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Review Article

## Recent Advances in Pickering Emulsions: Stabilizers, Applications and Future Directions

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### Abstract

Pickering emulsions are a type of dispersion in which solid particles stabilize the interface between oil and water, often offering greater stability than traditional emulsions that rely on surfactants. This review thoroughly examines the main factors that affect the performance and stability of Pickering emulsions, such as the particles' wettability, dimensions, morphology, and how densely they cover the interface. A crucial factor, particle wettability, is typically measured by the contact angle formed at the interface of the three phases. Crucial for effective emulsion stabilization, with particles exhibiting intermediate wettability ( $\theta \approx 90^\circ$ ) offering optimal stability. Particle size plays a complex role, with smaller particles enhancing stability by forming dense interfacial layers and resisting sedimentation. Non-spherical particles, such as rods, ellipsoids, and Janus particles, exhibit unique stabilization mechanisms due to their high aspect ratios and deformability at the interface. Surface coverage and electrostatic interactions also significantly impact emulsion stability. The review discusses the pickering emulsions offer several benefits compared to traditional surfactant-based emulsions. These advantages include enhanced stability due to the irreversible adsorption of solid particles at the interface, reduced reliance on synthetic surfactants, and improved biocompatibility and environmental friendliness, reduced toxicity and suitability for various applications. The preparation methods and characterization techniques for Pickering emulsions are summarized, along with their diverse applications in pharmaceuticals, biomedicine, cosmetics, and functional foods. Recent advances and future directions are explored, emphasizing the potential of engineered colloidal particles, multiparticle stabilization, and microencapsulation techniques for enhancing stability and functionality. This comprehensive review provides valuable insights into the fundamental principles and practical applications of Pickering emulsions, guiding future research and development in this promising field.

**Keywords:** Pickering emulsion, applications, future scope

## 1. Introduction:

Emulsions are mixtures formed by blending two liquids that do not naturally mix, typically using mechanical agitation along with a stabilizing agent like a surfactant.<sup>1</sup> To achieve the desired stability, an emulsifier is required to minimize the surface tension at the boundary between the oil and water phases. Emulsifiers are categorized according to their molecular composition and how they function. Depending on their chemical makeup, they can be grouped into synthetic compounds, naturally derived substances, finely divided solid particles, and supporting agents.<sup>2</sup> However, some emulsifying agents may pose risks to human health and contribute to environmental pollution, and certain widely used synthetic emulsifiers, such as carboxymethylcellulose and polysorbate-80, can disrupt essential intestinal microbes that are crucial for human health. This disruption has been shown to cause Mild inflammation, excessive weight gain, metabolic disorders, and various long-term inflammatory conditions observed in normal (wild-type) mice.<sup>3</sup>

To overcome these adverse effects, Emulsions can be stabilized using solid particles, and when this method is

employed, the resulting systems are referred to as Pickering emulsions.<sup>4</sup> The capacity of solid colloidal particles to provide physical stabilization to emulsions has been acknowledged since the early 1900s,<sup>5</sup> and significant Research has been carried out to explore how particle-stabilized interfaces form and behave in simplified systems. Nevertheless, the interest in applying Pickering emulsions within the food industry has emerged primarily in recent times. Provides two reviews of this topic. In the earlier review, he notes that "the proportion of this research directly applicable to foods is necessarily small". By 2012, however, he observes that "various types of dispersed particles of biological origin have recently been shown to be effective for Pickering stabilization of food-grade oil-in-water emulsions"<sup>6,7</sup>. In recent years, there has been growing interest in Pickering emulsions that are stabilized by solid colloidal particles. Because they do not require additional surfactants. These particles are typically eco-friendly and tend to be less toxic compared to conventional surfactants., perfectly aligning with consumer demand for "clean-label" products.<sup>8</sup> Currently, research on Pickering emulsions primarily is concerned with inorganic particles such as graphene, silica, calcium

carbonate, and  $\text{Fe}_2\text{O}_3$ , as well as biological particles such as soybean protein, zein, whey protein, starch, and cellulose. These solid particles typically adsorb irreversibly at the oil-water interface, forming a viscoelastic film through strong interfacial interactions. This led to the development of a dense three-dimensional network structure among the droplets, Granting Pickering emulsions outstanding resistance to droplet merging, Ostwald ripening, and phase separation by creaming.<sup>9</sup> The fundamental criteria for solid particles to act as effective stabilizers in Pickering emulsions include: (i) particles must be partially wetted by both the dispersed and continuous phases while remaining insoluble in either; (ii) particles should possess an optimal degree of partial wettability to promote strong adsorption at the interface.; and iii) Particles must be considerably smaller than the droplets in the emulsion, often by at least an order of magnitude. This requirement is similar to the importance of amphiphilic traits, as indicated by the Hydrophilic-Lipophilic Balance (HLB), in conventional emulsifying agents., These factors are crucial for stabilizing emulsions effectively.<sup>10</sup> The stability of Pickering emulsions largely hinges on how well particles wet the oil-water interface. This wettability is measured by the three-phase contact angle ( $\theta$ ), defined as the angle at the junction where the solid particle, the continuous phase, and the dispersed phase meet. For particles to effectively stabilize emulsions, their contact angle at the oil-water boundary should be near  $90^\circ$ , as this encourages optimal particle arrangement at the interface and forms a robust physical barrier that inhibits the merging of droplets.<sup>11</sup>

Pickering stabilization offers several important benefits, such as precise control over droplet size distribution and the formation of internal particle networks, reduced toxicity, absence of surfactants, and outstanding stability, resulting in a significantly prolonged shelf life that can span from several months to years.<sup>12</sup>

