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Review Article

Oleogels in Pharmaceutical Sciences: A Comprehensive Review

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Abstract:
Oleogels are gel-based semisolid systems that exhibit desirable rheological, physical, and chemical stability, making them highly suitable for formulation development. They have wide-ranging applications in the cosmetic, nutraceutical, and pharmaceutical industries. In the pharmaceutical field, oleogels are extensively utilized in topical drug delivery systems and as oil-based gel formulations, particularly for pediatric applications due to their versatility and ease of administration. Oleogels are non-crystalline, thermoreversible, viscoelastic systems composed of a lipophilic liquid phase, such as mineral oils, vegetable oils, or isopropyl myristate, structured by suitable gelling agents known as organogelators. These organogelators form a three-dimensional network through self-assembly and physical interactions, which entraps the liquid phase within the system. Due to their lipophilic nature, oleogels enhance drug penetration through the stratum corneum, thereby improving therapeutic efficacy in topical applications. Additionally, these systems are resistant to moisture and often do not require stabilizers or preservatives, offering an advantage over conventional hydrophilic gels. This article focuses on the components, formulation strategies, and recent advancements in oleogel-based products, highlighting their significant role and growing importance in pharmaceutical applications.
Keywords: Oleogels, Oleogelators, Gel-network Crystallization, Organogelators, Pharmaceutical Applications.

Introduction

The oral route is the most favored method of drug administration among the several dose forms that are given to the body. Although tablets and capsules are a common dosage form, they may have specific drawbacks when given to children who have significant swallowing difficulties. For this reason, we may prefer semisolid items over solid forms when administering medications. Similarly, topical semisolid formulations are a significant class of drug delivery technologies that are increasingly being used in therapy. However, because the skin is impenetrable, applying medications topically is difficult. Therefore, the primary obstacle is getting the medication through the skin by moving the drug molecule through the stratum corneum layer. Semisolid preparations, especially gel-based semisolid products with both solid and liquid components in their structures (also called organogels or oleogels), are used to structure liquid oils in a variety of industries, such as the pharmaceutical or

cosmetics industries, for drug delivery that can facilitate the consumption of a range of medications.

Oleogels are semisolid, non-crystalline, thermo-reversible viscoelastic systems made up of a lipophilic liquid phase (vegetable or mineral oils, isopropyl myristate) gelled with an appropriate gelling agent known as organogelators. Because of their lipophilic nature, oleogels can enhance drug penetration through the stratum corneum [3-5].

Gel-based pharmaceutical products are categorized as hydrogels, emulgels, organogels, or oleogels based on the polarity of liquid components. While oleogels, or organogels, have non-polar dispersion media like fixed oil, mineral oil, organic solvents, etc. that are gelled with an agent known as organogelators, hydrogels use water as the dispersion medium, which is subsequently gelled with a hydrophilic gelling agent.

The three-dimensional networked structure found in oleogel systems traps the polar phase. They are a better option for drug delivery systems than traditional gels because of their moisture resistance and lack of stabilizers or preservatives. They are more favored for topical application to spread uniformly as a film over the skin's surface for drug release because of their good organoleptic qualities, adequate extrudability and spread ability, high flexibility, and high thermal stability [6–11]. Therefore, the utilization of oleogels-based drug delivery devices is growing in pharmaceutical areas due to their simple preparation process and intrinsic long-term stability.

Components of an Oleogel System:

An oleogels system typically consists of two key components. Organic solvent is one of the components, while oleogelators, which immobilize the organic solvent, are the other. Oleogels are oil-structured, three-dimensional (3D) networks of crystalline particles that are created by chilling the hydrophobic solution, shearing it, or using heat. These techniques enclose the liquid oil in self-assembling fibers or polymers [12,13]. Compared to traditional semi-solids, oleogels have greater benefits and increase customer satisfaction [14,15]. Because oleogels are stable at high temperatures for extended periods of time, studies have been conducted to employ them for the oral administration of lipophilic substances [16–19]. There are two key components to this system, which are covered in more detail below.

