

The hydrophobicity of the nanoparticle surface plays a crucial role in determining their stability, dispersibility and in vivo performance. Bare Fe_3O_4 and $CoFe_2O_4$ nanoparticles are inherently prone to aggregation[67] in aqueous media due to van der Waals and magnetic dipole-dipole interactions, which reduces their efficiency as contrast agents[68]. By introducing a hydrophobic shell, such as alkylsilane-modified silica, polydimethylsiloxane (PDMS) or fluoropolymer coatings, the nanoparticles can be effectively shielded from direct contact with water, thus preventing uncontrolled agglomeration[69]. This hydrophobic barrier also protects the magnetic core from oxidation or dissolution in physiological environments, preserving its magnetic properties over time[70].

Moreover, hydrophobic surface functionalization enables the encapsulation of nanoparticles into hydrophobic polymer matrices or micelles, which can be further modified with amphiphilic ligands or surfactants to form stable aqueous dispersions[71]. This dual functionality ensures that while the core maintains its magnetic contrast capabilities, the hydrophobic shell contributes to long-term colloidal stability, enhanced circulation time and reduced non-specific interactions with biomolecules in the body. In advanced designs, hydrophobic coatings also allow the co-loading of hydrophobic drugs, making the nanoparticle system suitable for MRI-guided theranostics, where imaging and drug delivery are combined in a single platform[72].

Magnetic nanoparticles for magnetic hyperthermia: Magnetic nanoparticles such as Fe_3O_4 and $CoFe_2O_4$73 are widely studied for biomedical and energy-related applications due to their strong superparamagnetic behavior at the nanoscale[74]. One of their most important functions lies in magnetic hyperthermia, where these nanoparticles generate localized heat when exposed to an alternating magnetic field. This heat can be harnessed to kill cancer cells, enhance drug release or trigger other therapeutic responses[75]. The efficiency of magnetic hyperthermia depends not only on the intrinsic properties of the core-such as magnetic anisotropy, saturation magnetization and particle size-but also on the surface environment that surrounds the nanoparticles[76].

Hydrophobic surface coatings play a crucial role in controlling the performance of magnetic nanoparticles in hyperthermia[77]. By applying a hydrophobic shell, the nanoparticles become less prone to aggregation in aqueous media, thereby maintaining their superparamagnetic nature and enhancing their heating efficiency[78]. Hydrophobic shells also provide a barrier against oxidation and hydrolysis of the iron oxide or cobalt ferrite core, significantly improving long-term stability under physiological or humid conditions[79]. Furthermore, in biomedical contexts, hydrophobicity can be engineered to regulate interactions with lipid membranes, promoting selective adsorption, penetration or accumulation in hydrophobic cellular regions. This feature is particularly useful when designing nanoparticles for targeted hyperthermia in tumor tissues, which often have hydrophobic microenvironments[80].

Another functional advantage of hydrophobic shells in magnetic hyperthermia is the potential for dual functionality. The hydrophobic surface can serve as a reservoir for poorly water-soluble chemotherapeutic agents, allowing simultaneous drug delivery and localized heat generation upon magnetic stimulation[80]. This combination provides a synergistic effect: the heat enhances drug diffusion and tumor penetration, while the hydrophobic environment prevents premature drug release. Thus, hydrophobic coatings are not merely protective layers; they are functional design elements that enhance the therapeutic efficacy, stability and tunability of magnetic nanoparticles in hyperthermia applications.

Magnetic nanoparticles for targeted delivery systems: Magnetic nanoparticles, particularly iron oxide (Fe_3O_4) and cobalt ferrite ($CoFe_2O_4$), have gained extensive attention in targeted delivery systems due to their superparamagnetic behavior and ability to be manipulated under an external magnetic field[81]. Their small size, high surface-to-volume ratio and strong responsiveness to magnetic guidance make them promising carriers for therapeutic and diagnostic applications[82]. By functionalizing these nanoparticles with suitable surface coatings, they can be directed precisely to disease sites, such

as tumors or inflamed tissues, reducing off-target effects and improving therapeutic efficacy[83].

Hydrophobic surface modification plays a crucial role in enhancing the performance of magnetic nanoparticles in targeted delivery. A hydrophobic shell coating provides stability in biological environments by preventing rapid clearance or degradation in aqueous media[84]. Moreover, hydrophobic interactions enable the encapsulation or adsorption of poorly water-soluble drugs onto the nanoparticle surface, facilitating efficient drug loading. Once delivered to the target site, external stimuli such as pH changes, enzymatic activity or magnetic hyperthermia can trigger the release of the encapsulated cargo[85].

Additionally, hydrophobicity improves the nanoparticles' ability to interact with lipid-rich biological membranes, thereby promoting cellular uptake. This feature is particularly valuable in overcoming biological barriers, such as the blood–brain barrier or cellular lipid bilayers, which are inherently hydrophobic in nature. In this way, a carefully engineered hydrophobic shell not only protects the magnetic core but also enhances the bioavailability and delivery efficiency of therapeutic molecules[86]. Thus, combining magnetic responsiveness with controlled hydrophobicity makes Fe_3O_4 and $CoFe_2O_4$ nanoparticles powerful tools for next-generation targeted drug delivery systems. Overview of hydrophobisation of magnetic nanoparticles are shown in **(Figure 2)**.

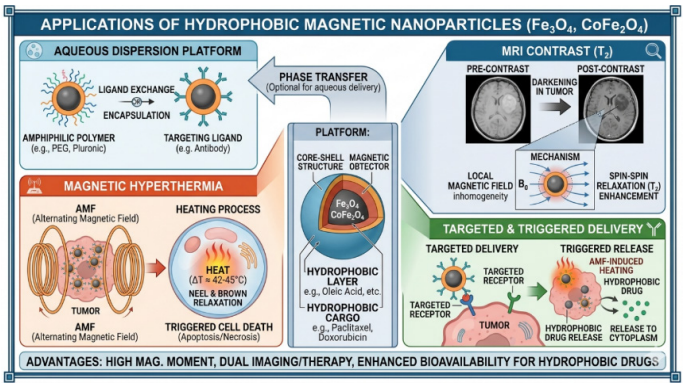


Figure 2: Overview of magnetic nanoparticles hydrophobisation.

Semiconductors (CdSe, ZnO, TiO_2, perovskites): Photocatalysis, light-harvesting, quantum dots.

SC for hydrophobic photocatalysis: Semiconductor nanoparticles such as CdSe quantum dots[87], ZnO nanocrystals[88], TiO_2 nanoparticles[89] and perovskite nanostructures[90] are widely studied for their superior optical and electronic properties, making them highly efficient photocatalysts. Their ability to absorb light and generate electron-hole pairs enable catalytic degradation of pollutants, hydrogen evolution, CO_2 reduction and solar-driven organic transformations. However, when these nanomaterials are used directly in aqueous systems, several challenges arise. They often suffer from photocorrosion, hydrolysis and surface recombination of charge carriers due to direct contact with water and reactive species[91]. In the case of perovskites, moisture sensitivity is a critical limitation that severely reduces long-term stability[92].

To overcome these issues, hydrophobic shell coatings around the semiconductor core play a crucial role. By encapsulating photocatalytic nanoparticles within a hydrophobic layer (such as alkylsilane-modified silica, fluoropolymers or hydrophobic ligands), the surface is shielded from water penetration, which suppresses degradation and enhances durability under operating conditions[93]. Moreover, the hydrophobic shell can control the interfacial interaction with reactants, selectively allowing organic molecules from an oil phase to interact with the photocatalytic core while repelling water molecules[94]. This phase-selective photocatalysis is particularly advantageous for oil-water separation systems, pollutant degradation in organic-rich environments and photocatalytic reactions involving hydrophobic substrates.

