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OLEOGEL: VERSATILE NOVEL SEMI-SOLID SYSTEM IN PHARMACEUTICALS

[1]Shivani P Chavan, [2]Shivali M Khandarkar, [3]Aditya V Palandurkar, [4]Sanjana R Bali, [5]Dr.Shivshankar Mhaske

[1]Student, [2]Student, [3]Student [4]Faculty, [5]Principal
Satyajeet College of Pharmacy, Mehkar, India.

Abstract : Oleogels are a promising class of gel-like materials formed by dispersing oils or lipids into a gel network using gelling agents. As drug delivery systems, oleogels offer significant advantages in terms of biocompatibility, stability, and controlled drug release, making them highly versatile for a wide range of pharmaceutical applications. Oleogels can be tailored for various delivery routes, including topical, transdermal, and controlled-release formulations, offering unique benefits in drug absorption, patient compliance, and therapeutic efficacy.The structure of oleogels, composed of natural or synthetic oils combined with gelling agents such as fatty acids, surfactants, or polymers, provides excellent stability and high drug-loading capacity. These systems can encapsulate both hydrophobic and hydrophilic drugs, making them suitable for a broad spectrum of therapeutic agents. Oleogels also exhibit non-greasy textures, improved skin permeability, and prolonged release profiles, which are especially beneficial for transdermal drug delivery.The versatility of oleogels lies in their tunable properties, including viscosity, gel strength, and drug release rate, which can be customized to optimize the delivery of specific drugs. This adaptability makes oleogels suitable for diverse applications, ranging from anti-inflammatory creams to more complex drug formulations for chronic diseases.In conclusion, oleogels are a highly versatile and innovative drug delivery platform with the potential to enhance drug bioavailability, improve patient adherence, and offer more effective treatments across various pharmaceutical fields.Their ability to deliver a wide range of drugs through controlled and sustained release positions oleogels as a promising solution in modern drug delivery systems.

IndexTerms **- oleogels, oleogelators, drug delivery, stability of oleogels**

I. INTRODUCTION

Oleogel, also known as organogel, is a semisolid formulation made by gelling oils with suitable gelling chemicals called organogelators.[124]There are two forms of gels: hydrogels, which contain a lot of water, and oleogels.[38] Semisolid dose forms retain their shape when subjected to external force due to their flexible nature.[135]An oleogel can be thought of as a significantly diluted system that, despite its high liquid content, has solid-like properties. [128] Oleogels are created by heating, shearing, and chilling a hydrophobic environment, resulting in a three- dimensional (3D) network of crystalline particles, self-assembled fibers, or polymers that entrap the liquid oil.[143] Oleogels are edible oils that have been immobilized within a thermoreversible, three-dimensional network of organogelators.[17] Oleogels, or vegetable oil- based organogels, have the ability to deliver hydrophobic or hydrophilic medicines in a targeted manner. Oleogels are non-Newtonian, which means they have shear thinning qualities that allow them to be injected with syringes while still maintaining their shape once implanted.[18] Oleogels have solid-like rheological qualities and thus keep their shape after implantation, while the medication slowly diffuses into the vitreous.[19] Oleogels are semisolid materials made up of an amphiphilic molecule, also known as organogelator, and a hydrophobic liquid. Oleogel microstructures consist of either permanent rigid networks or transitory semiflexible meshes, both of which are thermoreversible.[38]Oleogels have numerous advantages, including excellent emulsifying stability, a very simple preparation procedure,

low cost, and the capacity to form at low concentrations.[50,103]Oleogels are divided into two broad classes based on their molecular architecture: low-molecular weight oleogelators (LMOGs) and high-molecular weight oleogelators (HMOGs).[60]