## 2. The primary factors that govern the characteristics and stability of Pickering emulsions encompass the following elements.:

### 2.1 Wettability of Particles:

Pickering emulsions, first reported over a century ago, have garnered significant research interest owing to their unique stabilization mechanisms. The stabilization primarily depends on the assembly of solid particles at the oil-water interface, driven by the reduction in the interfacial area. A key factor influencing stability is the particle wettability, which is often characterized by the contact angle ( $\theta$ ) at the oil-particle-water interface. Hydrophilic particles ( $\theta < 90^\circ$ ) Favor oil-in-water (o/w) emulsions, while hydrophobic particles ( $\theta > 90^\circ$ ) stabilize water-in-oil (w/o) emulsions. Particles with  $\theta \approx 90^\circ$  offer maximum desorption energy, providing optimal stability.<sup>13</sup>

## Key Findings

### Wettability and Stability

Research has shown that extremely hydrophilic or hydrophobic particles result in the formation of unstable emulsions. Intermediate wettability ( $\theta \approx 90^\circ$ ) Enables particles to gather at the oil-water boundary, thereby improving stability. Surface modifications, such as silanization of  $\text{SiO}_2$  particles, enable tuning of wettability to achieve desired outcomes.<sup>14</sup>

### Role of Surface Homogeneity

The uniformity of the surface wettability also plays a critical role. Janus particles with dual surface properties (hydrophilic and hydrophobic) demonstrate superior emulsion stability by attaching strongly to the oil-water interface. Theoretical studies highlight that these particles achieve a minimum energy state when the interface is fully covered, leading to advanced applications and materials.<sup>14</sup>

### Measurement of wettability

The accurate characterization of particle wettability is essential. Methods include: the duration particles take to settle at the air-liquid interface, which is influenced by factors like their size and density. Microcalorimetry to measure the heat of immersion, providing reliable differentiation of particle wettability. Comparative studies have identified microcalorimetry as a robust technique for evaluating the particle wettability.<sup>15</sup>

### 2.2 Particle size:

The dimensions of particles employed in Pickering emulsions play a crucial role in determining the size of the droplets. emulsion stability, and stabilization mechanism. While particles are generally required to be much smaller than the targeted emulsion droplets, studies suggest that the size of particles has a multifaceted influence on the properties of the emulsion.

## Key Findings

### Particle Size and Emulsion Droplets

Particles At minimum, ten times smaller than the droplets in the emulsion. are ideal for stabilization<sup>16</sup>. Larger particles tend to have slower adsorption kinetics, resulting in larger droplets and less effective stabilization. For instance, 150 nm particles stabilizing 450 nm droplets may experience integrity loss during emulsification<sup>17</sup>.

### Size-Dependent Stability:

Smaller particles enhance emulsion stability by reducing sedimentation and improving droplet packing at the interface. Binks and Lumsdon demonstrated that decreasing the particle size improves resistance to sedimentation<sup>18</sup>. Similarly, smaller particles form denser interfacial layers, lowering the interfacial tension and preventing coalescence<sup>19</sup>.

## Particle Concentration and Droplet Size

The amount of particles needed to maintain droplet stability. increased proportionally with the particle size. Nan and colleagues demonstrated this correlation by utilizing chitosan-coated alginate particles of different sizes, revealing that bigger particles demand a greater quantity to effectively stabilize the emulsions.<sup>20</sup>

### Adsorption Efficiency:

Smaller particles adsorb more quickly due to their rapid kinetics, enabling them to pack efficiently at interfaces. In contrast, larger particles experience greater resistance to adsorption, which can lead to the formation of less stable emulsions. Dense interfacial arrangements from smaller particles improve stability against coalescence<sup>19</sup>.

Particle size profoundly influences the stability and droplet size of the Pickering emulsions. Smaller particles enhance the emulsion stability by forming dense interfacial layers, preventing coalescence, and resisting sedimentation. Larger particles, while still effective, require higher concentrations and have slower adsorption kinetics, making them less efficient for stabilization. These findings will guide the design of particle-based emulsifiers tailored for specific applications.

### Particle shape:

While early studies on Pickering emulsions primarily focused on spherical particles, recent research has explored the use of nonspherical particles, including rods, ellipsoids, fibres, cubes, peanuts, Janus particles, microbowls, and deformable nanogels, to stabilize emulsions<sup>21</sup>. These particles facilitated the formation of stable oil-in-water (O/W), water-in-oil (W/O), and multiple emulsions. Nonetheless, the mechanisms by which nonspherical particles stabilize these systems differ markedly from those of spherical particles and are still not completely understood.

## Key Findings

### Detachment Energy and Orientation:

For nonspherical particles, the calculation of detachment energy is more complex because standard equations for spherical particles are not applicable. The orientation and dimensions of a particle must be considered. De Folter *et al.* demonstrated that a modified form of the limited coalescence principle applies to cubic and peanut-like particles<sup>22</sup>.

### Aspect Ratio and Emulsion Stability:

The stability of emulsions is directly affected by the aspect ratio of non-spherical particles. According to findings by Madivala *et al.*, particles with higher aspect ratios contribute to greater emulsion stability and a larger volume of the dispersed phase. This improvement is due to the capacity of anisotropic particles to occupy more interfacial area, allowing for tighter packing, increased viscoelasticity, and enhanced overall stability.

<sup>23</sup>

## Deformability of Particles:

Flexible particles, such as rod-shaped cellulose nanocrystals or microgels, can deform at the droplet interface, improving interfacial anchoring and stability. For example, microgels undergo significant flattening upon adsorption, adopting morphologies such as "fried-egg" or "core-corona" shapes, which enhance interfacial coverage and stability<sup>24</sup>. Softer microgels outperform rigid ones in emulsification and stability<sup>25</sup>.

