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COMPREHENSIVE INSIGHTS INTO OLEOGELS

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ABSTRACT

In recent years, oleogels have emerged as crucial entities in the pharmaceutical, cosmetics, and food industries, owing to their unique properties and versatile applications. Oleogels are non-crystalline, thermoreversible matrices wherein solid components, known as oleogelators immobilize vegetable oils. This review explores the fundamental aspects of oleogels, including their preparation, characteristics, and role as effective drug delivery systems. Compared to conventional gels, oleogels offer enhanced stability against moisture and microbial growth, making them favorable for pharmaceutical formulations. Their rheological properties facilitate the controlled and sustained release of active pharmaceutical ingredients (API) across various routes of administration such as topical, transdermal, oral, and ocular applications. Moreover, oleogels exhibit good biocompatibility and biodegradability, further supporting their safety and efficacy in therapeutic use. This article summarizes the key points regarding oleogels, highlighting their composition, benefits, and applications across different industries, focusing on pharmaceuticals.

KEYWORDS: Oleogel, Oleogelator, Vegetable oil.

INTRODUCTION

In recent years, semi-solid products have acquired much importance in the pharmaceutical, cosmetics, food industries, etc. Semi-solid preparations with solid and liquid components in their structures have been regarded as gels. Gel-based semisolid products are more stable than other types.[1]

In general, based on the polarity of the liquid components, gel-based products may be categorized as hydrogels, emulgels, organogels, or oleogels. Oleogel is a type of gel in which vegetable oils are immobilized using a solid component. Oleogels have received great interest recently due to their potential to develop formulations for cosmetics, foods, and pharmaceuticals.[2] The process of inducing gelation is called oleogelation. The solid components that cause the gelation of oils are known as oleogelators. Examples of oleogelator are lecithin, sorbitan monostearate, ethyl cellulose, stearic acid, etc.

Oleogel is a non-crystalline, non-glassy, thermoreversible (thermoplastic) solid material and viscoelastic system in which the polar phase gets immobilized within spaces of the three-dimensional networked structure formed in the oleogel system.[3] Mainly, oleogels are resistant to moisture, and in the absence of stabilizers or preservatives, they are the preferred choice for drug delivery systems over conventional gels. Due to their good organoleptic characteristics, satisfactory extrudability and spreadability, high flexibility, and high thermal stability, they are chosen for topical administration to spread evenly as a film over the skin's surface for drug release.[4]

So due to the easy preparation method and inherent long-term stability, the use of oleogel-based drug delivery products is increasing in pharmaceutical fields. The present review deals with the components of oleogels, advantages, disadvantages, properties, and roles of oleogels in different drug delivery systems.

Components of An Oleogel

Generally, oleogels contain two components. One component is an oil and the other is an oleogelator/gelator.

1. Oil

Mainly vegetable oils were used as oils in oleogels. Oils help to improve the viscosity and make it easy to spread. Some examples of oils used in oleogels are soybean oil, sesame oil, olive oil, coconut oil, sunflower oil, etc.[5]

1.1 Soybean oil

Soybean /soyabean oil is a vegetable oil extracted from soybean seeds. The refined soybean oil contains linoleic acid (50-57%), linolenic acid (5-10%), oleic acid (17-26%), palmitic acid (9-13%), and stearic acid (3-6%). It is one of the most widely consumed cooking oils and the second most consumed vegetable oil. Soybean oil is used for oral, topical, and intravenous

administration of drugs. It is also used in the formulation of many drug delivery systems such as liposomes, microspheres, microemulsions, nanoemulsions, nanocapsules and organo/oleogel. It may also be used in cosmetics and it has emollient properties, it is used as a bath additive in the treatment of dry skin conditions.[6]

1.2 Sesame oil

Sesame oil is an edible vegetable oil derived from sesame seeds. The refined sesame oil contains arachidic acid (0.8%), linoleic acid (40.4%), oleic acid (45.4%), palmitic acid (9.1%), and stearic acid (4.3%). Sesamin, a complex cyclic ether, and sesamolin, a glycoside, are also present in small amounts. Sesame oil may be used as a solvent in the preparation of subcutaneous injections, oral capsules, rectal suppositories, and ophthalmic preparations. It may also be used in the formulation of suspensions and emulsions.[6]