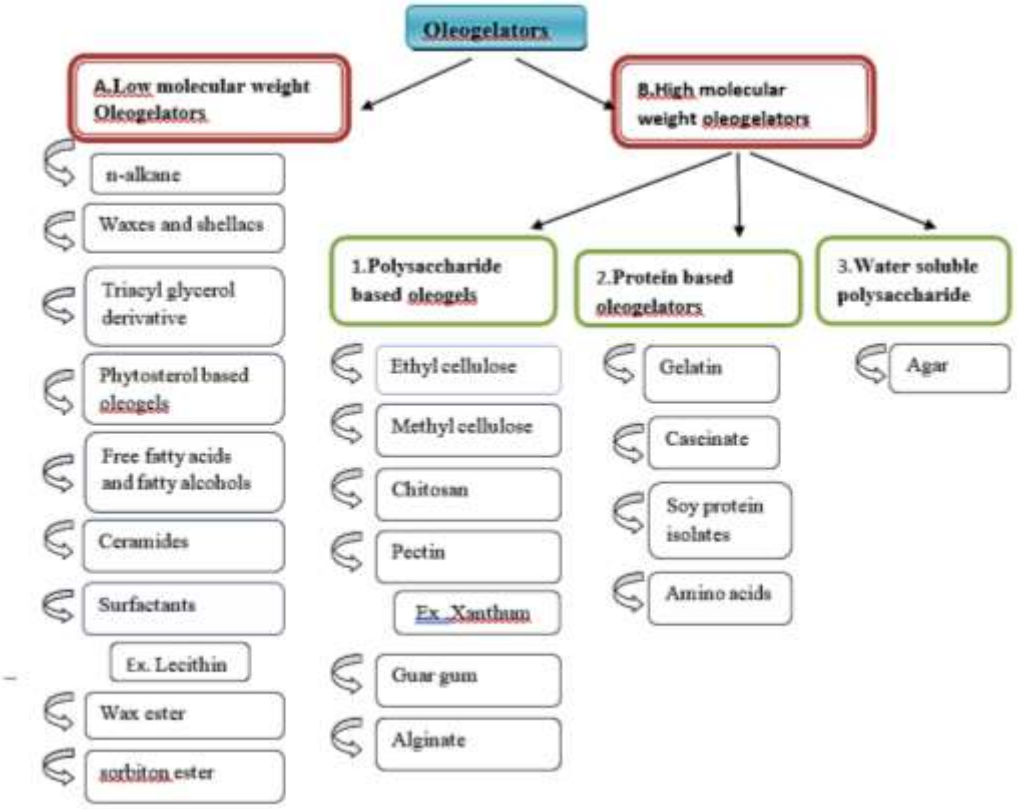

Fig1: Types of Oleogels

Saturated and trans fats add distinctive texture, oil binding, rheological qualities, and stability to traditional solid fats. [54,122] Oleogelators commonly include fatty acids, fatty alcohols, plant and animal waxes, sorbitan esters, phospholipids, ceramides, and phytosterols.[110] Organogels or oleogels are made up of a non-polar dispersion medium such as fixed oil, mineral oil, organic solvents, and so on, which is gelled using an agent known as an organogelator. [68] Gels are made up of three-dimensional fibrous networks of solid substances called gelators, which trap either polar (hydro-gel) or apolar (organogel) liquid phases. If the polar phase is vegetable oil, the organogel is referred to as oleogel.[64] The lipolysis of the enclosed hydrophobic molecules is significantly decreased. The reasons listed above can help explain why bioactive compounds are more stable when included into oleogels.[87] Sterol, sorbitan monostearate (Span 60), sorbitan monopalitate (Span 40), and derivatives of cholesteryl anthraquinone have been employed as organogelators in the creation of oleogels with various apolar solvents, including mineral oil, vegetable oils (such as sesame, rice bran, and sunflower oil), and organic solvents (such as cyclohexane, benzene, and carbon tetrachloride).[98] Compared to traditional distribution methods, organogels provide a number of significant advantages. [101] Numerous fields, including organic chemistry, environmental chemistry, pharmaceuticals, and cosmetics, have looked into the use of oleogels. [33] The organogels were stored at 5 ℃ for 24 h and then at 25 ℃ for 24 h before analysis, which was carried out at 15 ℃ to ensure solid condition at the time of analysis. [9] To make organogels, dissolve an organogelator in a heated (60-80 ℃) apolar phase then freeze to induce gelation. [130]The type of the gelling molecule will determine the ultimate features of the producing oleogels (for example, opacity, texture, melting temperature, and oil binding capacity). [106]The food sector has made extensive use of oleogels as oil migrating agents, fat blooming agents [128], for the delivery of hydrophobic and fat- soluble substances such vitamins and pharmacological moieties, and for cosmetic applications.

[46] Oleogels have been used for medicinal purposes such as topical wound healing. [139, 63] Oral mucosal administration. [127] Oral Delivery [6] Ophthalmic, the oleogel formulation comprises drug particles that serve as dispersed depots, resulting in longer release. [101,18] Topical.[67] Oleogels improve the solubility of hydrophobic drugs and have good rheological qualities. [111] Furthermore, because there is no water present, they are resistant to microbial infection and do not require the application of preservatives. [114,124] Oleogelation has been employed as a

developing and efficient physical strategy to convert low-viscous non-polar fluids into elastic solids, thanks to the formation of a 3D oleogelator network. [118]

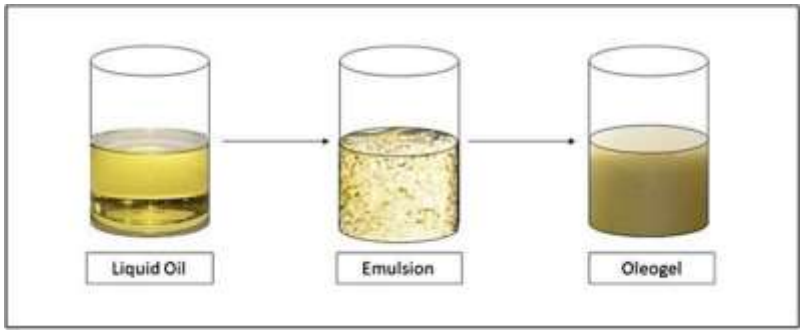

Fig2: visual difference between Liquid oil, Emulsion, Oleogel.