## Three-Phase Contact Angle Challenges:

Measuring the three-phase contact angle of deformable or non-spherical particles presents significant challenges. Coertjens *et al.* employed freeze-fracture shadow-casting cryo-SEM to determine the contact angle of ellipsoidal particles, highlighting the difficulties involved in evaluating these properties for soft and anisotropic particles<sup>25</sup>.

Non-spherical particles offer unique advantages for stabilizing Pickering emulsions by covering larger interfacial areas and improving packing efficiency. Their deformability further enhanced stabilization by facilitating interfacial anchoring. However, challenges, such as determining the detachment energy and contact angles for these particles, require advanced analytical techniques. This growing field highlights the potential of nonspherical particles to create stable and versatile emulsions for various applications.

## 2.3 Particles surface coverage:

The stability of particle dispersions in solution is largely influenced by their tendency to aggregate due to large surface areas. Ensuring dispersion stability generally relies on steric hindrance or electrostatic repulsion; however, these interactions can introduce an energy barrier that hinders the adsorption of particles at the oil-water interface. Frechette *et al.* explored this phenomenon using ion-pair gold nanoparticles, demonstrating that increasing the pH of the aqueous phase enhances electrostatic repulsion, leading to particle desorption from the interface. This reversible adsorption effect highlights the significant role of electrostatic forces in particle assembly and emulsion stability<sup>26</sup>.

Studies have further emphasized the impact of electrostatic forces on the adsorption behaviour of particles at interfaces and the subsequent stability of Pickering emulsions. These forces influence particle assembly, separation, and the overall integrity of emulsions<sup>27</sup>. introduced a theoretical model to optimize the surface graft concentration of particles, achieving a balance between partial wetting and colloidal stability. Experimental results supported their theoretical findings but also revealed limitations, such as the model's inability to account for the curvature of the water-in-oil (w/o) interface, which affects particle packing and interactions<sup>27</sup>. The stability of Pickering emulsions depends not only on particle wettability but also on electrostatic forces and surface properties. Although progress has been made, gaps in understanding remain. For instance, the interplay

between surface roughness and emulsion stability lacks consensus, and theoretical models require refinement to incorporate interface curvature and complex particle interactions. Expanding the scope of research and diversifying experimental systems will be crucial in establishing a comprehensive framework for designing stable Pickering emulsions.

### 3. Advantages of Pickering emulsions compared to traditional emulsions:

Pickering emulsions offer several benefits over traditional emulsions stabilized with surfactants or emulsifiers, making them highly adaptable and appealing for use in a wide range of industries. Their superior stability arises from the presence of solid particles at the oil-water interface, which act as a physical shield against droplet merging. This barrier significantly reduces the likelihood of coalescence—the merging of smaller droplets into larger ones—thereby greatly improving the emulsion's long-term stability.

Pickering emulsions also align with sustainability goals, as they often utilize biodegradable and biocompatible solid particles such as food-grade starch or biodegradable polymers. This renders them particularly well-suited for use in food, pharmaceutical, and environmental fields. In addition, their minimal dependence on surfactants adds to their appeal. Pickering emulsions mitigate environmental concerns because many surfactants are non-biodegradable and pose ecological risks upon release into the environment.

Due to their versatility in forming both basic and advanced formulations, these emulsions are extensively used in industries like food and beverage, pharmaceuticals, cosmetics, and materials science. By modifying the surface properties of stabilizing particles through coatings or functionalization, the wettability at the oil-water interface can be precisely controlled, allowing fine-tuning of the emulsion composition to meet specific requirements.

Additionally, Pickering emulsions offer superior stability compared to traditional emulsions, which translates to an extended shelf life and potentially reduces the reliance on preservatives. This enhanced stability, combined with their customizable properties, positions them as a valuable solution for improving emulsion performance across a wide range of uses.

Across the food and beverage industry, various microencapsulation techniques have been developed to protect probiotics from harsh conditions. Pickering emulsions, in particular, have gained traction as a promising method for encapsulation, as evidenced by a growing body of research in recent years. Their ability to encapsulate active ingredients while providing stability makes them an appealing choice for innovative product development in multiple fields.<sup>28</sup>

Oil-in-water (O/W) emulsions consist of tiny oil droplets dispersed within a continuous water-based phase. Unlike traditional emulsions, which rely on surfactants (emulsifying agents) for stabilization, Pickering emulsions are stabilized by solid colloidal particles.

These particles assemble into a compact layer at the oil-water boundary, serving as a physical barrier that prevents the merging of droplets. This leads to outstanding stability, primarily due to the irreversible attachment of particles and steric hindrance effects.<sup>29</sup>

### 4. Materials used as a stabilizer:

To efficiently stabilize Pickering emulsions, solid particles need to possess certain traits—such as suitable wettability, particle size, and surface characteristics—that control their ability to adsorb at the oil-water interface. This section focuses on the main types of solid particles used as Pickering stabilizers, with an emphasis on food-grade materials due to their safety and sustainability.