1.3 Olive oil

Olive oil is a liquid fat obtained by pressing whole olives. It is commonly used in cooking for frying foods or as a salad dressing. It can also be found in some cosmetics, pharmaceuticals, and soaps. Olive oil is a mixture of fatty acid glycerides. It mainly contains, Palmitic acid (7.5–20%), Palmitoleic acid (0.3–5%), Hepatodecenoic acid (0.3%), Stearic acid (0.5–5%), Oleic acid (55–83%), Linoleic acid (3.5–21%), Linoleic acid (40.9%), Arachidic acid (40.6%), Eicosaenoic acid (40.4%), Behenic acid (40.2%), Lignoceric acid (41%) and sterols are also present.

Olive oil has been used in enemas, liniments, ointments, and plasters. It has also been used in oral capsules and solutions and as a vehicle for oily injections including targeted delivery systems. It has been used in topically applied lipogels and has also been used to soften ear wax. In cosmetics, olive oil is used as a solvent, and also as a skin and hair conditioner. Types of products containing olive oil include shampoos and hair conditioners, cleansing products, topical creams and lotions, and sun-tan products.[6]

2. Oleogelator

Oleogelators are classified into two depending upon the nature of the gelators. They are, Low molecular weight oleogelators (LMWOG) and Polymeric oleogelators as shown in Fig. 1. Physical and chemical gels form when polymeric gelators trap the organic solvent by creating a network of entangled or cross-linked chains. Weak interchain interactions such as π-π stacking, van der Waals forces, and H-bonding, further enhance the physical stability of the

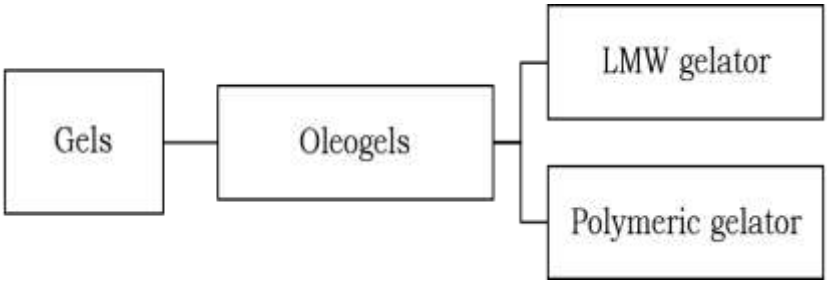

Figure No. 1: Classification of oleogels.

gels. Similarly, the formation of a sufficiently long aggregation to cause solvent gelation and overlapping in low molecular weight oleogelators depends on their physical interactions. Based on their composition and the kinetic characteristics of the aggregates, low molecular weight oleogels are classified as solid-fiber matrix or fluid-fiber matrix.[7]

2.1 Low molecular weight oleogelators (LMWOG)

Most low molecular weight oleogelators are highly stable and more efficient at low concentrations with different organic solvents. The solubility of gels with organic solvents is increasing by heating and smooth gelation by adding low concentration. These gelators are typically used for gelation at 0.5 and 3% w/v concentrations. These gelators have high gelation ability and the resulting gel will last for a long time. There are different types of LMWOGs including waxes, fatty acids and fatty alcohols, phytosterols, ceramides, lecithin, surfactant, etc.[8]

2.1.1 Waxes

Waxes are a class of organic compounds that are comprised of long alkyl chains including various functional groups such as fatty acid esters, fatty alcohols, aldehydes, ketones and aromatic compounds. The main component for the formation of gels is the wax ester, which constitutes 75% of the total mass of the wax. For example; Rice bran wax, carnauba wax, candelilla wax, beeswax, etc.[8,9]

2.1.1.1 Rice bran wax

Rice bran wax (Oryza sativa) (RBX) is a natural plant wax derived from rice bran, which is a byproduct of rice milling. It is mainly used in cosmetics, pharmaceuticals, food, polymer and leather industries. It contains long hydrocarbon chains of fatty acids (C_{16}-C_{32}) and fatty alcohols (C_{24}-C_{38}). RBX has a melting point range of 78-81ºC. Due to its long hydrocarbon chain, it develops strong intermolecular interactions that can crystallize readily to give unique

crystal morphology and physical properties. Also, the rate formation of oleogels was highest for RBX as compared to other waxes.[9,10]