Thus, surface and capillary forces primarily cause a non-flowing situation, while the chemical properties of the liquid oil remain unchanged. [155]Additionally, rheological testing have been used to forecast product behavior and offer insights to enhance overall performance and stability. [152]Although stability tests are often carried out at constant temperatures, tests undertaken under periodically altered settings can uncover deficiencies faster than storage at a constant temperature. [49]The advanced rheometric expansion system (ARES) from Rheometric Scientific in the United States was used to characterize the rheology of the organogel. [100] Bicontinuous colloidal systems known as organogels coexist in liquid and micro- heterogeneous solid (gelator) phases. The organic liquid component is scattered with intricate networks of strands that make up the solid portion. [31] Oleogelators can be distributed but precipitate to form a network after gelation because they are soluble in the oil. [151]Another potential use for oleogels is the delivery of bioactive compounds, which has drawn interest from the pharmaceutical sector recently. [23] Although very few such formulations have been studied, organogels can be employed in the pharmaceutical industry to deliver drugs and vaccines via various administration methods. [158]

HISTORY AND PROSPECTUS

II. Oleogels first appeared in the food industry, when engineers created structural agents to solidify liquid oils. [135] Oleogels have promising prospects in the pharmaceutical industry as carriers for drug delivery systems. Oleogels hold significant promise for future usage in personal care and pharmaceuticals, making them an exciting subject for further research and development. [104] The oleogelation subject is of considerable interest worldwide: search the ISI Web of Science Core Collection Clarivate Analytics (13.11.2019) by the keywords combination "food" and "organogels" or "oleogels" » on Topic fields. [72] The first attempts to use organogels for easy drug administration dates back to the late twentieth century. So far, the Scopus database has identified 3615 publications with the keywords oleogel or organogel. They include 3214 research papers, 180 reviews, 66 books, and their chapters. The breakdown by country is as follows. China has 111 published articles, followed by Canada with 69, the United States with 65, Brazil with 39, Italy with 35, Spain with 34, Mexico with 31, Belgium with 25, Iran with 23 and Portugal with 23. [28] In order to overcome the current drawbacks of polymer gels, including their high molecular weight, critical gelation concentration, sol–gel transition temperature, biocompatibility, processability, and thermo-reversibility, researchers have been actively working on the development of self-assembled low molecular weight gels for efficient tissue regeneration and wound closure applications. [139] More recently, oleogels have also been suggested as effective means of delivering nutrients and bioactive compounds while regulating fat digestion. [47] They have acquired popularity in the food sector due to their capacity to mimic the functionality of fats and supply bioactive ingredients, providing potential solutions to a variety of difficulties and prospects for the development of novel food items.

III. [80] Recent oleogel research has focused on identifying new compounds or combinations of chemicals that can act as structural agents, as well as developing new gelation processes. This includes investigating many kinds of structural agents, such as waxes. [22,85] and polymers. [43,129] Scientists are also looking into the usage of novel techniques like micro- and nanostructuring. [108,153,88] to control oleogel characteristics and produce unique structures with specified functions. [9,88] since from the late 1950's oleogelation has been discussed. Because of wide range of benefits oleogels are used in food industry from last decades. [54] Oleogels and organogels are popular topics, and while the number of publications on oleogels has increased from 13 to 151 in the last ten years (search Scopus from 2011 to 2021 using the keyword "oleogel"), the number of publications on organogels has remained relatively constant, increasing from 105 to 152. [81] Oleogelation has recently developed as a new and effective approach for structuring liquid oils into solid-like systems, which improves product performance in terms of texture, sensory qualities, and shelf life in the food, pharmaceutical, and cosmetic industries. [119]

PURPOSE OF OLEOGELS

Fats and oils are mostly triglycerides, which contain monounsaturated, polyunsaturated, and saturated fatty acids, as well as a few minor chemicals. In most situations, food products contain a combination of these triglycerides. [136,137] Saturated and trans fatty acids play an important role in food technology since they are responsible for certain aspects like as flavor, palatability, and texture. [25] Furthermore, the triacylglycerols form a supracolloidal network, converting fats into solid or solid-like materials and adding structure to food goods. [14,72] Oleogels can be utilized for long-term treatment because they can develop a drug depot when administered via injection. They entrap the liquid oil through self-assembly, making hydrophobic medicines easily ensnared within them. As a result, it is appropriate for pharmaceuticals classified as BCS II or IV due to their low aqueous solubility. [40] BCS class II medicines like as atorvastatin, carvediol, and ciprofloxacin can be utilized in oleogel formulations to improve their efficacy by targeting solubility and direct absorption in the body. [135]Whereas for BCS class IV medications such as amphotericin, ciprofloxacin, and neomycin, the important properties to improve are solubility and permeability. Such medications can be efficiently mixed with the oil. [143]

II. MATERIALS AND METHODS

Materials

Oil

In this instance, oil structure results from the restricted solubility of high melting TAGs in the oil; as a heated solution cools, the TAG molecules separate from the liquid soup to form crystals that connect to form a network. [69] In order to create coconut oil oleogel (COO), high oleic sunflower oil oleogel (HOSO), and sunflower oil oleogel (SFO), oleogels were made with various oils. [50] Oleogels are oil-structured systems composed of a gelling ingredient added to edible oil. [143] Oleogels can be made from a variety of vegetable oils, and their physical and structural properties are closely connected to those of the lipid phase. [144] Oleogelation can be accomplished using a variety of gelators, including phytosterol-oryzanol combinations and 12- hydroxystearic acid, which can self-assemble into crystalline fibers, entrapping the oil. [90] The authors determined that oils with higher levels of saturation strengthen the oleogel. [13]