#### 4.1. Inorganic Particles used as a stabilizer:

##### 4.1.1. silica:

Silica is among the most thoroughly researched solid particles used as Pickering emulsifiers. Widely utilized for their affordability, biocompatibility, low toxicity in living organisms, and antimicrobial properties, these particles cannot be directly used to stabilize Pickering emulsions because their surfaces are rich in hydroxyl groups (–OH)<sup>30</sup>. Numerous studies have demonstrated that unmodified silica, which is hydrophilic due to the presence of Si–OH groups on its surface, mainly stabilizes oil-in-water (O/W) Pickering emulsions. Conversely, silica that has been hydrophobically modified is more effective at stabilizing water-in-oil (W/O) Pickering emulsions. Consequently, numerous studies have focused on developing various modified silica types to achieve tailored properties for improved applications in Pickering emulsions, such as through polymerization.<sup>31</sup> A study explores Pickering emulsions stabilized by colloidal silica particles modified with both hydrophilic and hydrophobic groups to mimic surfactant properties. The hydrophilic component, mPEG silane, reduces surface charge, enhances surface activity, and allows pH-controlled particle flocculation, aiding emulsion formation and stabilization. Hydrophobic groups (propyl, methyl, or octyl silanes) balance the surface properties, enabling stable emulsions for over 1.5 years. Smaller droplets were achieved by optimizing particle size and functionalization balance. Key factors for effective emulsification include the ratio of hydrophobic to hydrophilic groups, silane addition order, suspension pH, salt concentration, and particle surface area relative to oil.<sup>32</sup>

Many studies have concentrated on modifying particle surfaces by adsorbing surface-active substances such as surfactants, proteins, and polymers—including chitosan, alginate, and cetyltrimethylammonium bromide (CTAB)—are used with materials like polystyrene, silica, and bentonite, respectively. This study concentrates on stabilizing Pickering emulsions (PEs) by employing SiO<sub>2</sub> nanoparticles (NPs) that have been surface-modified with the surfactant CTAB. Using the Taguchi method, the effects of temperature, pH, and CTAB/SiO<sub>2</sub> ratio on the surface properties and stabilizing performance of SiO<sub>2</sub> NPs were optimized. Results showed that temperature

and CTAB/SiO<sub>2</sub> ratio significantly influenced the zeta potential (ZP), with the optimal conditions being pH 5.5, CTAB/SiO<sub>2</sub> ratio of 2:8, and temperature at 50°C, achieving a ZP of -26 mV for maximum stability. The modified NPs were confirmed via FTIR, TGA, and contact angle measurements. The findings highlight the potential of these modified SiO<sub>2</sub> NPs for use in PEs, particularly in food and pharmaceutical applications, offering a robust and efficient approach to enhancing emulsion stability. The study provides valuable insights but could further explore scalability and application-specific performance.<sup>33</sup>

#### 4.1.2. Clay:

Clay minerals like montmorillonite, kaolinite, and laponite have attracted considerable interest as efficient stabilizers in Pickering emulsions because of their distinctive physicochemical characteristics. Their natural hydrophilicity, elevated aspect ratio, and surface charge enable them to stabilize both oil-in-water (O/W) and water-in-oil (W/O) emulsions, depending on how their surfaces are modified.<sup>34</sup>

Clay particles act as mechanical barriers at the oil-water interface, preventing droplet coalescence and imparting high emulsion stability. Additionally, their biocompatibility, abundance, and low cost further enhance their industrial relevance.<sup>35</sup>

Because of their hydrophilic properties, unmodified clay particles generally stabilize oil-in-water (O/W) emulsions. However, surface treatments, such as organophilic modifications with surfactants or polymers, can increase hydrophobicity, enabling stabilization of W/O emulsions. These modifications also allow tailoring of the interfacial properties to optimize emulsification for specific applications, including cosmetics, pharmaceuticals, and enhanced oil recovery.<sup>36</sup> Recent advancements have explored the role of clay shape (plate-like vs. rod-like), particle size, and wettability in affecting the stability of emulsions and the distribution of droplet sizes.

study explores the stabilization of toluene-in-water (O/W) Pickering emulsions using Laponite RD clay particles. Stable emulsions, resistant to creaming and coalescence for over six months, form only when the clay particles are flocculated, achieved by adding salt (NaCl) at intermediate clay concentrations. The average droplet size depends on the oil volume fraction but not on clay concentration and ranges from 10 to 28 μm. Ostwald ripening initially causes droplet size changes but halts over time due to equalized Laplace pressures. The hydrophilic nature of Laponite RD ensures all emulsions remain O/W, even at high oil fractions, regardless of whether non-polar or polar oils are used.<sup>37</sup>

Nanoclay-based Pickering emulsions have gained significant attention due to their cost-effectiveness, abundance, natural origin, non-toxicity, and biocompatibility. This review highlights recent advancements in nano clay-stabilized Pickering emulsions, discussing their stability, physicochemical properties, and diverse applications, including catalysis,

oil spill remediation, active compound encapsulation, and functional porous material production. Factors influencing stability include wettability, morphology, nano clay concentration, oil type and volume, pH, and ionic strength. Surface modifications and synergistic additives can enhance performance for environmental and industrial uses. Future research should focus on revealing stabilization mechanisms, exploring new materials to adjust nano clay wettability, and developing eco-friendly applications in food, drug delivery<sup>38</sup>. Overall, clay-based stabilizers offer a versatile, sustainable, and cost-effective approach to Pickering emulsions.)