2.1.1.2 Carnauba wax

Carnauba wax (CRX) is wax from the carnauba palm Copernicia prunifera leaves. It has a melting point range from 81-86 ºC and consists of a complex mixture of high molecular weight esters of acid and hydroxyl acids. CRX is used in cosmetics and edible films.[10]

2.1.2 Fatty acids and fatty alcohols

The biocompatibility of the fatty acids and fatty alcohols under a physiological environment enables them to behave as a good gelator to form oleogels. Mainly the vegetable oil had no large effect on the structure, but the ratio between acid and alcohol significantly influences the structure hardness.[8]

2.1.3 Phytosterols

Mixtures of phytosterols and phytosterols esters form highly stable oleogels. Phytosterols form transparent gel because of their small size and molecular arrangement. For example; a combination of γ-oryzanol and β-sitosterol mixtures produces a transparent gel in sunflower oil oleogel form with hollow tubules-like morphology. β-sitosterol with γ-oryzanol can form oleogels with high mechanical properties and high levels of transparency.[9]

2.1.4 Ceramides

Ceramide-based gels are derived from sphingosine and fatty acids. They help to retain skin moisture and protect from skin irritants and environmental pollution. A large variety of ceramide-based oleogelators are used for gelling vegetable oils and they are used in the food and cosmetic industries. The gelation ability of ceramides depends on the length of the fatty acid chains. Short-chain fatty acids have high gel-forming ability even at lower concentrations compared to longer-chain fatty acids.[8,11]

2.1.5 Lecithin

Lecithin is a phospholipid, extracted from various plants and animal tissues separately from the egg cell. Lecithin derived from natural sources can form gelled structures and has been caused by the presence of incomplete chemicals within its structure. Synthetic lecithin and hydrogenated soy lecithin failed to develop oleogels. Apart from the chemical structure, the purity is excluded Lecithin also plays a key role in the formation of oleogel. Experimental

results show that lecithin fails to initiate the gellification process of an apolar solvent when lecithin contains <95% phosphatidyl content. In the case of the combination of sorbitan with lecithin can form a gel at low concentrations at 4% w/w. The mechanical behaviour of gel depends upon the optimum ratio of lecithin and sorbitan ranging between 2:3 and 3:2 lecithin/sorbitan. These gelators form needle-like crystals of 10 μm. Lecithin-based oleogels were found to be thermodynamically strong, thermoreversible (sol-to-gel transition temperature at 40◦C), transparent, viscoelastic, and biocompatible.[12]

2.1.6 Surfactants

The compound that lowers the interfacial tension between two phases is called surfactant. The self-assembly of surfactant molecules can be influenced by many factors such as pH, ionic strength, concentration, nature of oil, type of solvent and temperature. The physical properties of the oleogels are affected by the surfactant and the type of oil. Surfactants form lamellar structures in oils, leading to crystal-like platelets which form a three-dimensional aggregate structure, thereby stabilizing the oleogels. Some examples of surfactants are sorbitan monostearate, glycerol monostearate and sorbitan monopalmitate.[8] Non-ionic surfactants such as spans and tweens offer several advantages over ionic surfactants including increased stability, flexibility in formulation and more compatibility. Certain spans and tweens are also highly effective solubilisers, dispersing agents and wetting aids.[13]

2.2 Polymeric oleogelators

Oleogelation in polymeric materials takes place through the process of swelling, whereby physical interactions produce network structures. Mainly it is observed in star-shaped, linear and hyperbranched polymers. The gels obtained from these oleogelators show low gel-to-sol transition temperatures compared to LMWOGs. Some of the common polymeric oleogelators are cellulose derivatives, proteins, etc.[8]

2.2.1 Cellulose derivatives

Ethyl cellulose, a linear polysaccharide, is the first polymer used as an oleogelator. Ethyl cellulose-based gels were obtained by dissolving it in liquid oil above the glass transition temperature, i.e. 140 °C, and cooling down to room temperature. Ethyl cellulose is widely used in oral and topical pharmaceutical formulations. In topical formulations, it serves as a thickening agent in creams, lotions, or gels, provided an appropriate solvent is used. Ethyl cellulose has been studied as a stabilizer for emulsions and is used in cosmetics and food products.[14,15]