Oleogelators

The semi-solid medications known as oleogels, or organogels, are made by jelling lipophilic liquids using applicable chemicals called organogelators. [127] Oleogels, which are structured liquid oil painting matrices made with colorful gelatinizing agents (also appertained to as oleogelators or organogelators), similar as long- chain adipose acids, vegetable- grounded waxes, mono- and di- glycerides, alcohols, and phospholipids, may be a implicit remedy for the issues preliminarily mentioned. [62]Gelators should be" generally honored as safe" (GRAS) or deduced from natural sources for use in food or medicinal operations. [23] Organogelators must be safe, comestible, effective, extensively available, and nicely priced in order to satisfy abecedarian requirements. [53]Oleogels are systems that act solids and are grounded on the gelation of organic detergents (similar as oil painting ornon-polar liquid) using oil painting-answerable polymers or low- molecular-weight factors. These substances, appertained to as organogelators, produce a three- dimensional gel network that's thermoreversible and traps liquid organic detergents.[96] Using gelatinizing agents, the oleogelation process produces asemi-solid oil painting form. [97]oil painting structuring is grounded on the physical metamorphosis of dissolved gelators in a lipid terrain where the chemical parcels remain constant throughout the process. [44] Gelators that promote solid structure growth are classified into two types low molecular weight oil painting gelators (LMOG) and high molecular weight oil painting gelators (HMOG). [97] Polymeric organogels can deliver medicines in a pulsatile (on- off) manner in response to stimulants. So far, there have been many reports of stimulants-sensitive polymeric organogels. [154]Oleogelators are amphiphilic composites with a hydrophilic head and hydrophobic tail(s). An ideal oleogelator should be natural and biodegradable. [141]

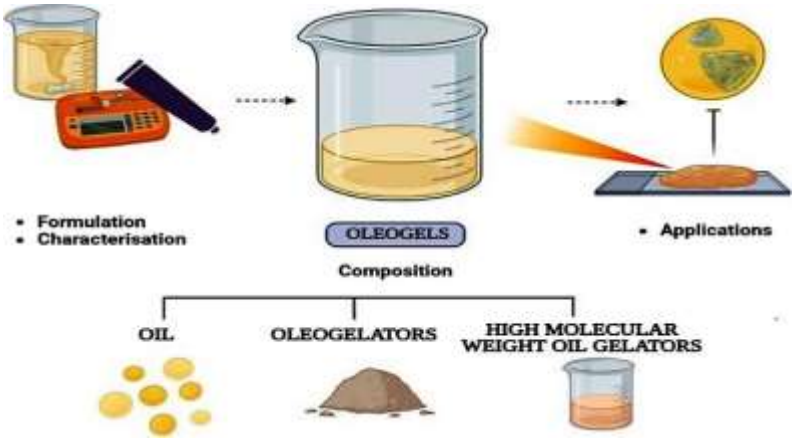

Fig3: Composition of Oleogel

A. Low molecular weight oleogelators

Low molecular weight organogelators (LMOGs) are appealing due to their thickness in the generation of organogels and tone- assembled fibrillar networks. [107] Low-molecular-weight gelators (LMWGs) (<3000 Da) such as waxes, monoglycerides, fatty acids, alcohols, Span 60, and phytosterol/oryzanol combinations are intensively investigated due to their ease of synthesis (direct dispersion), high gelling capacity, and availability. [77] It is worth mentioning that low molecular weight oleogels are typically made directly. Waxes are the most extensively investigated low molecular weight oleogelators, whereas EC polymer is the sole high molecular weight oleogelator employed in direct oleogel synthesis. [3]Oleogelators should normally have the following characteristics: (i) lipophilic and interacting parts, (ii) surface activity, (iii) thermoreversible features, (iv) natural origin, and (v) GRAS status. [92, 123]

1. n-alkane

N-alkanes, often known as paraffin, are saturated hydrocarbon chains composed of hydrogen- bonded carbon bonds. These compounds in organic solvents can form lamellar crystal formations. [135]Some of the most basic organogelators are n-alkanes. [31]An alkane hydrocarbon has the capacity to gel edible oils.[113] Modification of alkane chains with functionalized groups along the primary alkane chain was also proposed as a means of improving network stability and functionality. [102] Functional groups, such as the hydroxyl end group, can be introduced at any point along the carbon chain, including the end and the center.[132]