#### 4.1.3. Hydroxyapatite and calcium phosphate:

Hydroxyapatite (HAp) is composed of tricalcium phosphate and calcium hydroxide, represented chemically as Ca<sub>10</sub>(PO<sub>4</sub>)<sub>6</sub>(OH)<sub>2</sub>. It features a calcium-to-phosphorus (Ca/P) ratio of 1.67. Deviations from this ratio can lead to phase impurities caused by the formation of other calcium phosphate compounds. However, solubility is affected by characteristics like particle shape, size, porosity, and degree of crystallinity.<sup>39</sup> Hydroxyapatite (HAp) has emerged as a promising stabilizer for Pickering emulsions due to its unique physicochemical properties, including biocompatibility, nontoxicity, and high surface area. As a naturally occurring mineral in bone and teeth, HAp exhibits excellent bioactivity, making it an ideal candidate for applications in biomedicine, cosmetics, and pharmaceuticals. Earlier research has demonstrated that hydroxyapatite (HAp) nanoparticles can stabilize oil-in-water (O/W) Pickering emulsions when the oil phase contains ester groups or when polymers with ester functionalities are present in the oil. However, HAp nanoparticles by themselves are not effective emulsifiers for these systems. Further investigations revealed that the stabilization of these emulsions largely depends on the interactions between the polymer terminal groups and HAp nanoparticles at the oil-water interface. These interactions also help regulate the droplet size and product morphology. For instance, polystyrene (PS) molecules with different end groups, such as carboxyl or ester groups, and varying molecular weights, were used to study their effects on HAp nanoparticle-stabilized droplets and microstructures. In another study, the interaction between HAp nanoparticles and carbonyl or carboxylic acid groups was explored, where poly(ε-caprolactone) (PCL) It was incorporated because it can dissolve in a wider variety of organic solvents than other polyesters such as poly(L-lactic acid) (PLLA) and poly(L-lactide-co-glycolide) (PLGA), allowing the use of non-halogenated solvents.<sup>40,41</sup> Recently, hydroxyapatite (HAp) particles have been utilized to prepare vitamin E-loaded Pickering emulsions (PEs). These emulsions underwent in vitro digestion and bioaccessibility assessments before being incorporated into fortified food products. The findings revealed that the vitamin E-loaded PEs stabilized by nano-hydroxyapatite (n-HAp) particles disintegrated in gastric conditions and formed aggregates in the intestinal environment. Additionally, these PEs were incorporated into food systems such as gelatin and milk,

the bio accessibility of vitamin E increased significantly—by 3.3 and 6 times, respectively—compared to PEs without food matrix incorporation. This highlights the beneficial effect of incorporating PEs into food matrices to enhance vitamin E bio accessibility.<sup>42</sup>

#### 4.2. Organic Particles used as a stabilizer:

Natural organic particles possess key qualities such as renewability, biocompatibility, cost-efficiency, and biodegradability, while also enabling the valorisation of biomass and industrial byproducts, making them sustainable and versatile materials for various applications.

##### 4.2.1. Starch:

Starch is widely regarded as safe (GRAS), non-allergenic, easily accessible, and cost-effective. Native starch can be modified through several techniques to provide a variety of functional properties suitable for different uses. Recently, certain modified starches have become popular for stabilizing Pickering emulsions. For instance, nano starch shows promise as an effective food-grade stabilizer due to its biodegradability, safety, small particle size, and large specific surface area. Various methods to prepare nano starch, including acid hydrolysis, ultrasonication, and nanoprecipitation, have been explored, with particular focus on hydrophobic modifications to improve its emulsifying capabilities.<sup>43</sup> An oil-in-water (o/w) Pickering emulsion was developed using methyl salicylate as the active ingredient and octenyl succinic anhydride (OSA)-modified quinoa starch. The resulting emulsion exhibited properties suitable for topical application.<sup>44</sup> Starch acquires amphiphilic properties due to the hydrophobic nature of the OSA group, making it well-suited for Pickering emulsion applications. By combining the hydrophobic and steric characteristics of octenyl succinic anhydride (OSA) with the distinctive branched architecture of starch, these derivatives demonstrate outstanding abilities in stabilization, encapsulation, interfacial behaviour, thermal stability, nutritional value, and rheology. This review explores the methods of synthesis, structural analysis, and how the botanical source and structural features affect the physicochemical properties of OSA-modified starches. Gaining insight into these factors may facilitate the design of customized macromolecules with enhanced properties tailored for specific uses.<sup>45</sup>

This review highlights the latest developments in starch modification techniques for the preparation of Pickering emulsions.<sup>46</sup> The article gives the several methods of starch modifications for example, Size reduction-based modifications, Hydrolysis for starch nanocrystal production, Starch nanoparticles can be produced through techniques such as non-solvent precipitation, ultrasonication, ionic gelation, milling, high-pressure treatment, and modification with octenyl succinic anhydride (OSA), as well as by combining starch with non-starch particles. Various starch sources—including native starches from corn, potato, rice, wheat, and tapioca—are utilized in Pickering emulsions, often after

undergoing physical or chemical modifications. Among these, quinoa starch is frequently preferred for emulsion formation due to its excellent functional properties.<sup>47</sup>

##### 4.2.2. Cyclodextrin:

Cyclodextrins are highly effective stabilizers, ideal for creating long-lasting and uniquely functional emulsions, Like Pickering emulsion. Cyclodextrins (CDs) are cyclic oligosaccharides widely used in pharmaceutical applications. The naturally occurring forms— $\alpha$ ,  $\beta$ , and  $\gamma$ -cyclodextrins—consist of six, seven, and eight glucose units, respectively. To enhance their water solubility and improve their capacity to dissolve hydrophobic substances, these molecules are often chemically modified through processes such as hydroxypropylation or methylation of their hydroxyl groups. Commonly used derivatives in pharmaceutical formulations include hydroxypropyl- $\beta$ -cyclodextrin (HP $\beta$ CD), hydroxypropyl- $\gamma$ -cyclodextrin (HP $\gamma$ CD), and randomly methylated  $\beta$ -cyclodextrin, all of which exhibit improved properties. More recently, a novel methylated  $\beta$ -cyclodextrin known as Crysmeb has been developed, providing a more biocompatible option compared to other methylated cyclodextrins.<sup>48</sup> Cyclodextrins, with their unique molecular structure and natural amphiphilic properties, have exhibited an impressive capacity to create host-guest inclusion complexes with small hydrophobic molecules in water-based solutions. This characteristic has been effectively utilized in the formation of Pickering emulsions. Their distinctive structure and stabilizing capabilities make cyclodextrins highly promising for creating stable emulsions and introducing novel functionalities.<sup>49</sup> Compared to traditional surfactants, cyclodextrin nanoparticles offer the advantages of biocompatibility, environmental friendliness, and versatility in modifying surface properties. Their application spans various industries, including food, pharmaceuticals, and cosmetics, where they are used to stabilize emulsions and encapsulate active ingredients. Research has emphasized their ability to create stable high internal phase emulsions (HIPEs) and efficiently deliver bioactive substances.<sup>50</sup>