2.2.2 Proteins

Proteins are substances that are highly hydrophilic and made up of a sequence of amino acids. They do not dissolve well in oil, which makes it difficult to use them as oleogelators. The oil droplets in an oil-in-water emulsion are stabilised by a cross-linked monolayer of protein adsorbed at the surfaces. When the water is evaporated, it results in a dry foam network-like structure, where the air gaps are filled by oil and the proteins are present on the walls. The physical properties of the gel can be varied by altering the diameter of the droplets in the emulsion.[13]

Advantages of Oleogels

- Ease of preparation
- Cost reduction
- Longer shelf life
- Short-half-life drug used
- Thermodynamically stable
- Controlled release of drugs
- Reduces the frequency of drug dosing.
- Avoid first-pass metabolism.
- Oleogels are moisture-insensitive.
- More stable than other types of gel
- Both hydrophobic and hydrophilic drugs can be incorporated.[16,17]

Disadvantages of Oleogels

- Should be stored in a proper condition.
- The oleogels have a greasy property.
- Less stable to temperature.[13]

Methods of Oleogelation

The formation of oleogels is carried out using three methods. They are; fluid-filled fiber mechanism, solid fiber mechanism and hydration method.

i. Fluid-filled fiber mechanism

A non- polar solvent is combined with surfactant combination, and reverse micelles are formed. The addition of water to it produced tubular reverse mixture. Addition of more water leads to the development of 3D network.[18,19]

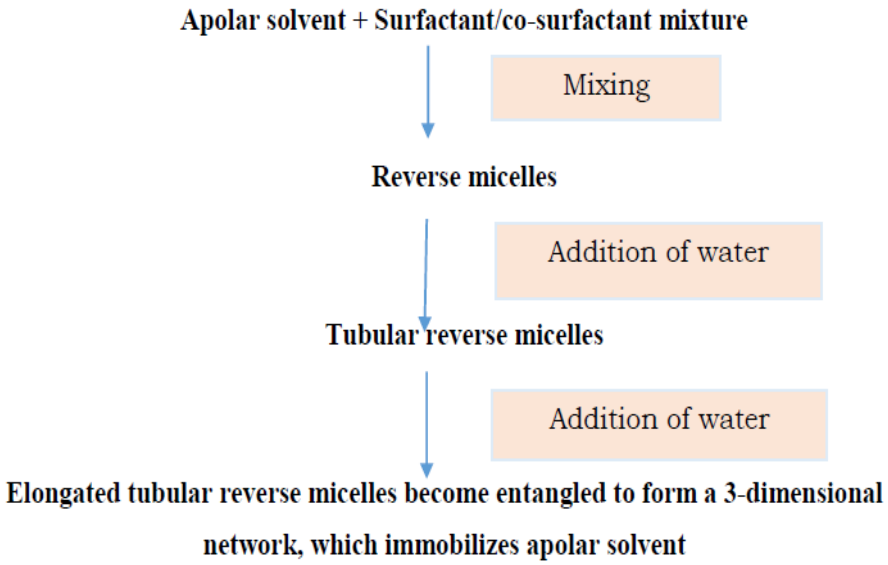

Figure No. 2: Fluid filed fibre method.[20]

ii. Solid fiber mechanism

A solid oleogelator is added to the organic solvent, and the mixture is heated at a temperature higher than the solid oleogelator's melting point, after the complete dissolution of the gelator into the solvent. The solution is placed for cool down. Finally, the 3D networked oleogel is formed.[19,21]