Additionally, the hydrophobic coating improves charge separation efficiency by reducing surface trap states where electron–hole recombination typically occurs in bare nanoparticles. In some cases, shells can also be engineered to be semipermeable, enabling diffusion of desired reactants while blocking deactivating species such as oxygen or hydroxyl radicals. As a result, hydrophobic core-shell semiconductor nanoparticles combine the intrinsic light-harvesting and catalytic activity of the semiconductor core with the environmental protection and selectivity provided by the shell, making them highly promising candidates for robust and efficient photocatalysis in complex multiphase systems.

Semiconductor core–shell nanoparticles for light harvesting: Semiconductor nanoparticles such as CdSe[95], ZnO96, TiO_2[97] and emerging perovskite[98] nanocrystals have attracted considerable attention in light-harvesting applications due to their unique size-dependent optical and electronic properties[99]. At the nanoscale, these materials exhibit strong absorption across the UV-visible spectrum, high exciton generation efficiency and tunable band gaps, making them ideal candidates for photovoltaic devices, photocatalytic water splitting and photodetectors[100]. However, the intrinsic sensitivity of these semiconductors to aqueous environments, oxygen and high humidity often results in rapid photodegradation, loss of crystallinity or surface defect formation. To address this challenge, the introduction of a hydrophobic shell around the semiconductor core is a crucial design strategy[101].

The hydrophobic shell plays multiple roles in ensuring the long-term stability and efficiency of semiconductor-based light-harvesting systems[102]. Firstly, it prevents water infiltration and suppresses hydrolytic or oxidative decomposition, which is particularly critical for perovskite nanocrystals[103] that are notoriously unstable in moist conditions. Secondly, the shell reduces non-radiative recombination pathways by passivating surface defects, thereby enhancing photoluminescence and charge separation efficiency. In dye-sensitized and quantum dot-sensitized solar cells, hydrophobic encapsulation improves interfacial compatibility with organic electrolytes, ensuring sustained photoresponse over extended operation[104]. Furthermore, the hydrophobic nature of the shell enhances dispersion of these nanoparticles in nonpolar solvents and polymer matrices, enabling the fabrication of flexible, water-resistant optoelectronic devices[105].

Overall, the integration of hydrophobic shells with semiconductor nanocrystals not only preserves their intrinsic optical properties but also introduces environmental resistance, which is indispensable for real-world light-harvesting applications. By combining nanoscale optoelectronic performance with macroscopic durability, hydrophobic core–

shell architectures open pathways for stable, efficient and scalable solar energy conversion technologies.

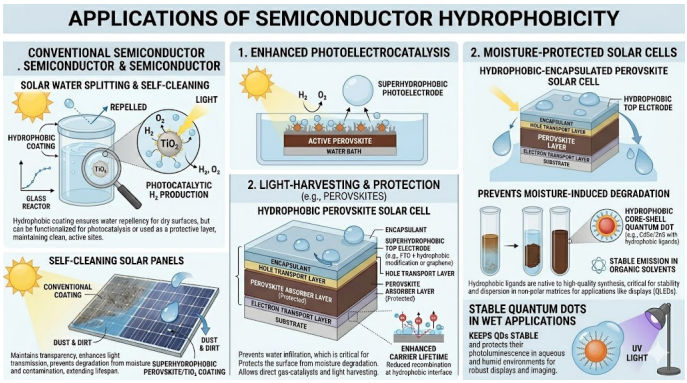


Figure 3: Overall semiconductor hydrophobicity.

SC for superhydrophobic quantum dot: Semiconductor nanocrystals such as CdSe[106], ZnO, TiO_2 and halide perovskites are widely recognized as quantum dots due to their tunable size-dependent optical and electronic properties[107]. Their ability to confine charge carriers within nanometer-scale dimensions imparts unique photoluminescence, quantum confinement and excitonic effects, making them highly valuable in optoelectronic devices, photocatalysis and biomedical imaging[108]. However, the performance and stability of these nanomaterials are strongly influenced by their surface chemistry, particularly when exposed to aqueous or humid environments.

In this context, hydrophobic shell coatings play a critical role in preserving the intrinsic functions of semiconductor quantum dots. For instance, CdSe[109] and perovskite quantum dots are prone to oxidation and hydrolysis, which rapidly degrade their photoluminescence efficiency. Encapsulating these cores within a hydrophobic shell prevents moisture penetration, thereby enhancing chemical stability and extending their operational lifetime in practical applications[110]. Similarly, TiO_2 and ZnO nanoparticles used in photocatalysis or solar cells often benefit from surface modification with hydrophobic ligands, which regulate charge transfer pathways and improve compatibility with organic matrices in hybrid devices[111].

Hydrophobic functionalization also directly impacts colloidal stability and processability[112] of quantum dots. By rendering their surfaces water-repellent, these nanoparticles preferentially disperse in nonpolar solvents, facilitating their integration into polymer films organic light-emitting diodes (OLEDs) or triboelectric nanogenerators[113] where hydrophilic surfaces would otherwise cause aggregation. Moreover, hydrophobicity allows selective interfacial localization of semiconductor nanoparticles at oil-water boundaries, enabling novel applications in photocatalytic emulsions and interfacial energy harvesting[114].

Overall, the hydrophobic shell is not a passive coating but an active design element that governs both the stability and the functional deployment of semiconductor quantum dots. It ensures that the core retains its optical and electronic properties while enabling compatibility with diverse nonpolar environments, making hydrophobicity a central factor in advancing next-generation devices and energy materials. Overall application of hydrophobic semiconductor is shown in **(Figure 3)**.

Carbon-based nanostructures (graphene QDs, CNTs, nanodiamonds): Conductivity, mechanical reinforcement, drug carriers

Carbon-based nanostructures with conductivity against hydrophobicity: Carbon-based nanostructures such as graphene quantum dots (GQDs)[115], carbon nanotubes (CNTs)[116] and nanodiamonds[117] have emerged as versatile functional cores in nanoscale materials owing to their excellent electrical conductivity, high surface area and chemical stability[118]. These properties make them attractive for applications in electronics, energy storage, catalysis and sensing. However, the effectiveness of these nanostructures is often dictated by their surface chemistry. Most pristine carbon nanostructures are intrinsically hydrophobic due to the dominance of sp^2- or sp^3-hybridized carbon frameworks, which limits their dispersion in aqueous media but enhances their performance in organic solvents, polymer matrices and hydrophobic environments[119].

Hydrophobicity plays a critical role in determining the interfacial behavior of carbon-based nanostructures. For instance, CNTs exhibit high intrinsic conductivity, but their tendency to aggregate in water restricts uniform distribution. A hydrophobic shell coating around CNTs can improve compatibility with nonpolar solvents or polymer hosts, enabling percolated conductive networks within insulating matrices[120]. Similarly, graphene quantum dots, which often show partial hydrophilicity due to oxygen-containing groups, can be rendered hydrophobic by surface modification[121]. This hydrophobic character is essential in applications such as oil-water separation membranes[122], triboelectric nanogenerators and organic photovoltaics, where resistance to water and stable charge transfer across nonpolar phases are required[123].

Nanodiamonds, although less conductive than CNTs or GQDs, provide robust mechanical reinforcement when integrated into composites. Their hydrophobic modification ensures better interfacial bonding with polymer matrices, minimizing charge leakage and enhancing dielectric strength[124]. Furthermore, hydrophobicity protects the conductive pathways of carbon nanostructures from moisture-induced degradation, maintaining high electron mobility even under humid conditions. In energy harvesting devices, for example, this combination of conductivity and water repellence ensures stable triboelectric performance[125].

In summary, carbon-based nanostructures inherently offer excellent conductivity, but their hydrophobic functionalization or shell coating is crucial for achieving stable dispersion in nonpolar environments, preventing water-induced defects and maintaining reliable electrical performance. This balance between core conductivity and hydrophobic surface chemistry underpins their use in electronics, coatings, sensors and energy nanodevices.