1. Oils: The oil phase in an oleogels system is responsible for the gel property and the uptake of lipid molecule droplets in the gastrointestinal phase. The gelation process of oleogels is mostly predicted by the chemical structure of oil, namely the quantity of unsaturated fatty acids and the length of the fatty acid chains. Long carbon chains are found in gels with greater stiffness. Additionally, the higher degree of unsaturation in the oil phase, which creates a more uneven spatial arrangement, can occasionally be the cause of enhanced hydrophobicity of oil. Because there are more connections in this kind of system, the increased hydrophobicity feature improves the solubilization of non-polar structurants by creating stronger gels [20–22].

2. Oleogelators: They are of two types

Low molecular weight oleogelators:

These oleogelators feature a stable crystal network and self-assembling characteristics. This is mostly caused by physical interactions, such as hydrophobic and hydrogen bonding forces or Vander Waals forces.

The family of waxes and phytosterols, such as mono-acylglycerol, are significant examples of low molecular weight oleogelators. The characteristics of sitosterol/γ-oryzanol, fatty acids and fatty alcohols [23,24], lecithin/tocopherol25, lecithin/phytosterols26, and waxes/monoacyl glycerols are covered here.

1. Waxes and shellac:

By entangling oil within their pores and adsorbing oil onto the network's surface, these low molecular weight oleogelators create a three-dimensional network [27,28]. Wax is

heated in liquid oil above its melting point to create this kind of oleogel, which is then cooled to 27°C under shear or quiescent conditions. The gelation property influences the behavior of gelation and is dependent on the quality of the oil. This kind of oleogelator is more commonly utilized in food-based products because it is readily available. By creating a water-in-oil type structured emulsion, their thermo-reversible nature occurs [29,30]. Examples of waxes used in edible oleogels in this category are sunflower, carnauba, rice bran, and candelilla wax [31].

2. Phytosterols based oleogels:

β-Sitosterol and γ-Oryzanol are oleogels based on phytosterols. The phytosterols A hydroxyl group is joined to the third carbon of the A ring in sitosterol's steroid skeleton. Similarly, the 17th carbon of D ring [32] has an aliphatic side chain linked to it. γ-oryzanol contains a combination of ferulic acid esters of phytosterols such as campesteryl ferulate, 24-methylenecycloartanyl ferulate, cycloartanyl ferulate, and triterpene alcohols [33]. The prepared oleogels' ability to absorb oil is determined by the ratio of β-Sitosterol and γ-Oryzanol combined with vegetable oils [34]. A review of the literature indicates that the crystallization behavior of the phytosterol-based oleogel [35] can be predicted by adding monoglyceride, which functions as an emulsifier.

High molecular weight oleogelators:

By creating a three-dimensional network by hydrogen bonding, these oleogels, which are composed of high molecular weight structurants such proteins and polysaccharides, can encapsulate oil.

This kind of oleogelator's viscoelastic characteristics are primarily determined by the molecular weight, conformation, and concentration of the polymers. Various instances of this category include.

Polysaccharide based oleogels:

The pharmaceutical industry makes extensive use of oleogels based on polysaccharides. Ethyl cellulose is a significant class of polysaccharide-based oleogels that gel in liquid oils because of its hydrophobic nature [36,37]. Ethyl cellulose forms a stiff, three-dimensional entangled network that aids in oil entrapment when heated above its glass transition temperature, or 130°C, and then cooled. The mechanical characteristics of ethyl are influenced by several factors.

Cellulose oleogels, such as molecular weight, the impact of polymer concentration, gel cooling rate, type of surfactant, polymer to surfactant ratio, etc. Thus, adding surfactants causes the polymer's molecular weight to rise, improving gel hardness and gel point temperature and creating more elastic gels [39, 40].