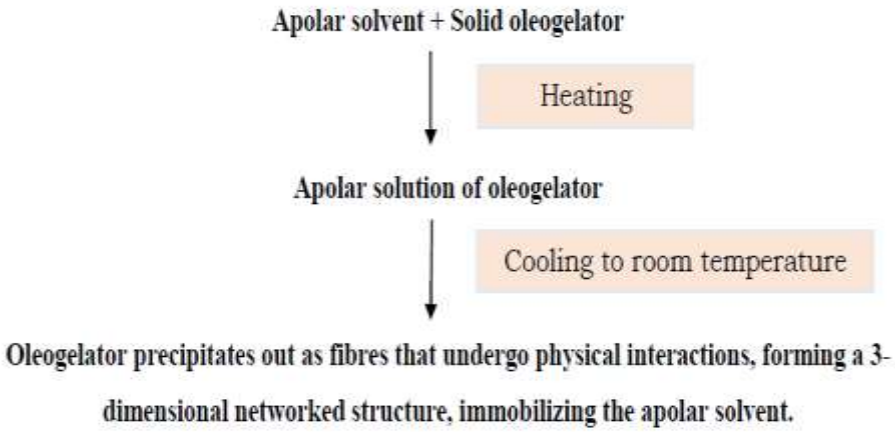

Figure No. 3: Solid fibre method.[21]

iii. Hydration method

The gel is prepared using hydrated inorganic chemicals, which produce a dispersion in the addition of water or other agents such as propylene glycol, hydroxyl propyl cellulose, etc. It was used for the enhancement of gel formation.[18,19]

Mechanism of Formation of Oleogels

Normally, gels are obtained by mixing the gelator into the organic solvent at a high temperature (higher than the melting point of gelators) followed by cooling at room temperature. Cooling the super-heated solution will decrease the affinity of solvent molecules and the oleogelator which leads to the self-assembling of oleogelators into solid aggregates via physical intermolecular interactions. The assemblies take place in three ways: (1) Crystallization, (2) Precipitation and (3) gelation.[8]

Properties of Oleogels

Viscoelasticity

Viscoelastic means a material having both viscous and elastic properties. Generally, oleogels have an elastic behaviour at low shear rate. As the shear rate increases, it will disrupt the physical interaction in gel; further, it tends to flow.[22]

Non-birefringence

Non-birefringence means that the material does not split a light wave into two distinct beams with different velocities when it passes through the gel. The morphology of oleogels shows dark matrix in the microscope, which indicates that the oleogel is optically isotropic, meaning its optical properties are the same in all directions.[22]

Thermoreversibility

When the oleogels are heated at high temperatures above their critical temperature, they lose their solid matrix-like structure and begin to flow, which results in the gel-to-sol transition. However, it can be reversed by cooling down.[13]

Thermostability

Oleogels are thermos stable in nature. The ability of oleogelators to undergo self-assembly and form gels under suitable conditions which depends the thermostability of the oleogels.[23]

Optical clarity

Oleogels can be transparent or opaque depending on the composition and mode of aggregation of the oleogels.[12]

Biocompatibility

Biocompatibility refers to the ability of a material to interact with biological systems without causing adverse effects. The biocompatibility of oleogels depends on the biocompatibility of the oleogelator chosen. Generally, oleogels are biocompatible.[23]

Chirality

Low molecular weight gelators influence the growth and stability of solid fibre networks due to their chirality. However, chirality does not affect fluid fibre networks. The presence of chiral centers within the gelators aids in forming a compact molecular structure, which enhances the thermodynamic and kinetic stability of the oleogels system.[20]

Applications of Oleogels

Oleogels are gaining popularity due to their potential to improve health outcomes and their versatility in different applications. Their ability to combine the benefits of oils with the structural properties of gels opens up new possibilities for innovation in multiple fields. some key areas where oleogels are making an impact.

1. Drug Delivery Systems

The use of oleogels as a drug delivery vehicle was quite limited due to their non-biocompatible nature. Recently, focus on the synthesis and construction of biocompatible structural agents for oleogelation has made them suitable for drug delivery.