Carbon-based nanostructures with mechanical reinforcement against hydrophobicity: Carbon-based nanostructures such as graphene quantum dots (GQDs), carbon nanotubes (CNTs)[126] and nanodiamonds[127] have emerged as highly effective reinforcing agents in polymer and composite systems due to their exceptional mechanical, electrical and thermal properties[128]. At the nanoscale, these materials possess extremely high aspect ratios, strong covalent bonding within the carbon lattice and outstanding stiffness, which translate into

remarkable mechanical reinforcement when incorporated into host matrices[129]. The presence of a hydrophobic shell around these carbon-based cores further enhances their compatibility with non-polar or hydrophobic polymer matrices such as polydimethylsiloxane (PDMS), polystyrene or fluoropolymers. This hydrophobic modification minimizes aggregation, improves dispersion within the matrix and maximizes the effective stress transfer between the nanofiller and the polymer chains[130].

Hydrophobicity also plays a crucial role in maintaining the integrity and durability of reinforced composites under humid or aqueous environments. Carbon nanostructures often tend to form agglomerates due to strong π–π stacking or hydrogen bonding interactions, which can compromise mechanical reinforcement[131]. By introducing a hydrophobic shell, these particles resist moisture-induced aggregation and maintain stable interfacial adhesion with the surrounding polymer[132]. This results in composites with enhanced tensile strength, modulus and wear resistance, even under harsh environmental conditions. Moreover, the hydrophobic shell serves as a barrier against oxidative or hydrolytic degradation of the carbon nanostructures, ensuring long-term stability[133].

Thus, hydrophobic core-shell designs of carbon-based nanostructures not only exploit the intrinsic strength of graphene, CNTs and nanodiamonds but also provide superior environmental resistance and processability. This makes them particularly attractive in applications such as aerospace composites, protective coatings, flexible electronics and tribological systems, where mechanical robustness must be maintained alongside water-repellent and anti-fouling properties.

Carbon-based nanostructures as drug carriers: Role of hydrophobicity: Carbon-based nanostructures such as graphene quantum dots (GQDs), carbon nanotubes (CNTs)[134] and nanodiamonds (NDs) have emerged as versatile nanocarriers for therapeutic applications[135]. Their high surface area, tunable surface chemistry and unique electronic structure enable efficient interaction with biomolecules. A critical factor in their drug delivery performance is surface hydrophobicity, which strongly influences drug loading, stability and release kinetics[136].

Hydrophobicity plays a dual role in drug loading. Many anticancer and antiviral drugs are poorly water-soluble and inherently hydrophobic, which limits their bioavailability. The hydrophobic domains of carbon nanostructures provide favorable noncovalent interactions, such as π–π stacking, van der Waals forces and hydrophobic interactions, to encapsulate or adsorb these molecules. For instance, CNTs and graphene sheets can act as nanoscale "hydrophobic reservoirs" for aromatic drugs like doxorubicin, allowing high loading efficiency without covalent modification. Similarly, nanodiamonds with surface hydrophobic facets can stably adsorb hydrophobic therapeutic agents, preventing premature release in aqueous media[137].

Beyond drug loading, hydrophobicity also governs the biological fate and pharmacokinetics of carbon nanocarriers. A moderately hydrophobic surface improves cellular uptake by facilitating interaction with lipid bilayers, enabling efficient translocation across membranes. This is particularly beneficial for cancer therapy, where rapid and selective uptake into tumor cells is desired[138]. At the same time, excessive hydrophobicity can induce aggregation in physiological fluids and nonspecific protein adsorption, reducing circulation time and increasing clearance by the immune system[139]. Therefore, fine-tuning hydrophobicity-often by functionalizing shells with amphiphilic polymers, surfactants or biomolecules-is crucial for balancing drug loading capacity with biocompatibility[140].

In summary, carbon-based nanostructures leverage their hydrophobic surfaces to act as efficient drug carriers for hydrophobic therapeutics, enabling enhanced solubility, targeted delivery and controlled release. However, their performance is highly dependent on optimizing the degree of hydrophobicity: too little results in poor drug affinity, while too much compromises colloidal stability and safety. Thus, controlled surface engineering strategies are essential to harness the full potential of hydrophobic carbon nanocarriers in nanomedicine. Overview of hydrophobisation of carbon-based nanoparticles are shown in **(Figure 4)**.

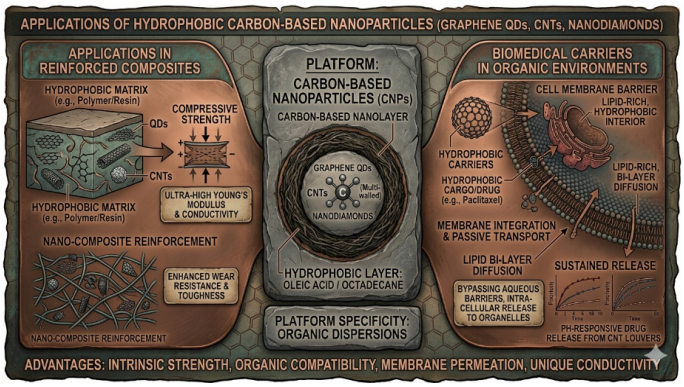


Figure 4: Hydrophisation of carbon-based nanoparticles overview.

Hydrophobic Shell Coatings at Nanoscale

Silane-based shells (e.g., octadecyltrichlorosilane, fluoroalkylsilanes): Give strong water repellency and chemical robustness

Silane-based coatings, such as octadecyltrichlorosilane (OTS) and fluoroalkylsilanes[141], are widely employed to impart hydrophobicity and chemical robustness to nanoparticles[142]. These molecules contain reactive silane groups ($–SiCl_3$ or $–Si(OR)_3$), which readily hydrolyze and condense with surface hydroxyl groups present on inorganic nanoparticle cores such as silica, titania or metal oxides. Through this covalent anchoring, a stable monolayer or thin shell of silane molecules forms around the nanoparticle. The long alkyl chains of OTS or the fluorinated segments of fluoroalkylsilanes orient outward, creating a low-surface-energy outer layer that effectively repels water and resists wetting[143]. At the nanoscale, this modification not only makes individual particles hydrophobic but also protects them against hydrolysis, oxidation and aggregation in aqueous systems. Furthermore, the chemical robustness of the siloxane network ensures that the hydrophobic shell remains stable under harsh environmental conditions, making silane-coated nanoparticles attractive for applications in self-cleaning coatings, oil-water separation and controlled drug delivery in hydrophobic environments. The combination of strong covalent bonding with tunable surface chemistry gives silane-based shells a unique advantage in designing durable and multifunctional core-shell nanoparticles[144]. Overview of hydrophobisation of silane-based shells nanoparticles are shown in **(Figure 5)**.

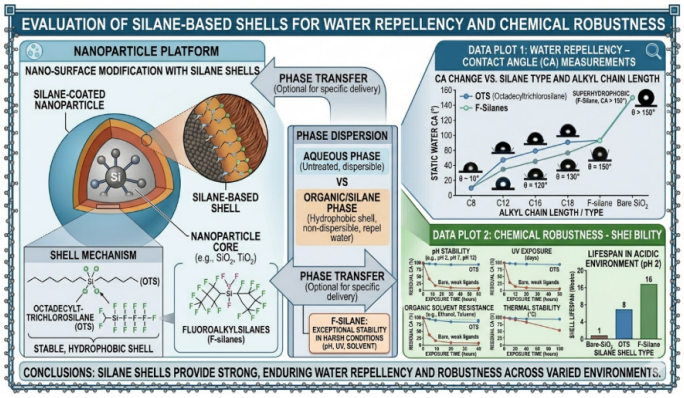


Figure 5: Overview of hydrophobisation of silane-based shells.