2. Waxes and shellacs

Waxes are lipophilic chemical compounds made of long alkyl chains (>16◦C) with functional groups including carboxyl, hydroxyl, ketones, aldehydes, and esters. [70] Waxes from diverse sources may differ in chemical composition, influencing melting and crystallization capabilities, gelation points, and crystal shapes. [79] According to a recent study, oleogel strength in wax organogels revealed a negative correlation with fatty alcohol concentrations and a positive correlation with wax esters, hydrocarbons, and free fatty acid concentrations. Therefore, a range of chemical classes can gel wax oleogels. [66] The wax esters, which make up over 75% of the mass of these waxes, are the ingredient that causes organogelation. These are aliphatic waxy esters with 44–64 carbons found in rice bran wax. [14]

Natural waxes (from plants and animals) are inexpensive and considered food grade. The most sought-after waxes for use in food applications are candellilla wax, carnauba wax, rice bran wax, beeswax, sunflower wax, and plant shellac wax/resin. Sunflower wax concentrations for oil structuring fall between 0.5% to 10%. [28] Natural waxes typically have a melting point ranging from 50 to 80 degrees Celsius. [70] Examples of the most thoroughly studied waxes are sunflower wax [79], rice bran wax [45], carnauba wax[12], bees wax[71], candelilla[15], berry wax[79], and fruitwax [117]. Waxes are chemically diverse compounds made of long-chain esters derived from fatty acids and alcohols. They are readily available and can shape oil at low concentrations (0.5-4% w/w), and many waxes are agricultural byproducts. [148] Furthermore, wax-based oleogels have proven to be extremely effective because, even at low concentrations, they can crystallize into a well-formed network with significant oil-binding capabilities. [65] Beeswax, a crystalline solid with a distinct honey aroma, is generated by Apis mellifera, Apis cerana, Apis florae, and other bee species for honeycomb production. [95,88] Apis mellifera bees produce two types of wax: yellow and white beeswax. [88]

Shellac-Chemically, it is a complex blend of polar and nonpolar components, including polyhydroxy polycarboxylic esters, alcohols, acids, and alkanes. Shellac, as a complex mixture of polar and nonpolar components, is amphiphilic and tends to self-assemble into supramolecular structures depending on the solvent characteristics. [48] The rate at which shellac crystallizes is heavily influenced by chilling speed. [135] Shellac, a natural resin derived from the secretions of the lac bug Laccifer Lacca, has long been used in pharmaceutical, food, and cosmetic applications. [120]

3. Triacyl glygerol derivative

The majority of oil-structuring solutions rely on low-molecular weight oil gelators, which replicate triacyl glycerides' (TAG) inherent propensity to self-assemble and crystallize into an orderly fat network. [133] Increases structural power and oil loss (Blake & Marangoni, 2015). Fully hydrogenated oils, commonly known as hardfats, are low-cost, readily available substances with a homogeneous composition primarily made up of saturated high melting point triacylglycerols. [8] When high melting TAGs in hardfats are utilized alone, at least 10% is required to generate an oleogel. [51] Low-molecular weight oil gelators, which replicate the natural capacity of triacyl glycerides (TAG) to self-assemble and crystallize to form an ordered fat network, are the foundation of the majority of oil-structuring techniques. The crystal lamellae structures that make up this network are known as nano-platelets. [34] The ultimate network structure and features are caused by larger-scale crystal clusters that are formed by the interaction of these self-assembling one-dimensional stackings. [123] TAG molecules crystallize into a network of crystals that physically bonds the liquid oil inside through a temperature- induced process. [133]

4. Phytosterols based oleogels

Phytosterols are recommended for oleogel manufacturing due to their natural and healthful properties. Unfortunately, the oleogel created with phytosterol alone (e.g., β-sitosterol, the most abundant phytosterol) is unstable, causing sitosterol crystals to aggregate and settle to the bottom. [160] Plant sterols, a type of steroidal chemicals found naturally in plants, are free of saturated fats and have a cholesterol-lowering impact. [5] Phytosterols have been found to offer a variety of health benefits, including lowering plasma levels of low-density lipoprotein (LDL) cholesterol. [24]

5. Free fatty acid and fatty alcohol

The development of mixed crystals between the two fatty components, which results in a small crystal size and nearly complete crystallization of the two fatty components in oil (high solid fat content), is directly related to the improvement of oleogel characteristics. [20] The length of the fatty alcohol chain and the cooling rate used during crystallization have a significant impact on the structure and physical characteristics of fatty alcohol oleogels. It turned out that fatty alcohols were a highly effective stabilizer for peanut oil. [122] Combinations of fatty acids and alcohols with appropriate chain lengths demonstrate the capacity to form distinct edible oils. Mixtures of fatty acids and alcohols with the same chain lengths, at 5% (w/w) in sunflower oil, demonstrated a synergistic effect below 20 °C at composition ratios of 7:3 and 3:7 (w/w). [57]

6. Ceramides

Creating ceramide-based oleogels has various health benefits, including good moisture retention capacity, [94] preventing colon cancer, modifying cell development, differentiation, and programmed cell death. [142] Ceramide has recently been discovered to be capable of self-assembling into fiberous crystals, resulting in a 3D network caused by hydrogen bonds that entraps vegetable oil. [94] Ceramide, the primary lipid species of the stratum corneum, is a kind of molecule in which a sphingosine is connected to a fatty acid via an amide bond. Ceramide is widely used in skincare products due to its exceptional moisture retention ability. For example, dietary CER has been linked to the ability to inhibit colon cancer. [93]