1.1 Topical/ transdermal drug delivery system

Skin is the largest organ in the body, containing a large number of blood vessels that help in the transport of drugs. Generally, drugs are absorbed into the blood vessels by applying them topically in the form of ointments, creams, pastes, and gels. In gels, they have become more significant due to their simple absorption process through the skin layers.[11]

Olmesartan (OLM) oleogel formulations, composed of Tween 20, Aerosil 200, and lavender oil, improve the therapeutic efficacy and bioavailability of the drug and decrease OLM side effects. The oleogel formulation is considered a promising carrier for the transdermal delivery of OLM, as it can avoid its oral problems, increase therapeutic efficacy, decrease oral side effects, and improve bioavailability.[24]

Sorbitan monostearate-based oleogels for topical drug delivery have shown controlled release of metronidazole.[25]

Lecithin-based organogel is used as a matrix for transdermal delivery systems due of its ability to improve the transport rate of the bioactive agents apart from their proven long-term biocompatibility and low irritability potential. The aromatic tetra-amidines loaded in lecithin organogel reduce the tumor cell growth in mice xenografted with the highly tumorigenic cell line by transdermal administrations. The methyl nicotinate incorporated within lecithin gel showed almost complete percutaneous absorption in human models, inducing erythema in a short period.[23]

1.2 Parental drug delivery system

Oleogels can be used for long-term treatment because they can form a drug depot when administered through an injection. Tenofovir drug-loaded chitosan nanoparticles were formulated and spray-dried. The NPs were incorporated with the oleogel matrix and their stability was tested using non-newtonian rheological properties. More than 40-60% sustained release was observed compared to chitosan nanoparticles and standards.[26]

1.3 Ocular drug delivery system

Dexamethasone was loaded into oleogels that had ethyl cellulose as the gelator at a concentration of 10% (wt.%) in soybean oil, exceeding the drug's solubility limit, and then expelled from the syringe to form cylindrical rods for prolonged drug delivery. Drug release was evaluated in a buffer under sink conditions through intra vitreal Injections and showed that there is an increase in the release from 600-3000hour.[27]

Nanocomposite oleogel formulations for ocular drug delivery made up of groundnut oil (GNO), stearic acid (SA) and graphene oxide (GO). These oleogels are designed to enhance the corneal permeation of the ciprofloxacin. The mechanical stability of the oleogels significantly increased when the GO concentration was >0.015 wt%. The absorption of medication through the cornea doubled when the oleogel contained 0.05% GO. The developed oleogels exhibited strong antibacterial properties against E. coli and showed cytocompatibility against human mesenchymal stem cells. These results demonstrate the potential of nanocomposite oleogels based on GNO/SA/GO for ocular drug delivery applications.[28]

1.4 Oral drug delivery system

Jasmine floral wax and wheat germ oil-based oleogel for oral delivery of curcumin to assess the release of curcumin from the oleogel. An increase in the concentration of jasmine floral

wax resulted in the increased partitioning of the curcumin into the formulation. This affects the drug release and it comes in terms with Korsmeyer−Peppas model parameters of curcumin release. Oleogel provides satisfying results in in-vitro release. These findings could be important for the application of oleogels in the food and pharmaceutical industries for loading and delivering bioactive components.[29]

2. Nutraceutical Applications

Nutraceuticals are products derived from food sources that are claimed to provide additional health benefits beyond the basic nutritional value found in foods. Depending on the jurisdiction, these products may claim to prevent chronic diseases, improve health, delay the aging process, increase life expectancy, or support the structure or function of the body. β-carotene, lycopene, curcumin, docosahexaenoic acid (DEA), eicosapentaenoic acid, linoleic acid, linolenic acid, the tannins, flavanols, and phytosterols are a few examples of soluble in fat nutraceutical substances. By employing oleogels for controlled release, these substances will increase the therapeutic efficacy.[11] Policosanol and olive oil-based oleogel were created to give the nutraceutical component ferulic acid orally. Due to its antioxidant qualities, ferulic acid has potential uses in treating diabetes, Alzheimer's disease, cancer, and other degenerative disorders.[30]

3. Medicinal Applications

Oleogels and their composites can be used for the treatment of many diseases such as rheumatoid arthritis, osteoarthritis, Reiter’s syndrome, dysmenorrhoea, postoperative pain, pyrexia, and acute gout treatment.