Polymeric shells (PDMS, polystyrene, fluoropolymers): Flexible, tunable thickness, biocompatibility options

Polymeric shells such as polydimethylsiloxane (PDMS), polystyrene and fluoropolymers are widely used to coat nanoparticles due to their flexibility, tunable thickness and surface functionalization options[145]. When applied as a shell around functional nanoparticle cores, these polymers provide a protective barrier that enhances stability, dispersibility and compatibility in diverse environments[146]. PDMS coatings are particularly attractive because of their inherent biocompatibility, elasticity and hydrophobicity, which make them suitable for biomedical nanoparticles designed for drug delivery, biosensing or implantable devices[147]. Polystyrene shells, on the other hand, offer rigid encapsulation and controllable porosity, allowing nanoparticles to entrap hydrophobic drugs or act as carriers in controlled release systems. Fluoropolymer coatings impart exceptional chemical resistance and superhydrophobicity, which are highly advantageous for nanoparticles intended for harsh environments, self-cleaning surfaces or anti-fouling applications[148].

At the nanoscale, the thickness of these polymeric shells can be precisely tuned during synthesis using techniques such as emulsion polymerization[149], surface grafting[150] or layer-by-layer deposition[151,152], enabling control over particle stability, drug release kinetics or interaction with biological membranes. Importantly, polymeric shells also serve as a platform for further functionalization, such as attaching targeting ligands, stimuli-responsive groups or stealth coatings like polyethylene glycol (PEG) to enhance circulation time in vivo. Thus, coating nanoparticles with PDMS, polystyrene or fluoropolymers not only provides structural protection and hydrophobicity but also significantly expands their functional versatility for biomedical, environmental and industrial applications[153].

Alkyl-thiol self-assembled monolayers (SAMs): On Au or Ag nanoparticles, making them hydrophobic

Alkyl-thiol self-assembled monolayers (SAMs) are one of the most widely used strategies to impart hydrophobicity to metallic nanoparticles, particularly gold (Au) and silver (Ag). The strong affinity of thiol (-SH) groups toward noble metal surfaces allows the spontaneous formation of ordered monolayers when alkyl-thiol molecules are introduced to colloidal suspensions of these nanoparticles[154]. Upon adsorption, the thiol head group covalently binds to the metallic surface, while the long alkyl chain extends outward, creating a densely packed, hydrophobic organic shell. This coating not only stabilizes the nanoparticles against aggregation but also significantly alters their surface wettability, rendering them hydrophobic in aqueous environments[155]. The hydrophobic shell serves multiple functions: it provides a protective barrier against oxidation or surface fouling, enhances the solubility of nanoparticles in nonpolar solvents and facilitates their incorporation into organic matrices or polymer composites[156]. Furthermore, the modularity of alkyl-thiols enables fine control over surface properties-by varying chain length, branching or terminal functional groups, researchers can tune the degree of hydrophobicity or introduce additional functionalities such as bioactive ligands[157]. In essence, SAM-coated Au and Ag nanoparticles act as engineered nanostructures where the metallic core retains its plasmonic, catalytic or sensing capabilities, while the hydrophobic shell ensures environmental stability and selective interaction with nonpolar systems[158].

Hybrid shells (silica modified with alkyl chains): Combine rigidity + hydrophobicity.

Hybrid silica shells modified with alkyl chains are widely employed as nanoparticle coatings to integrate the structural benefits of silica with the surface properties of organic groups[159]. Silica itself provides a rigid, chemically stable and transparent shell that protects the nanoparticle core from aggregation, oxidation and dissolution[160]. However, pristine silica is inherently hydrophilic due to surface silanol groups, which limits its use in nonpolar environments and reduces compatibility with hydrophobic drugs or matrices. To overcome this, silica surfaces are chemically modified with alkyl chains, such as octadecyl (C18) or fluoroalkyl groups, through silane coupling reactions[161]. The result is a hybrid shell that retains the mechanical robustness and permeability control of silica while introducing a hydrophobic exterior[162].

This hybrid modification plays a crucial role in tuning interfacial interactions at the nanoscale. For example, a hydrophobic silica shell can improve dispersion of nanoparticles in organic solvents or polymer matrices, making them highly suitable for coatings, composites and biomedical formulations. In drug delivery, the alkyl-modified silica shell acts as a hydrophobic reservoir for poorly soluble therapeutic molecules, while the rigid silica framework ensures controlled release and prevents premature leakage[163]. In environmental and catalytic applications, the hydrophobicity of the modified silica shell allows selective adsorption of organic pollutants or enhances catalytic efficiency in biphasic systems. Importantly, the combination of silica's rigidity and alkyl-chain hydrophobicity provides long-term stability in aqueous and biological environments, making these coatings versatile and durable[164]. Overview of hydrophobisation of nanoparticles are shown in **(Figure 6)**.

Unique Features at Nanoscale

Surface-dominated behavior: The hydrophobic shell completely dictates the wettability and colloidal stability of individual nanoparticles

At the nanoscale, particles exhibit surface-dominated behavior, meaning their physicochemical properties are largely controlled by the chemistry of their outermost layer rather than the bulk composition of the core165. When nanoparticles are coated with a hydrophobic shell, this outer layer entirely dictates their

wettability, dispersion and interfacial interactions. For instance, a magnetic nanoparticle such as Fe_3O_4 has an intrinsically hydrophilic oxide surface, but once coated with alkylsilane-modified silica or long-chain fatty acids, the particle behaves like a hydrophobic entity-dispersing preferentially in oils and repelling water[166]. Similarly, gold nanoparticles capped with alkylthiols or fluorinated ligands exhibit strong water repellency and remain colloidally stable in nonpolar solvents, despite gold itself being metallic and hydrophilic in nature. In another case, quantum dots encapsulated in a hydrophobic polymer shell (e.g., polystyrene or PDMS) become highly stable in organic phases and can be embedded into polymeric films without aggregation[167]. These examples illustrate how the hydrophobic coating governs nanoparticle stability and interaction with surrounding media, regardless of the intrinsic chemistry of the core. Thus, by tailoring the shell chemistry, researchers can control colloidal behavior, dispersion environments and surface wettability-key parameters for applications ranging from oil–water separation and drug delivery to self-cleaning coatings and emulsion stabilization[168].

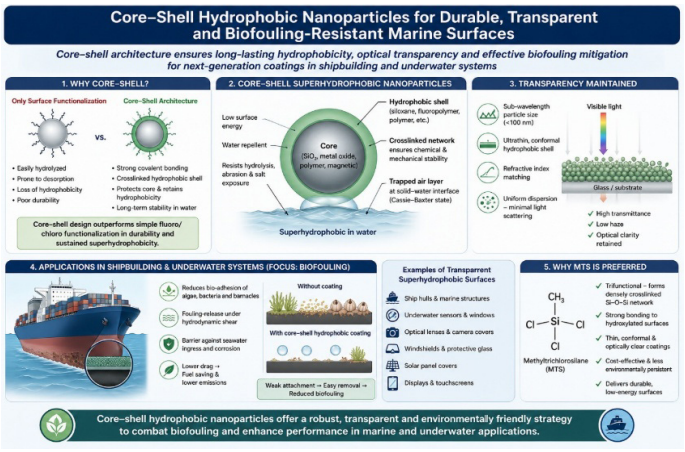


Figure 6: Application of hydrophobic nanoparticles assisting in water repelling surfaces.