1. Protein-based oleogels:

Because proteins are hydrophilic, they cannot be used to create oleogels. However, scientists may create protein-based oleogels utilizing the emulsion template approach and solvent exchange method. Solvents such as acetone or tetrahydrofuran (THF) are employed in the solvent exchange process. Similar to this, a new method known as high internal phase pickering emulsions (HIPEs) is utilized to formulate oleogels in the emulsion template method. This method involves first creating an emulsion using protein as an emulsifier, then removing the water phase. Here, oleogels are made from proteins such gelatin, soy protein, and b-lactoglubulin [41,42,43].

2. Oleogels with modifications:

Because oleogels are more stable and have features that make them excellent for usage in a variety of medicinal items, scientists are attempting to alter these properties by creating optimal oleogels for use in a variety of food products. By employing cutting-edge methods known as the "high-intensity ultrasound technique," which aids in figuring out an oleogelator's crystallization behavior and improves the oleogel's rheological, textural, and thermal properties [44].

Formulation of Oleogels:

The following gelation mechanisms can be used

to create oleogels.

1. Fatty acid crystallisation:

Long hydrocarbon chain oleogelators such as natural waxes, fatty acids, fatty alcohols, phytosterols, sorbitan esters, monoacyl glycerols, diacylglycerols, and phosphor lipids are employed in the fatty acid crystallization process for oleogel formulations.

When these hydrocarbon chains reach a specific concentration range, they begin to form crystallite conformations that store oil and encourage solid structure when cooled. Because of their high melting point, long chain length, and low polarity, the waxes cause oleogelation at low concentrations, which results in the creation of a network of needle-shaped crystals that trap oil and cause oleogelation [45]. Nevertheless, rather of needle-like structures, sunflower wax, bee wax, and mineral wax [46] formed plate-like crystal structures.

2. Polymeric networks:

Polymer networks are used in the formulation of oleogels. Food-grade polymers such as hydroxypropyl methylcellulose, methyl cellulose, ethyl cellulose, chitosan and chitin, zein, β-lactoglobulins, gelatine, ethylene-vinyl acetate copolymer, etc. are employed as oleogelators to engulf the oil phase in order to create oleogels.

3. Self-assembled fibrillar networks:

In this method of creating oleogels, self-assembled fibrillar networks induce oleogelation in non-polar solvents through non-covalent interactions such as H-bonding, π-π stacking, electrostatic, and van der Waals interactions, creating a three-dimensional network that traps the oil [48]. The shape, hardness, and compactness of the oleogels generated during this process are determined by many environmental parameters such as storage temperature and cooling rate [49].

Pharmaceutical Applications of Oleogels:

Oleogels are widely used in many different fields, as Figure 1 illustrates.

Bioactive delivery:

In addition to aiding in the regulation of drug release, the oleogels system has targeted the release of lipid-soluble nutraceuticals such as carotene, lycopene, co-enzyme Q10, docosahexaenoic acid, eicosapentaenoic acid, tannins, etc. These nutraceuticals have been shown to provide various health benefits to humans, such as reducing platelet aggregation, blood viscosity, and fibrinogen, as well as lowering the incidence of chronic diseases like cancer and cardiovascular diseases [50]. This bioactive lipid soluble chemical is encapsulated in oleogels, which improves their solubility in the gut and aids in regulating medication release. In addition to the aforementioned lipid compounds, oleogels also contain a variety of carbohydrates, such as modified and hydrolyzed starches, cellulose derivatives, proteins, such as whey proteins, caseinates, and gelatine, and gums, such as exudates and extracts.

Topical Applications:

Topical formulations are the preferred method of drug delivery since they increase drug permeability and lessen the adverse effects of the oral route. Therefore, oleogel formulations have been effectively studied as dermal pharmaceuticals [52] because they aid in the penetration of medications such as nitroglycerine, scopolamine, nicotine, clonidine, fentanyl, estradiol, testosterone, lidocaine, and oxybutinin. By eliminating the severe gastrointestinal irritation that occurs when aceclofenac is administered orally, scientists are developing a topical transdermal drug delivery oleogels system for aceclofenac [53,54].