The research showed that cyclodextrins (CDs) can form complexes with linear oil molecules, allowing them to effectively stabilize emulsions. The amphiphilic nature of  $\beta$ -cyclodextrins ( $\beta$ -CD) was enhanced by esterifying them with octadecenyl succinic anhydride (ODSA) under alkaline conditions, resulting in ODS- $\beta$ -CD particles with degrees of substitution (DS) between 0.003 and 0.019. The addition of hydrophobic chains to the  $\beta$ -CD structure caused changes in their morphology, surface charge, particle size (ranging from 449 to 1484 nm), and contact angle (from 25.7° to 47.3°), all of which significantly impacted their emulsification properties. Moreover, the  $\beta$ -CD crystals underwent a structural change—from a cage-like to a channel form, and eventually to an amorphous state—which further enhanced their ability to emulsify.<sup>51</sup>

### 4.3. New organic–inorganic hybrid polymers as Pickering emulsion stabilizers:

Polyphosphazene (PZS) microspheres, featuring a unique hybrid structure that combines organic and inorganic components in a cyclomatrix framework, have been developed as effective stabilizers for creating

Pickering emulsion droplets. Phosphazene polymers are hybrid materials that combine organic and inorganic components, incorporating elements like phosphorus, nitrogen, carbon, sulphur, and oxygen. They are known for their remarkable thermal stability, biocompatibility, and resistance to solvents.<sup>53</sup>

**Table 1: Preparation and characterization techniques of Pickering emulsion:**

S.N.	Emulsification method	Description	Advantages	Ref
1.	High-shear mixer	High-speed mechanical mixing to disperse solid particles and emulsify phases.	a. Simple b. Fast Processing c. scalable	53,54
2.	High-pressure homogenization	continuous emulsification process that uses a high-pressure valve to transform and disperse one liquid into another uniformly.	a. Small Droplet Size b. Suitable for large-scale, continuous emulsion production. c. high Efficiency	55,56,60
3.	Rotor-stator Mechanism	Rotor-stator homogenizers consist of rotating blades (rotor) within a stationary sheath (stator), which homogenize samples using mechanical tearing and shear forces.	Easy Installation and Use Low Investment Cost Versatility Rapid Process Small Volume Capability	57,58,59
4.	Probe Sonication	Probe sonication uses high-frequency ultrasonic waves emitted from a probe to generate cavitation, creating intense shear forces to break droplets.	a. Produces very fine emulsions with narrow droplet size distribution b. homogeneous Emulsions c. Generates intense localized energy, requiring less emulsifier or stabilizer.	61,62
5.	Micro fluidization	Microfluidic technology is a technique for emulsification that generates droplets of the dispersed phase one by one as the continuous phase within microchannels shears them.	a. Superior Stability b. Gentle Processing c. Nano-Sized Emulsions d. Requires less energy compared to high-pressure homogenizers due to its controlled flow mechanism	63,64
6.	Membrane emulsification	Membrane emulsification is a process where the dispersed phase is forced through a microporous glass or ceramic membrane into the continuous phase, allowing droplets to stabilize at the interface.	a. The size of the droplets can be accurately regulated by choosing the appropriate membrane pore size. b. Environmentally Friendly c. comprises two methods: direct membrane emulsification (DME) and premixed membrane emulsification (PME).	65
7.	Ultrasonic emulsification	Ultrasound equipment utilizes acoustic cavitation—the creation and collapse of air bubbles—to produce powerful physical effects like strong shear forces, turbulence, and shock waves.	a. Small Droplet Size b. Versatility (suitable for emulsification, extraction, emulsion polymerization) c. Fast Process d. Uniform Emulsions	66,67

8.	Hand-shaking	A simple, low-energy method where the dispersed and continuous phases are manually mixed by vigorous shaking to form emulsions stabilized by particles.	a. Requires no specialized equipment, making it accessible for basic lab work. b. Low Cost c. Quick Setup	68
9.	Syringe mixing	A low-energy technique that mixes oil and water phases by repeatedly pumping them between two syringes linked by a three-way stopcock.	a. Suitable for small-scale and on-site emulsification without specialized devices b. Adjustable Parameters	69

## 6. Applications of Pickering emulsion in various fields:

The increasing attention toward Pickering emulsions is driven by progress in particle synthesis methods and the development of novel colloids with tailored surface characteristics. These emulsions are celebrated for their stability and high payload capacity, making them ideal for various applications. Most notably, the biocompatibility of the particle stabilizers used in these systems enhances their appeal, especially in sensitive fields like pharmaceuticals and cosmetics. This combination of factors not only boosts their performance but also opens the door to innovative solutions across multiple industries, making Pickering emulsions a promising area of research and application.