Interfacial activity: These particles migrate to oil–water interfaces and stabilize or destabilize emulsions (Pickering emulsions)

Nanoparticles coated with hydrophobic or amphiphilic shells exhibit remarkable interfacial activity, enabling them to spontaneously migrate to the oil–water interface and stabilize emulsions through the Pickering effect[169]. Unlike traditional surfactants that lower interfacial tension through molecular adsorption, coated nanoparticles form a rigid, irreversible barrier at the droplet interface, preventing coalescence and phase separation[170]. For example, silica nanoparticles coated with alkyl or fluoroalkyl chains preferentially anchor at oil-water interfaces, where the silica core provides rigidity while the hydrophobic shell enhances affinity toward the oil phase. Similarly, magnetic nanoparticles functionalized with hydrophobic ligands can be used to not only stabilize emulsions but also break them on demand by applying an external magnetic field, offering controllable separation processes[171]. In another case, carbon nanotubes modified with polymeric shells align at interfaces, improving emulsion stability in polymer blends and enhancing mechanical reinforcement. These examples highlight how nanoparticle coatings determine wettability balance, allowing selective partitioning at fluid interfaces. By tailoring the shell chemistry, nanoparticles can either stabilize emulsions for applications in food, cosmetics and drug delivery or act as demulsifiers in oil–water separation, making interfacial activity a key functional property of coated nanosystems[172].

Stimuli-responsiveness: If the shell is smart (e.g., pH-, temperature- or light-responsive), the core's properties can be exposed or hidden

pH-responsive hydrophobic coatings: Hydrophobic coatings can be engineered to respond selectively to pH, which is particularly important in biomedical environments where normal tissues (pH ~7.4) differ from tumor or endosomal compartments (pH ~5–6). For instance, silica nanoparticles grafted with hydrophobic alkyl chains linked via pH-labile bonds remain water-repellent and shield the core under neutral conditions. In acidic environments, the linker hydrolyzes, making the shell more hydrophilic and permeable. This switch allows the release of hydrophobic drugs or the exposure of catalytic cores only in targeted acidic sites, thereby combining stability in circulation with selective activation[173].

Temperature-responsive hydrophobic coatings: Thermo-responsive hydrophobic shells exploit polymers such as poly(N-isopropylacrylamide) (PNIPAM), which change their hydration state with temperature. Below their lower critical solution temperature (LCST), these coatings are swollen and more hydrophilic, restricting access to the core. Above the LCST, the polymer chains collapse into a more hydrophobic state, increasing permeability or exposing the nanoparticle surface. For example, PNIPAM-modified magnetic nanoparticles have been developed for hyperthermia-assisted drug delivery, where local heating switches the shell from protective to releasing mode, enabling on-demand drug release in tumor tissues[174].

Light-responsive hydrophobic coatings: Light-triggered systems integrate photo-responsive hydrophobic groups such as azobenzene derivatives into silica or polymeric shells. In the dark or under visible light, these coatings maintain a compact hydrophobic barrier, keeping the core protected. Upon UV irradiation, cis–trans isomerization of azobenzene alters the shell polarity and structure, increasing permeability and allowing drug molecules or ions to escape. For example, azobenzene-functionalized hydrophobic silica shells have been used to regulate the release of hydrophobic anticancer drugs, offering remote, non-invasive control over therapeutic action[175].

Redox-responsive hydrophobic coatings: Redox-sensitive systems rely on incorporating hydrophobic disulfide-linked groups in the nanoparticle shell. In extracellular environments, the hydrophobic shell remains intact, protecting the core. Once inside cells, the higher concentration of glutathione (a reducing agent) cleaves the disulfide linkages, disrupting the hydrophobic barrier and turning the surface more hydrophilic. This mechanism is particularly effective in nanodiamond or carbon nanotube carriers, where the hydrophobic shell ensures stable circulation, but the redox environment inside cells enables targeted release of hydrophobic drugs[176].

Magnetic/other stimuli in hydrophobic coatings: Hydrophobic coatings can also be designed to respond to magnetic fields or mechanical forces. For example, hydrophobic polymer-coated Fe_3O_4 nanoparticles can be guided magnetically to a target site, where external heating or oscillating fields alter the shell morphology, increasing its permeability. In such cases,

hydrophobicity ensures core protection during transit, while the external stimulus enables site-specific activation[177].

In summary, stimuli-responsive hydrophobic coatings act as smart barriers: under normal conditions they protect the nanoparticle core and prevent premature interactions, while external triggers (pH, temperature, light, redox or magnetic fields) convert the shell's hydrophobic behavior to a more permeable or hydrophilic state. This dual function provides precise spatiotemporal control, making them highly valuable for drug delivery, catalysis and sensing applications.

Enhanced durability: Protects delicate nanoscale cores (e.g., quantum dots or perovskites) from oxidation, hydrolysis or aggregation

Hydrophobic coatings significantly enhance the durability of delicate nanoscale cores such as quantum dots, perovskite nanocrystals or metallic nanoparticles, which are otherwise prone to rapid degradation. For instance, quantum dots often suffer from oxidation and surface defect formation when exposed to moisture or oxygen, leading to quenching of their photoluminescence[178]. By encapsulating them within a hydrophobic shell, such as alkylsilane-modified silica or fluoropolymer layers, water molecules are effectively repelled from the surface, thereby preserving their optical stability. Similarly, perovskite nanoparticles are highly sensitive to hydrolysis and thermal instability, which limits their use in optoelectronic devices[179]. A hydrophobic shell, often made from long-chain organic ligands or PDMS coatings, acts as a barrier against humidity and solvent penetration, dramatically improving their environmental stability without altering their intrinsic electronic properties. Metallic nanoparticles such as silver or iron oxide also benefit from hydrophobic coatings, where the protective shell prevents aggregation, surface oxidation or corrosion, ensuring long-term dispersibility and retention of magnetic or catalytic activity. Thus, hydrophobic coatings not only provide water repellency but also serve as a shielding layer that safeguards the delicate nanoscale core from chemical and physical stress, thereby extending the functional lifetime of these advanced nanomaterials[180].

Applications of Hydrophobic Core–Shell Nanoparticles

Emulsion stabilization or breaking: Used in controlled emulsification, oil recovery and water treatment

Hydrophobic core–shell nanoparticles can act as effective agents in emulsion control, either by stabilizing emulsions (Pickering emulsions) or by breaking them (demulsification), depending on how they are designed. In stabilization, the hydrophobic shell enables nanoparticles to adsorb at the oil–water interface, where they lower interfacial tension and prevent coalescence of droplets. For example, silica nanoparticles coated with long-chain alkyl groups or fluorosilanes exhibit partial wettability, allowing them to lodge at the interface of oil and water droplets181. This interfacial anchoring creates a mechanical barrier that hinders droplet fusion, resulting in highly stable emulsions used in food formulations, drug delivery emulsions and enhanced oil recovery (EOR). A common example is alkyl-silane modified silica nanoparticles, which stabilize water-in-oil (W/O) emulsions for crude oil extraction by ensuring that small, dispersed droplets remain stable even under high temperature and salinity conditions[182].

Conversely, hydrophobic core–shell nanoparticles can also function as emulsion breakers (demulsifiers). In this role, the hydrophobic coating helps nanoparticles preferentially migrate into the oil phase, where they disrupt the protective surfactant layer at the droplet interface. This destabilization leads to coalescence and phase separation, allowing efficient removal of water from oil or contaminated mixtures[183]. For instance, magnetic Fe_3O_4 nanoparticles coated with hydrophobic polymers can be introduced into oil–water emulsions, where they break the emulsion by collapsing interfacial films[184]. These particles can then be magnetically recovered and reused, making the process both efficient and sustainable. Similarly, hydrophobic carbon nanotube–silica hybrids have been used to break industrial wastewater emulsions by selectively interacting with oil droplets, enabling rapid separation and purification[185].

Thus, through careful tuning of surface hydrophobicity and interfacial activity, core-shell nanoparticles can serve as dual-purpose agents-either stabilizers for creating controlled, long-lived emulsions or demulsifiers for breaking stubborn emulsions in oil recovery and water treatment. The choice of function depends primarily on the balance of hydrophobic-hydrophilic interactions at the nanoparticle surface, making them highly versatile in interfacial engineering.