Lecithin organogels act as potential drug delivery vehicles for many biologically active agents that can treat aging of cells.[31] Ointments based on oleogels have many advantages when compared to conventional ointments. For example, cyclosporine encapsulated in sorbitan monooleate gelator administrated orally displayed enhanced activity. In addition, oleogel derived from Eudragit L and S finds potential in rectal drug delivery. Hydrophilic vaccines can also be delivered by microemulsion-based oleogels. They produce a depot effect by slowly releasing the antigens.[23]

4. Lubricants

Lubricants are used to improve mechanical performance and lower friction between moving parts. Oleogels made from castor oil and ethyl cellulose, α-cellulose, and methyl cellulose

blends are treated as environmentally friendly lubricating greases with high thermal and mechanical stability. Oleogels composed of sorbitan, glyceryl monostearate, and various vegetable oils are biodegradable alternatives to lubricating greases. Using low-viscosity oils like rapeseed and soybean oils generate oleogels with high values for linear viscoelastic functions.[11]

5. Food Industry

The main objective of using oleogels in the food industry is to replace solid fats which have adverse health effects. These constituents of oleogels are rich in unsaturated fats which are healthier than solid fat. In recent years, oleogels are used in ice cream, chocolate, margarine, shortening, and meat products. Frankfurters made using an organogel emulsion with γ-sitosterol/β-sitosterol and sensory analysis showed no significant difference between sausages prepared with 10% sunflower oil gel compared to control sausages that were rich in saturated fatty acids.[32]

EC oleogel could also be applied to reduce the cooked frankfurter sausages instead of beef fat, it showed no significant difference in chewiness and firmness compared to the control product made with beef fat.[33] Rice bran wax and soybean oil-based oleogel for the production of healthy and very soft margarine and shortening the concentration of Saturated fatty acids (SFA)and Trans fatty acids (TFA).[34]

The addition of beeswax/sunflower oil-based oleogel significantly affects the rheological and printing properties of the potato starch-protein (PS-WP) system in 3D printing. An oleogel content of 30% was found to be the best for 3D printing. In addition, a printing temperature of 30°C, a printing speed of 40 mm/s, and a nozzle diameter of 1.2 mm were the optimal combined parameters for the best printing effect.[35]

6. Cosmetic Applications

The skin care products mainly are emulsion based which means they contain water and oil phases. In addition, there are still products with only an oil phase, this belongs to the oleogels category. They are recommended for skin problems or used as dermatological cosmetics.

Unlike emulsions, the skin will hydrate gradually when using oleogels. It reduces the transdermal water loss (TEWL). In addition to that, natural water-retaining substances like

e.g. urea, which also has antipruritic effects, can be integrated into oleogels. Unlike water-containing emulsions, there will be no problem here with the long-term stability of urea.[2]

The high lipid content products reduce skin roughness. Hence, oleogels are recommended for hand care products and general skin protection purposes. It is also recommended for the care of the lips for cold protection products as well as for the care around the eye where spreading formulations as well as such containing emulsifiers. Dry and cracked foot skin will become soft and smooth. They are also recommended for the supportive care of diabetic skin, perianal skin disorders, and decubitus oleogels can also be applied as a sun protection product provided that appropriate sun protection filters are included.[33]

CONCLUSION

Oleogels represent a promising advancement in the field of pharmaceuticals, cosmetics, and food industries, offering unique properties and versatile applications. Their non-crystalline, thermoreversible nature, combined with their resistance to moisture and microbial growth, makes them ideal for developing stable drug delivery systems. The ease of preparation, cost-effectiveness, biocompatibility, and biodegradability of oleogels further enhance their attractiveness for therapeutic use. With their ability to control and sustain the release of active pharmaceutical ingredients across various administration routes, oleogels provide a significant advantage over conventional gels. The comprehensive understanding of their composition, preparation methods, and potential applications highlights the transformative potential of oleogels in improving health outcomes and product innovation across multiple industries. As research continues to advance, oleogels are expected to play a pivotal role in developing next-generation formulations, ensuring enhanced efficacy and safety for consumers.

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Conflicts of Interest

The authors declare that they have no conflict of interest.

ABBREVIATIONS

API	Active Pharmaceutical Ingredient
LMWOG	Low molecular weight oleogelators
RBX	Rice bran wax
CRX	Carnauba wax
OLM	Olmesartan
GNO	Groundnut oil
SA	Stearic acid
GO	Graphene oxide
DEA	Docosahexaenoic acid
EC	Ethyl Cellulose
SFA	Saturated fatty acids
TFA	Trans fatty acids
PS-WP	Potato starch- protein
TEWL	Transdermal water loss

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