Oleogels in cosmetics:

The majority of today's cosmetic formulations are emulsion-based goods made of oil and water. Oleogels cosmetic pharmaceutical formulations soften and smooth rough and cracked foot skin since they include oils. Additionally, the formulations of oleogels are advised for the supportive treatment of diabetic skin, perianal skin conditions, and sun protection products that shield skin from damaging sunlight [55,56].

Nutraceutical applications:

A pharmaceutical substitute with physiological advantages is called a nutraceutical. These days, researchers are changing the physical characteristics of oils to make them more like fats and by adding particular molecules (polymers, amphiphiles, and waxes) to the oils to create oleogels.

For instance, ethylcellulose has a lot of promise for binding oil at concentrations of 10% and less, creating oleogels with a wide range of characteristics that can be applied topically.

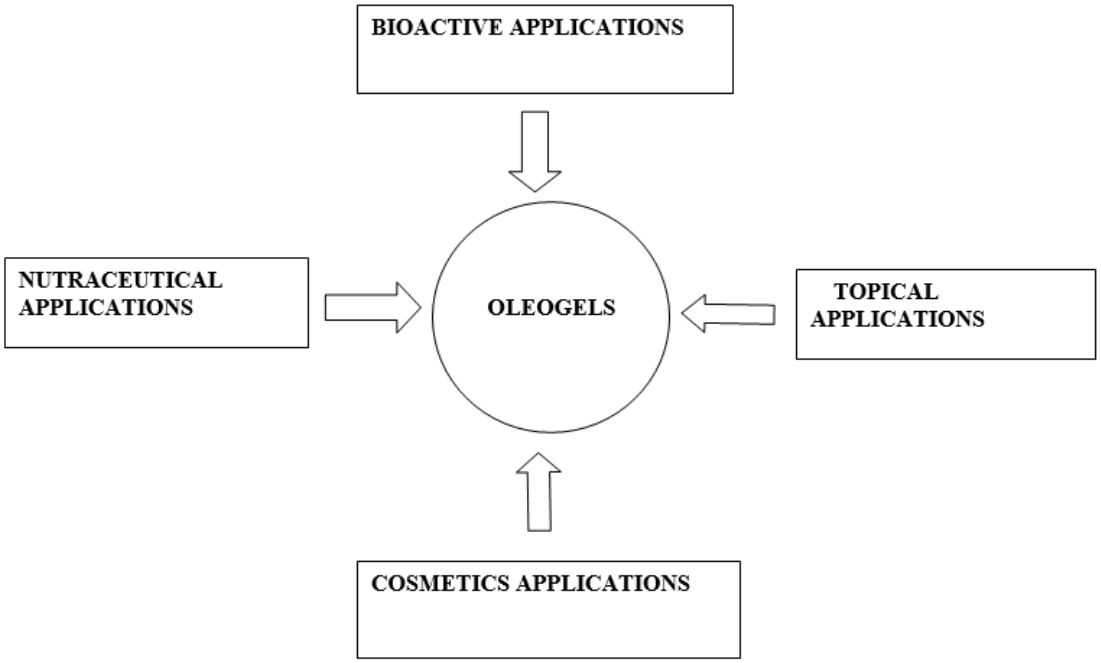

Figure 1. Dıfferent Roles of Oleogel System

Conclusions:

The usage of oleogels in food goods, pharmaceuticals, and drug delivery systems has risen significantly during the past few decades. Numerous oleogel formulations in solid, liquid, and semisolid dosage forms have been introduced to the market in recent years. Physical and microbiological stability issues are present in a variety of topical formulations, such as ointments, lotions, and gels.

According to recent studies of the literature, oleogel is a promising base for a variety of medications that can be used to create topical formulations that are resistant to microbiological activity and have superior chemical and physical stability when compared to traditional topical base formulations. The use of oleogels in a variety of pharmaceutical applications, such as topical formulations, bioactive delivery, and baked goods, implies that they can be applied on a wider commercial scale.

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