Drug delivery in hydrophobic environments: Hydrophobic shell increases loading of poorly water-soluble drugs

One of the most important applications of hydrophobic core-shell nanoparticles is in drug delivery of poorly water-soluble therapeutics. Many potent drugs, such as paclitaxel, curcumin and doxorubicin, have very low aqueous solubility, which severely limits their bioavailability and therapeutic efficacy. By engineering a hydrophobic shell coating around nanoparticles, researchers create a surface that has a strong affinity for such drugs, allowing them to be efficiently loaded through hydrophobic interactions, van der Waals forces and π–π stacking. The rigid core-often made of silica, polymer or magnetic nanoparticles-provides stability and functionality, while the hydrophobic shell serves as a reservoir for lipophilic drugs[186].

For example, silica nanoparticles modified with long-chain alkyl silanes (such as octadecyltrimethoxysilane) have been used to encapsulate paclitaxel, where the hydrophobic shell prevents premature release in aqueous environments and allows gradual drug diffusion once inside the body[187]. Similarly, polymeric core–shell nanoparticles with polylactic-co-glycolic acid (PLGA) cores and hydrophobic outer layers have been employed for the delivery of curcumin, improving its stability and therapeutic action against cancer cells[188]. In another case, magnetic nanoparticles coated with a hydrophobic polymer shell enable dual functionality: the hydrophobic layer loads drugs like doxorubicin, while the magnetic core allows for targeted delivery under an external magnetic field[189].

The advantage of such hydrophobic shells lies in their ability to bridge the solubility gap between lipophilic drugs and aqueous biological fluids. Once administered, these core–shell nanoparticles can circulate stably in the bloodstream, with the hydrophobic shell protecting the drug cargo from premature clearance[190]. At the target site, stimuli such as pH change, enzymatic activity or external triggers (magnetic field, light, ultrasound) can induce drug release, ensuring localized delivery with reduced systemic toxicity. Thus, hydrophobic core–shell

coatings provide a powerful strategy for translating insoluble drugs into clinically viable formulations by combining high loading efficiency, controlled release and enhanced therapeutic performance[191].

Nano-triboelectric materials: Hydrophobic shell enhances charge retention under humid conditions

Nano-triboelectric materials are engineered to harvest mechanical energy by generating surface charges through contact electrification. However, a persistent challenge in these systems is their reduced efficiency under humid conditions, as water molecules from the environment adsorb onto the material surface, forming a conductive layer that dissipates charges[192]. To overcome this limitation, nanoparticles with hydrophobic core–shell coatings are integrated into triboelectric devices to maintain charge retention. The hydrophobic shell acts as a moisture barrier, preventing the adsorption of water molecules and thereby preserving the insulating nature of the triboelectric surface[193].

For example, silica nanoparticles coated with fluorosilane or long-chain alkylsilanes have been incorporated into polymer matrices such as PDMS. The rigid silica core contributes to nanoscale roughness, while the hydrophobic shell reduces surface energy, making the composite highly water-repellent[194]. This hierarchical structure not only enhances triboelectric output by providing more contact area but also minimizes charge leakage in humid air. Similarly, carbon-based nanoparticles (graphene, CNTs or nanodiamonds) coated with hydrophobic shells are used in flexible triboelectric nanogenerators (TENGs). Their conductive cores facilitate charge transfer, while the outer hydrophobic functionalization ensures that charges are not neutralized by environmental moisture[195]. In another example, perfluoropolymer-coated $BaTiO_3$ nanoparticles have been embedded into elastomer films, where the ferroelectric core enhances dielectric constant and the hydrophobic shell protects against humidity-induced performance loss.

These strategies demonstrate how core–shell nanoparticles with hydrophobic coatings provide a dual advantage: (i) the core imparts functional enhancement such as increased permittivity, roughness or conductivity, while (ii) the hydrophobic shell safeguards surface charges under humid environments[196]. As a result, triboelectric nanogenerators fabricated with such hybrid nanoparticles can sustain high output voltages and stable performance even in real-world, moisture-rich conditions, which is essential for wearable electronics, self-powered sensors and rural energy-harvesting applications.

Nanocatalysts in biphasic reactions: Core remains active in organic phase, while shell prevents aqueous poisoning

In biphasic catalysis, the reaction takes place between two immiscible liquid phases, typically an organic phase containing reactants and an aqueous phase used for extraction or separation. Nanoparticles as catalysts are highly attractive in these systems due to their large surface area and tunable activity, but they face a major limitation: many active cores (such as noble metals or transition-metal oxides) are sensitive to aqueous deactivation processes, including oxidation, hydrolysis or poisoning by polar molecules[197]. To overcome this, researchers design hydrophobic core–shell nanocatalysts, where the catalytic core remains exposed to the organic phase, while the hydrophobic shell provides a barrier against water molecules. This ensures that the catalyst retains high activity in the organic phase without being deactivated in the aqueous environment[198].

A clear example is Pd nanoparticles coated with hydrophobic silica shells for use in aqueous–organic biphasic hydrogenation and C-C coupling reactions. The hydrophobic shell prevents water and polar species from adsorbing onto the Pd surface, while allowing organic reactants to diffuse through and access the catalytic sites[199]. Similarly, magnetic Fe_3O_4 nanoparticles coated with hydrophobic polymers can catalyze esterification or hydrocarbon oxidation reactions in organic solvents, while remaining protected from aqueous oxidation; the magnetic property also allows easy recovery from the mixture. Another prominent case is gold nanoparticles with thiol-alkyl self-assembled monolayers, which retain high stability in biphasic oxidation of alcohols: the alkyl-thiol layer shields the Au surface from water but allows oxygen and hydrophobic alcohols to access the catalytic sites[200].

This design strategy exploits the amphiphilic nature of the biphasic system: the hydrophobic shell localizes the nanocatalyst at the organic-water interface or within the organic phase, ensuring efficient contact with organic reactants while simultaneously blocking aqueous poisoning. By combining the activity of the catalytic core with the selectivity and protection of the hydrophobic shell, such core–shell nanoparticles achieve long-term operational stability, recyclability and improved efficiency in biphasic chemical transformations.

Self-cleaning and anti-fouling nanocoatings: When dispersed in polymer matrices, they create water-repellent films

Self-cleaning and anti-fouling nanocoatings rely on the integration of hydrophobic core–shell nanoparticles into polymer matrices to impart water repellency and surface protection[201]. In these systems, the core provides functionality (rigidity, durability or optical/catalytic properties), while the shell-functionalized with hydrophobic groups such as fluorocarbons, long alkyl chains or polydimethylsiloxane (PDMS)-ensures that the final surface strongly resists wetting[202]. When these nanoparticles are uniformly dispersed within a polymer film, they create a hierarchical micro-nano roughness combined with low surface energy, leading to the Lotus effect. Water droplets bead up and roll off instead of spreading, removing dirt and contaminants in the process-hence, the coating exhibits self-cleaning behavior[203].

For example, silica nanoparticles coated with octadecylsilane have been incorporated into polyurethane films to generate transparent, water-repellent coatings for glass and textile surfaces[204]. Similarly, TiO_2 nanoparticles encapsulated with hydrophobic shells have been embedded into polymer layers, where they not only resist fouling but also provide photocatalytic degradation of organic residues under sunlight, enhancing the self-cleaning effect[205]. Another example involves fluoropolymer-coated nanodiamonds, which have been dispersed in epoxy coatings for marine applications, where the hydrophobic shell prevents biofouling by microorganisms, algae or barnacles, thereby extending the service life of ship hulls and underwater structures[206].