7. Surfactants

The impacts of surfactant addition to EC- grounded oleogels were delved in terms of the chemical composition of the surfactant's" head" and" tail" groups. Four distinct surfactants with similar chemical structures were studied sorbitan monostearate(SMS), sorbitan monooleate(SMO), glycerol monostearate(GMS), and glycerol monooleate(GMO).[58]Structurants can take numerous forms, although they're frequently classified as crystalline patches, polymeric beaches, flyspeck- filled networks, and liquid crystalline mesophases. [146] Amphiphilic chemicals have been set up to structure waterless and organic mediums, performing in gels with colorful characteristics. Surfactants, for illustration, are amphiphilic motes that have the eventuality to tone- assemble into colorful structure blocks. Surfactants can tone- assemble into colorful structure blocks, including globular micelles, spherical micelles, vesicles, and lamellar structures. [149]In surfactants, hydrophilic halves engage via hydrogen cling, while hydrophobic halves interact with organic liquid oil painting. These structure block infrastructures were dissembled by calculating critical quilting parameters(CPPs). [147]

Ex. lecithin :-

A vital element of all living cells, lecithin is a lipid patch with a special molecular structure that enables it to carry out a variety of tasks. Because of its emulsifying, density- modifying, stabilizing, solubilizing, and penetration- enhancing rates, lecithin is used considerably in the food, medicinal, and ornamental diligence. [89]Along with other composites like triglycerides and adipose acids, it's made up of a complex admixture of acetone- undoable phosphatides, substantially phosphatidyl choline, phosphatidyl ethanolamine, phosphatidyl serine, and phosphatidyl inositol. [157, 89]Lecithin is generally attained from oil painting- bearing seeds similar as soybeans, rapeseeds, and sunflower kernels, with soybeans being the most studied source. [55, 156] Beast sources of lecithin include egg thralldom and marine creatures, with egg thralldom being high in content and marine creatures known for their high phospholipid composition. [61]Lecithin is a naturally being phospholipid combination that's considerably employed in a variety of operations, including beast and mortal diets, specifics, and

cosmetics. [41] Lecithins have long been employed as a surfactant and demitasse habit modulator (changing crystal clear morphology according on food matrix, physical, and colloidal state). [42]still, because of their water perceptivity, lecithin oleogels have limited use in food. [116] From a medicine delivery perspective, lecithin organogels(LO) are particularly intriguing systems due to their biocompatibility, amphiphilic nature, enabling dissolution of numerous medicine classes, and saturation enhancing capabilities. Lecithin's amphiphilic nature allows it to take on a variety of forms, including mono- and bimolecular flicks, vesicles, liquid chargers, mixes, and, most importantly for this review, organogels. [158]

8. *Wax esters*

Sunflower wax and rice bran wax are considered single-component waxes since wax esters account for about 90% of their total makeup. [88] The oleogelation method is an emerging technology that has been extensively researched using plant wax esters such as beeswax, carnauba wax, candelilla wax, sunflower wax, and rice bran wax, among others. [39]

9. *Sorbitan ester*

Lecithin, sorbitan esters, polysorbate, and mono-and diglycerol are a few emulsifiers that are frequently utilized in the food business. With a hydrophilic–lipophilic (HLB) value of 4.7, sorbitan monostearate (Span 60) is a lipophilic emulsifier that is suitable for use in food. It gives food items their mouthfeel and softness and is a component of several readily accessible shortenings. [84] Sorbitan esters, often known as spans, are biodegradable nonionic surfactants that have fatty acid chains as the hydrophobic groups and sorbitol as the hydrophilic headgroup.They are frequently employed as emulsifying agents in the food, cosmetics, and pharmaceutical sectors. [162]

B. High molecular weight oleogelators

These are high molecular weight structures, such as proteins and polysaccharides, that may trap oil by using hydrogen bonds to create a three-dimensional network. The viscoelastic characteristics of oleogels made with polymeric oleogelators are significantly impacted by the molecular weight, concentration, and conformation of the polymers. [13]

1. *Polysaccharide based oleogelators*

a. Ethylcellulose

Because of its hydrophobicity, ethylcellulose has been found to be an effective oleogelator by direct dispersion, either alone or in combination with surfactants. [73] Ethylcellulose (EC) is the only known food-grade polymer-type oleogelator that is derived from natural sources and has a high gelling capability. However, oleogels stabilized simply by EC were inhomogeneous and had a low oil retention capacity. [76] EC is a linear polysaccharide derived from cellulose by replacing some of the functional hydroxyl groups on the repeating glucose units with ethylene groups along the polymer backbone. [77]

b. *Methylcellulose*

When creating oleogel systems, hydrophilic substances like hydroxypropyl methylcellulose (HPMC) and methylcellulose (MC) can be employed as structuring agents. Because of the hydrophobic substituents on its cellulose framework, MC has a naturally surface-active nature.