### 6.1 Pharmaceutical applications:

Pickering emulsions, stabilized by solid particles instead of conventional surfactants, have attracted significant interest across multiple industries because of their unique properties. In the pharmaceutical sector, these emulsions play a crucial role in improving the bioavailability of hydrophobic drugs and enabling controlled release. They are especially effective for creating nanocarriers that enhance targeted delivery and minimize side effects. Additionally, their inherent stability allows for the encapsulation of sensitive compounds, safeguarding them from degradation and ensuring their efficacy.<sup>70</sup>

Pickering emulsions, stabilized by unmodified nanocellulose, exhibit excellent structural stability and biocompatibility, making them promising candidates for drug delivery and production. These emulsions are particularly advantageous for applications requiring uniform mixtures or effective release systems, including pharmaceuticals, cosmetics, food products, fuels, and even as models for advanced materials like porous structures, liquid foams, emulsion films, and aerospace components such as core-shell nanofibers and hollow nanotubes.<sup>71</sup> What sets Pickering emulsions apart is their ability to incorporate specific functionalities into their stabilizing particles. For instance, these particles can provide oxidation resistance, UV protection, and responsiveness to environmental changes, as well as unique electromagnetic properties. This versatility opens up exciting possibilities for developing innovative products across a range of industries, from health care

to aerospace, by leveraging the unique attributes of these emulsions and their derivatives.<sup>72</sup>

Pickering emulsions stabilized with modified starch particles show great potential for use in pharmaceutical applications.<sup>73</sup> This research investigates the application of octenyl succinic anhydride (OSA)-modified starch nanoparticles sourced from corn, potato, and pea as stabilizers for Pickering emulsions, demonstrating improved stability under environmental stress, minimizing oil separation, and effectively preserving the bioactive compound curcumin.

Pickering emulsions present a valuable strategy in pharmaceuticals by increasing the stability and bioavailability of active compounds. Their distinctive characteristics, resulting from solid particles at the interface, enhance drug delivery systems. Overall, they offer significant potential for developing innovative drug formulations and targeted treatment methods.<sup>74</sup>

### 6.2 Biomedical Application:

Pickering emulsions hold significant potential in fields such as drug delivery and medical imaging due to their enhanced stability, capacity to transport substantial quantities of materials, and customizable characteristics based on the nanoparticles used for stabilization<sup>75</sup>.

In biomedical applications, the emulsifiers used in Pickering emulsions commonly consist of inorganic or organic particles such as silica or magnesium hydroxide, as well as polymers like poly (lactic-co-glycolic acid) (PLGA), poly(N-isopropylacrylamide) (pNIPAM), polystyrene, and poly (methyl methacrylate) (PMMA). Recently, there has been increasing interest in utilizing natural polysaccharides, including cellulose, starch, chitosan, and alginic acid. These materials are appealing because they are biocompatible, non-toxic, and biodegradable, making them suitable for developing biologically relevant Pickering emulsions.<sup>76</sup>

Pickering emulsions can also be valuable for biorecognition and bio separation by using a technique called molecular imprinting. This approach typically involves creating molecularly imprinted polymers (MIPs) that can recognize specific molecules with strong binding and high selectivity, similar to how natural systems like antibodies work.<sup>77</sup>

The wide range of applications for Pickering emulsions highlights their significant potential in topical biomedicine, offering distinct advantages over

traditional formulations. The improved effectiveness of these emulsions can be attributed to the design of the particulate emulsifiers used. These particles typically exhibit superior adhesion, improving their ability to attach to the skin and boost delivery efficiency without requiring chemical penetration enhancers. This enhanced delivery performance, along with extended shelf life and advancements in biocompatible and biodegradable emulsifiers, positions Pickering emulsions as a promising choice for topical applications.<sup>76,78</sup>

### 6.3. Cosmetic application:

Using Pickering emulsions in cosmetics can help reduce the toxicity and irritation associated with traditional surfactants. Moreover, solid particles enhance droplet stability, making these emulsions ideal for encapsulation, controlled release, and transdermal delivery of active compounds.<sup>79</sup> Recently, Pickering emulsions have attracted considerable attention due to their promising uses in various cosmetics and personal care products, such as skin moisturizers, sunscreens, whitening agents, anti-aging formulations, antiperspirants, deodorants, and hair care items.<sup>80</sup> The rising use of Pickering emulsions in the cosmetics industry is fueled by consumer preferences for safer, environmentally friendly products and the sector's aim to develop affordable, sustainable solutions. These emulsions improve the application and absorption of formulations in specific cosmetic areas. Creating a curcumin-loaded Pickering emulsion with biocompatible ingredients meets the increasing demand for eco-conscious and effective cosmetic products. This formulation not only improves sun protection and antioxidant benefits but also enhances the spread ability and absorption of active ingredients in skin care applications.<sup>81</sup> Incorporating Pickering emulsions in cosmetic formulations offers a safer alternative to traditional surfactants, minimizing irritation and enhancing stability. Their ability to encapsulate active ingredients like curcumin aligns with consumer preferences for eco-friendly products that deliver effective benefits. In summary, Pickering emulsions offer a valuable breakthrough for creating innovative and eco-friendly cosmetic products.