The success of these nanocoatings lies in the synergy between the polymer matrix (providing mechanical strength and adhesion) and the hydrophobic shell-coated nanoparticles

(imparting surface roughness and water repellency). This combination creates robust, durable films that are useful in applications ranging from architectural glass, textiles and consumer electronics to marine and biomedical devices, where both self-cleaning and anti-fouling properties are critical.

Synthesis Approaches

Miniemulsion/seeded emulsion polymerization: Hydrophobic polymer shells around inorganic or metallic cores

Miniemulsion polymerization for hydrophobic shells: In miniemulsion polymerization, a mixture of monomer, surfactant, co-stabilizer and nanoparticles is dispersed into nanoscale droplets (50–500 nm) under high shear or ultrasonication[207]. Each droplet effectively acts as a nanoreactor, within which polymerization takes place. The nanoparticles (e.g., silica, TiO_2, Fe_3O_4 or quantum dots) are introduced into the monomer phase and stabilized at the droplet interface, where they become encapsulated as the polymer shell forms[208]. The use of hydrophobic monomers such as styrene, butyl acrylate or methyl methacrylate ensures that the resulting polymer shell is water-repellent. The chemistry is usually initiated by oil-soluble radical initiators (e.g., AIBN, benzoyl peroxide), which diffuse within the droplets and trigger chain-growth polymerization. The dynamic confinement of nanoparticles inside these monomer droplets ensures uniform encapsulation and as polymerization proceeds, the particles become permanently coated with a hydrophobic polymer shell[209].

Example: Hydrophilic silica nanoparticles have been successfully encapsulated with polystyrene shells via miniemulsion polymerization. The silica cores were first surface-treated with surfactants to anchor them at the monomer–water interface, followed by radical polymerization of styrene, leading to silica@PS core–shell nanostructures that exhibit strong hydrophobicity[210].

Seeded emulsion polymerization for controlled shell growth: Seeded emulsion polymerization[211] uses pre-formed nanoparticles as "seeds" onto which a polymer shell is grown in a stepwise manner. Unlike miniemulsion, where encapsulation happens inside droplets, this approach allows controlled shell thickness and layer-by-layer growth[212]. The seeds (e.g., gold nanoparticles, Fe_3O_4 or TiO_2) are dispersed in water with surfactants and monomers are slowly added to the system. Polymerization is initiated by water-soluble initiators (e.g., potassium persulfate, APS) and radicals generated in the aqueous phase migrate to the hydrophobic monomer-swollen nanoparticles. The monomer then polymerizes directly at the nanoparticle surface, forming a hydrophobic polymer shell around each core. By controlling the feed rate and monomer composition, precise shell thickness and functionality (e.g., fluoropolymers for extreme hydrophobicity, acrylates for tunable surface chemistry) can be achieved[213].

Example: Magnetic Fe_3O_4 nanoparticles have been coated with poly (methyl methacrylate) (PMMA) shells using seeded emulsion polymerization. The PMMA coating not only provided hydrophobicity, making the particles dispersible in organic solvents, but also enhanced stability against oxidation and aggregation, while preserving their superparamagnetic properties[214].

Dynamics of hydrophobization: In both methods, the core–shell assembly is driven by interfacial chemistry:

- Hydrophobic monomers preferentially surround the nanoparticle surface, creating a low-energy interface.
- Radical initiators localize polymerization within droplets (miniemulsion) or on swollen nanoparticles (seeded emulsion).
- Polymer chains crosslink around the cores, forming a continuous hydrophobic layer that acts as a barrier against water, oxygen and reactive species.

Thus, the hydrophobization of nanoparticles via emulsion-based polymerization is not just a coating process, but a dynamic self-assembly coupled with in-situ polymer growth, resulting in stable and functional hydrophobic nanostructures.

Stepwise Mechanism: Hydrophobizing Nanoparticles via Miniemulsion Polymerization[215]

Step 1: Preparation of Miniemulsion Droplets

- A mixture of styrene monomer + oil-soluble initiator (AIBN)216 is dispersed in water using surfactants (e.g., SDS, CTAB) and a co-stabilizer (hexadecane).
- Under ultrasonication or high-shear mixing, nanoscale droplets (50–200 nm) are formed.
- Silica nanoparticles (SiO_2) are introduced, often pre-treated with a coupling agent (e.g., 3-(trimethoxysilyl)propyl methacrylate, MPS), so they can anchor at the oil–water interface.
- Each droplet acts as a nanoreactor containing both the monomer and the nanoparticle.

Step 2: Initiation of Radical Polymerization

- The oil-soluble initiator AIBN decomposes thermally in the hydrophobic styrene phase:
- These free radicals initiate polymerization of styrene monomers inside the droplet:

Step 3: Growth of Polymer Chains Around the Nanoparticle

- As polymerization proceeds, polystyrene chains grow inside the droplet and because the silica surface is functionalized with methacrylate groups, covalent bonding occurs:
- This covalent anchoring ensures the polymer chains "graft" directly onto the nanoparticle surface.
- The polymer grows outward, gradually forming a continuous hydrophobic shell around the silica core.

Step 4: Formation of Core–Shell Nanoparticles

- Each droplet solidifies into a silica@PS nanoparticle with a rigid silica core and a hydrophobic PS shell.
- The shell thickness depends on the monomer-to-core ratio and polymerization time.
- The final product is a hydrophobized nanoparticle, dispersible in organic solvents and exhibiting water repellency.

Why Hydrophobization Works Here

- **Silica (hydrophilic):** naturally unstable in hydrophobic environments, aggregates in water.
- **Polystyrene shell (hydrophobic):** shields the core, repels water and ensures colloidal stability.

- **Chemistry:** methacrylate silane coupling ensures polymer–inorganic linkage, preventing desorption of the coating.
- **Dynamics:** Mini emulsion confinement guarantees that polymerization occurs around individual nanoparticles, avoiding bulk polymer formation.

Other Examples

- Fe_3O_4@PMMA nanoparticles: Superparamagnetic cores coated with PMMA via seeded emulsion polymerization, making them hydrophobic and stable in organic solvents.
- TiO_2@PS nanoparticles: Photocatalytic TiO_2 stabilized with PS shells to improve dispersion in hydrophobic paints and coatings.

Stepwise Mechanism: Hydrophobizing Fe_3O_4 Nanoparticles with PMMA Shells:

Step 1: Surface Preparation of Fe_3O_4 Nanoparticles

- Bare Fe_3O_4 nanoparticles are hydrophilic due to hydroxyl groups on their surface (–OH).
- To enable polymer shell attachment, they are often functionalized with vinyl- or methacrylate-silane coupling agents (e.g., 3-(trimethoxysilyl)propyl methacrylate, MPS).
- This introduces C=C double bonds on the Fe_3O_4 surface, making it chemically compatible with methyl methacrylate (MMA) monomers.

Step 2: Emulsion Formation

- A water phase containing surfactant (e.g., SDS) is prepared.
- MMA monomers are added to the aqueous phase, forming nanosized droplets stabilized by the surfactant.
- The functionalized Fe_3O_4 nanoparticles are dispersed in the system, acting as "seeds" for polymer growth.

Step 3: Initiation of Polymerization

- A water-soluble initiator (e.g., potassium persulfate, KPS) decomposes in the aqueous phase:
- These sulfate radicals enter the monomer-swollen seed particles or droplets, initiating free-radical polymerization of MMA.

Step 4: Growth of PMMA Shell on Fe_3O_4 Cores

- Polymerization proceeds via radical chain growth:
- Because Fe_3O_4 surfaces are functionalized with vinyl groups, grafting-from polymerization occurs directly at the nanoparticle interface.
- PMMA chains grow outward from the surface, forming a uniform hydrophobic shell around the Fe_3O_4 core.