[11]

c. *Chitosan*

Chitosan is a biopolymer with favorable health qualities derived from the deacetylation of chitin, a naturally occurring polymer similar to cellulose. Chitosan is a promising oleogelator that has been successfully used in the creation of oleogels; however, due to its limited solubility in oil, oleogels must be produced indirectly. [105]

d. *Pectin*

Pectins are commonly derived from citrus fruit peels and albedo, apple pomace, banana peels, and sugar beet pulp. Pectins have useful applications due to their low toxicity and distinctive features, particularly in the food industry, where they are commonly employed as gelling agents, thickeners, emulsifiers, and stabilizers. [88]

Ex. Xanthum gum

Xanthan gum is largely answerable in both cold and hot water. When dissolved in cold water at low attention, it generates high-density results(from 100 to millions of mPa•s) with a weak gel character, stability across a wide temperature range, and pH 3- 10. Because of these characteristics, it's employed in the culinary sector as a thickening or suspending component in dressings, gravies, quick goodies, dairy and confection products, and fruit authorities. [88] Xanthan gum was employed to stabilize the polymeric network, which is an oleogel made of sunflower oil painting, gelatin, and xanthan gum. [97]

a. *Alginates*

Alginates are natural polysaccharides that are becoming increasingly popular in the food and pharmaceutical industries due to their physicochemical and health benefits. Because of their outstanding physicochemical qualities, alginates have been effectively employed to create oleogels. High molecular weight alginate (ALG) (350,000-500,000 Da) was combined with sodium caseinate (SC) protein to stabilize the structure of the resulting oleogel. [97]

g. Guar gum

Furthermore, in addition to XG, additional polysaccharides such as guar gum (GG) and locust bean gum (LBG) could be utilized as thickeners. [109]

2. Protein based oleogelators

In addition to polysaccharides, proteins can be used, albeit their potential as an oil structuring agent has received little attention. Protein may be more fascinating than other structural agents that have already been extensively researched in numerous ways: the nutritious benefit of proteins is widely recognized, consumers embrace them, and they are label-friendly. Such approved additives may improve the utilization of structured oils, or oleogels, in a variety of food applications. [73]

a. *Gelatin*

Gelatin/xanthan gum-based oleogels have a hardness range of 0.4e3.0 N and a high stiffness (G0 > 10.0 kPa). [141]

b. *Caseinate*

In some situations, such a stable interface was already achieved using protein-only produced oleogels containing 2% sodium caseinate. [73] The primary originality of this strategy is the use of emulsions as a template, with a combination of CNC (Pickering stabilizer) and sodium caseinate (surface active) allowing for room temperature drying. [16]

c. *Soy protein isolate*

Soy protein isolate, a plant-based protein, was used in this work to develop oleogel due to its amphiphilic characteristics and use as a food emulsifier. Chemical, physical, and enzymatic changes can improve soy proteins' emulsification abilities. The addition of polysaccharides is one technique to improve the emulsifying activity and stability of the emulsion. [125] Soy protein isolate (SPI) is commonly utilized to protect physiologically active components and as the wall material for oil microcapsules. [10] Because of its outstanding gelation capabilities, soy protein is regarded as a viable choice for vegetable protein manufacturing. It has been frequently utilized as a high-quality animal protein alternative since 1990. [88]

d. *Amino acids*

Amino acids start interacting with one another when the proteins unfurl, generating cross-links and aggregates that form a three-dimensional network. This network creates a gel-like substance by trapping water and other chemicals. [88]

3. Water based polysaccharides

Polysaccharides are among the most prevalent macromolecular polymers in nature, with sources including algae, plants, animals, and microbes. Hydrophilic polysaccharides, for example, can be successfully used in the formulation of emulsion-based foods like oleogel.

Ex. Agar

Agar is an interesting polysaccharide that could one day be utilized to structure oils and so make oleogels. This polysaccharide is derived from kelp of the Gelidiaceae and Gigartinaceae families. It is composed of two monomers: agarose and agaropectin. As a result, it is frequently utilized in the cosmetics sector to produce fat-free creams and masks since it increases the lubricity and adherence of these products. Agar also functions as a thickener, stabilizer, and emulsifier. [97]

III.Methods of preparation of oleogel

Oleogels are formed by dispersing a structural agent into an oil phase using a variety of processes such as hot or cold homogenization, solvent evaporation, and melt mixing. Each approach has distinct advantages and is chosen based on the nature of the structuring agent and the desired qualities of the finished oleogel product. [135]