### 7. Recent advances, future direction and challenges:

Pickering emulsions have gained significant attention recently, with their applications expanding across diverse fields such as material science, engineering, biochemistry, food, and cosmetics. Recent advancements in particle synthesis have endowed these emulsions with unique functionalities that traditional surfactant-stabilized emulsions lack. Many studies have highlighted the important role of Pickering emulsions in areas such as interfacial catalysis, biomedicine, drug delivery, and functional materials, indicating that the development of tailored colloidal particles will be a key focus in future research.<sup>82</sup>

Pickering emulsions hold significant potential in functional foods, but their commercial application is limited by susceptibility to degradation, rancidity, and

microbial spoilage due to exposure to air, light, and moderate temperatures. A promising strategy to overcome these challenges is transforming Pickering emulsions into powder via microencapsulation, which improves the shelf life and stability of the encapsulated bioactive compounds. This process involves trapping the bioactive substances inside the emulsions (core) while using stabilizers and protective wall materials (shell) to shield them from external environmental influences.<sup>83</sup>

In the context of food applications, both active and bioactive compounds are important. Active compounds are intended to enhance food stability, while bioactive compounds may provide health benefits. As consumers become more health-conscious and environmentally aware, there is a growing demand for foods that offer convenience, safety, and health benefits. The nutraceutical market is projected to grow substantially in the near future, offering strong potential for the application of Pickering emulsions in bio-derived films and coatings. These emulsions can act as efficient carriers, enhancing the controlled release and absorption of bioactive ingredients<sup>84,85</sup>.

Most Pickering emulsions traditionally rely on a single type of stabilizing particle, although a few studies have explored the use of combinations of rigid particles. In uncommon instances, dual particles with differing characteristics—such as varying softness, hydrophobicity, or surface charge—have been utilized together to achieve co-stabilization. Blending diverse emulsifiers can introduce multifunctional capabilities and responsive behaviours, highlighting the investigation of multiparticle interactions at the emulsion interface as a compelling area for future research and development.<sup>86</sup>

In summary, Pickering emulsions are poised to revolutionize various industries, particularly in functional foods, by enhancing the stability and bioavailability of bioactive compounds through innovative microencapsulation techniques. Their ability to incorporate diverse stabilizers offers exciting possibilities for developing multifunctional applications. As research advances, the exploration of engineered colloidal particles and multiparticle interactions will likely uncover new opportunities in this dynamic field. Overall, the future of Pickering emulsions looks promising, with ample opportunities for research and application across various industries.

### 8. Conclusion:

In conclusion, Pickering emulsions represent a novel type of dispersion where solid particles stabilize the oil-water interface, providing superior stability and reduced toxicity compared to conventional surfactant-based emulsions. This overview emphasizes the key elements that affect the stability and performance of these emulsions, such as the wettability, size, shape, and surface distribution of the stabilizing particles. Optimal stabilization occurs with particles having intermediate wettability and smaller sizes that create dense interfacial layers, while non-spherical particles improve packing and surface coverage. The versatility of both

organic and inorganic particles as stabilizers is emphasized, particularly focusing on food-grade materials. Additionally, the review notes recent advancements aimed at enhancing biocompatibility and functionality through engineered particles and microencapsulation techniques. Pickering emulsions find significant applications across various fields,

including pharmaceuticals, biomedicine, and cosmetics, presenting promising potential for future research and innovation. Overall, this comprehensive coverage illustrates not only the fundamental principles of Pickering emulsions but also their practical applications and the direction for future investigations in this evolving field.

**Table 2: Patents on Pickering Emulsions:**

Sr.no	Patent Number	Title/Description	Assignee	Date of Patent	Key Components	Application	Ref
1.	US 10,925,279 B2	PICKERING EMULSION FORMULATIONS	Syngenta Participations AG, Basel (CH)	Feb 23 2021	a) Colloidal solid b) Dispersed emulsion phase with: - Oily liquid - Solid in oily liquid - Solid dispersed in oil - Colloidal solid at the interface - Ostwald ripening inhibitor	Agricultural pest control	<sup>87</sup>
2.	US 2013/03371 58A1	Pickering Emulsion for Producing Electrically Conductive Coatings and Process for Producing a Pickering Emulsion	BAYER INTELLECTUAL (57) PROPERTY GMBH, Monheim (DE)	Dec. 19, 2013	-Solvent not miscible with water -Sterically stabilized silver nanoparticles	Producing conductive coatings	<sup>88</sup>
3.	US 2014/03297 35 A1	FRAGRANCING COMPOSITION OF PICKERING EMULSION TYPE	LOREAL, Paris (FR)	Nov. 6, 2014	-Dispersed oily phase (apolar hydrocarbon-based oil) -Continuous aqueous-alcoholic phase (C-C monoalcohol) -At least 1% by weight of fragrant substances -Magnesium silicate particles	-Cosmetic formulations -Enhancing texture and stability -Providing fragrance	<sup>89</sup>
4	US 2016/03030 04 A1	COMPOSITION OF PICKERING EMULSION COMPRISING LOW AMOUNT OF ALCOHOL	L'OREAL, Paris (FR)	Oct. 20, 2016	-Dispersed fatty phase (silicone oils, hydrocarbonated oil, or mixture) -Continuous aqueous phase (C1-C4 alcohol, 0.1% to 20% by weight) -Hydrophobic particles (hydrophobic silicas,	Cosmetic formulations	<sup>90</sup>

					starch, talc, etc.)		
5	US 2023/00645 13A1	FINELY DIVIDED AQUEOUS PARTICLE- STABILIZED PICKERING EMULSION AND PARTICLE PRODUCED THEREFORM	Wacker Chemie AG, Munich(DE)	Mar.2,20 23	-aqueous particle stabilized Pickering emulsion  -stabilized by finely divides silica	-for particle production	<sup>91</sup>
6	US 2024/01649 97A1	PICKERING EMULSIONS STABILIZED BY PARTICLE	Firme ich SA. Satingny (CH)	May.23,2 024	-liquid emulsion  -stabilized by organic and inorganic particle  -ethanol free formulation.	-Industrial need	<sup>92</sup>

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