Step 5: Final Core–Shell Nanoparticles (Fe_3O_4@PMMA)

- The result is a superparamagnetic Fe_3O_4 core encapsulated by a hydrophobic PMMA shell.
- The PMMA coating:
  - Prevents oxidation of Fe_3O_4 cores (which otherwise rust in water).
  - Stops aggregation by steric stabilization.
  - Hydrophobizes the surface, allowing dispersion in organic solvents or hydrophobic matrices.
  - Improves durability for biomedical and environmental applications.

Why PMMA Shell Gives Hydrophobicity

- PMMA has a low surface energy and resists wetting by water.
- Once coated, Fe_3O_4 nanoparticles no longer interact strongly with aqueous media, but instead disperse well in hydrophobic solvents (toluene, chloroform) or polymer films.
- This makes Fe_3O_4@PMMA ideal for magnetic nanocomposites, coatings and targeted delivery systems.

Sol-gel condensation at nanoscale: Silica-based shells, functionalized with hydrophobic groups

The sol–gel condensation method is one of the most widely used techniques for fabricating silica-based hydrophobic shells around nanoparticles. The process begins with the hydrolysis of alkoxysilanes such as tetraethyl orthosilicate (TEOS) in the presence of water and a catalyst, leading to the formation of silanol groups[217]. These silanol groups undergo condensation to generate Si–O–Si linkages, which progressively deposit as a silica network around the nanoparticle core. At the nanoscale, controlling the reaction conditions such as pH, catalyst concentration and surfactant templates is essential to ensure that silica nucleation occurs preferentially on the nanoparticle surface rather than in solution, resulting in a uniform and conformal shell[218]. To impart hydrophobicity, the silica shell is functionalized with organosilanes containing nonpolar substituents such as octadecyl, methyl or fluoroalkyl chains. This can be achieved either by co-condensing TEOS with hydrophobic silanes during shell growth or by post-grafting hydrophobic silanes onto surface silanol groups of preformed silica shells. The resulting hybrid shells combine the rigidity and protective function of silica with the water-repellent character of the organic groups[219].

Fe_3O_4 magnetic nanoparticles coated with silica and modified using octadecylsilane achieve superhydrophobicity, enabling efficient separation of oil from water owing to both their magnetic responsiveness and water-repellent surfaces[220]. In contrast, CdSe/ZnS quantum dots encapsulated in silica shells and further functionalized with fluoroalkylsilanes gain long-term optical stability; the hydrophobic coating prevents quenching by moisture and extends their usability in bioimaging and optoelectronics[221]. TiO_2 nanoparticles, when encapsulated with methylated silica shells, exhibit hydrophobic photocatalytic activity, allowing them to degrade organic pollutants under humid or multiphase conditions without being deactivated by water adsorption[222]. Finally, silver nanoparticles hydrophobized with silica–alkyl silane shells display enhanced stability against oxidation and aggregation, thereby maintaining their antimicrobial efficiency in aqueous and biological environments[223]. Taken together, these examples highlight how the choice of core material dictates the functional outcome of hydrophobization, while the silica-alkyl hybrid shell consistently provides durability, water repellency and compatibility with diverse applications ranging from environmental remediation to nanomedicine.

Ligand exchange/self-assembly: Replacing hydrophilic ligands with hydrophobic ones on metallic or semiconductor nanoparticles

Ligand exchange and self-assembly are widely used strategies to hydrophobize metallic and semiconductor nanoparticles by replacing surface-bound hydrophilic ligands with hydrophobic ones[224]. Nanoparticles synthesized in aqueous media are typically stabilized by hydrophilic capping agents such as citrate, poly (acrylic acid) or short-chain amines, which maintain colloidal stability but restrict their use in nonpolar solvents and hydrophobic environments[225]. In the ligand exchange process, these hydrophilic ligands are substituted with hydrophobic molecules-commonly long-chain alkylthiols, alkylamines or hydrophobic phosphonic acids-that exhibit stronger affinity for the nanoparticle surface. For metallic nanoparticles like gold or silver, thiol groups (-SH) form strong covalent Au-S or Ag-S bonds, anchoring long alkyl chains to the surface and creating a hydrophobic corona[226]. For oxide or semiconductor nanoparticles such as TiO_2, ZnO or CdSe quantum dots, hydrophobic carboxylates, phosphonates or silanes coordinate to surface metal sites, replacing labile hydroxyl or hydrophilic ligands[227].

The dynamics of this process are driven by thermodynamic stability and surface affinity. Strongly binding ligands displace weaker ones through competitive adsorption and the hydrophobic tails self-assemble into ordered monolayers or brush-like structures, lowering the surface energy[228]. For instance, citrate-capped gold nanoparticles can be rendered hydrophobic by treatment with dodecanethiol, resulting in a spontaneous phase transfer from water to nonpolar solvents like toluene[229]. Similarly, oleic acid or oleylamine ligands are used to functionalize CdSe and perovskite nanocrystals, imparting hydrophobicity and improving their stability in organic phases for optoelectronic applications[230]. This ligand exchange not only alters surface wettability but also dictates colloidal behavior, interparticle spacing and compatibility with polymer or solvent systems. By carefully controlling ligand concentration, solvent polarity and reaction time, researchers can achieve well-dispersed, hydrophobically stabilized nanoparticles with tunable surface chemistry[231].

Click chemistry or grafting-from polymerization: Covalently attaching hydrophobic brushes at the nanoscale

Hydrophobisation of nanoparticles via click chemistry or grafting-from polymerization is a powerful strategy to covalently attach hydrophobic polymer brushes at the nanoscale, thereby tuning surface wettability and stability[232]. In the click chemistry approach, nanoparticles such as silica, gold or iron oxide are first functionalized with reactive groups (e.g., azides or alkynes). Hydrophobic ligands or polymers bearing complementary groups are then covalently "clicked" onto the surface through highly efficient reactions like copper(I)-catalyzed azide–alkyne cycloaddition (CuAAC)161. For example, silica nanoparticles modified with azide groups can undergo CuAAC with alkyne-terminated long-chain alkyl or fluoroalkyl polymers, producing a dense hydrophobic coating that resists water infiltration. In the grafting-from polymerization method, the nanoparticle surface is initially functionalized with initiator groups, from which hydrophobic polymer chains are directly grown by controlled radical polymerizations such as atom transfer radical polymerization (ATRP) or reversible addition–fragmentation chain transfer (RAFT)[233]. For instance, iron oxide nanoparticles coated with ATRP initiators have been used to grow polystyrene or poly (tert-butyl acrylate) brushes, creating a hydrophobic corona around the magnetic core[234].

The dynamics of these mechanisms are critical: in click chemistry, the reaction proceeds with nearly quantitative yield under mild conditions, ensuring uniform and robust hydrophobic modification without damaging the delicate nanoparticle core[235]. In grafting-from polymerization, the polymer chain length and density can be precisely controlled, enabling tuning of the hydrophobicity, steric stabilization and interfacial behavior of the coated nanoparticles. This covalent tethering ensures long-term stability against desorption or ligand exchange, unlike physically adsorbed surfactants. Overall, these approaches provide a versatile and durable route to hydrophobization, with applications in drug delivery, self-cleaning coatings, emulsification control and nanocomposite fabrication[236].

Conclusions

In this review, we focus on the existing class of hydrophobic core–shell nanoparticles, examining their diverse applications where the hydrophobic shell plays a decisive role. Particular emphasis is placed on how the incorporation of a hydrophobic shell imparts functionalities that are distinct from those of bare core nanoparticles, thereby enabling exclusive and advanced applications in areas such as drug delivery, catalysis, self-cleaning coatings and environmental remediation. We further highlight the unique features of these core–shell systems, discuss their ability to carve out new technological niches and outline the synthesis techniques by which nanoscale cores are coated with hydrophobic entities. Collectively, this review aims to provide a comprehensive understanding of the design strategies, functional advantages and application potential of hydrophobic core–shell nanoparticles.

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