1. *Homogenization*

Hot homogenization unites the oil phase with the oleogelator, the material responsible for the gel network, at temperatures over their melting points. This helps to disperse oleogelator molecules of particles throughout the oil phase. [13] The mixture is next homogenized, either using a high-speed blender or a high-pressure homogenizer, to provide substantial shear pressures. [80] This step breaks up any aggregates and ensures that the oleogelator is evenly distributed throughout the oil phase, resulting in a stable and homogenous gel network. [35] Unlike hot homogenization, which involves heating the combination, cold homogenization combines the oleogelator and the oil phase at or below ambient temperature. Cold homogenization is frequently favored when working with temperature-dependent oleogelators or when the desired gel structure is better accomplished at lower temperatures in the final product. [29] After homogenization, the liquid cools, causing the gel network to form as the temperature drops. [112]The oil phase (40% of the emulsion) was added to the aqueous phase and homogenized using a homogenizer at 10,000 rpm for 5 minutes. After homogenization, the emulsion was cooled to +4°C and stored overnight until use. [27] Oil-in-water (O/W) emulsions comprising 50 wt% canola oil were produced with 1 and 4 wt% CPI using six passes of high- pressure homogenization at 20,000 psi. [56] During homogenization, fat particles are broken down, resulting in stable homogenous emulsions of liquids that can form suspensions. [36] Oleogel-in-water emulsions were created using two separate emulsification methods: hot and cold. The first involved high-pressure homogenization of EC-oil and water at a temperature over the gelling point of EC, whilst the latter entailed dispersing set EC-oleogels in water via high-speed mixing at a temperature below the melting point of EC oleogels. [29]For fifteen minutes, the prepared oleogel combination solution was further homogenized at 500 bar. Following homogenization, the mixed solution was promptly cooled to 4 °C in an ice bath. [91]

2. *Solvent Evaporation*

The solvent evaporation technique entails dissolving the oleogelator in a suitable solvent before blending it with the oil phase. The solvent is removed by carefully regulating the mixture's evaporation, leaving the oleogelator. Various oil structuring techniques are currently in use and spread across the oil phase. [13] This approach is especially useful when working with oleogelators that are difficult to dissolve in oils or when careful control of the gel structure is required. [115]

3. *Melt blending*

Melt blending entails rapidly combining the oleogelator and oil phases at a temperature above the oleogelator's melting point. However, it may not be appropriate for all oleogelators, particularly those that are prone to structural breakdown or alterations at high temperatures. [26] This can be accomplished using an appropriate heating technology, such as a water bath or a heating mantle. It is critical to keep the temperature above the gelator's melting point while remaining below the temperatures at which oil and medication deteriorate. [68] The most typical approach for oil gelation is to disperse the high-melting gelator directly into the oil at a temperature above its melting point, then cool to cause the gelator to crystallize or self- assemble into a network structure that traps the oil. [145]

HYBRID OLEOGELS

The so-called hybrid gels, also known as bigels, are created when hydrogels and oleogels are combined. These gels have both hydrophilic and lipophilic properties. Biphasic systems known as hybrid gels combine oil-based gels (oleogels) and water-based gels (hydrogels). Like hydrogels and oleogels, the usage of hybrid gels in food items must be concentrated on their integration in intricate food systems. The potential of these features can also be increased by adjusting the formulation and combining different materials. [1] Using a mechanical mixer set at 600 RPM for 45 minutes and a 24-hour equilibration period at room temperature prior to each analysis, oleogel and hydrogel fractions were combined in various ratios to create hybrid gels (HGs). [30] Hybrid oleogel formulations made up of two different kinds of gelator molecules take use of synergistic effects, which may improve the functionality and performance of the oleogel. Furthermore, control problems can occasionally limit

gelator concentration, which can be resolved by combining gelators. By combining HMOG and LMOG, hybrid oleogel takes use of the variations in the physical characteristics of these gelation processes. [60]

MECHANISM OF GEL FORMATION

The interfacial characteristics of oil bodies are known to be significantly influenced by oleosins, which are embedded in phospholipid monolayers. The oleosin molecule's hydrophobic core domain extends inward into the interior of the triacylglycerol and the hydrophobic acyl interior of the phospholipid, forming an 11 nm stalk-like structure. [83]

CHARACTERIZATION OF OLEOGELS

Oleogels are evaluated by a variety of tests and analyses in order to understand their physical, chemical, and functional properties. [135]

1. *Organoleptic characters*

The consistency, roughness, grease, color, texture, and physical appearance of the formulation are assessed. The oleogel is checked visually as well as by applying it to the skin and rubbing a small amount between fingers. The test is performed on oleogels to determine the sensory qualities and physical properties that influence their acceptance and performance. [52]

2. *Gel- sol transition*

The sol-gel transition refers to the development of bonds between particles or molecular species, which result in a three-dimensional solid network. This transition turns a sol, or colloidal solution, into a gel. [126] Stable oleogels must retain their gel structure during processing and storage to avoid phase separation or syneresis. Understanding the parameters that influence the gel-sol transition, such as choosing optimal gelling agents and formulation variables, makes it possible to improve the long-term stability of oleogel-based drug delivery systems. Managing drug release rates is critical for the gel-to-sol transition in drug delivery systems like Oleogels. [135]

3. *PH determination*

The pH of olmesartanoleogel formulations was measured using a pH meter (Thermo Scientific, Waltham, MA, USA). The pH meter was calibrated using a standard buffer solution before use. The research was conducted three times. The mean value ± SD was then calculated. [124]The pH of oleogel ranged from 6.29 to 6.48, matching the normal pH of humans. [99]

4. *In vivo study*

The preclinical test in rabbits had three goals: (i) to assess the safety and tolerance of the aqueous (niosomes and micelles) and non-aqueous (oleogel) formulations; (ii) to track the amount of epalrestat in the tear fluid; and (iii) to quantify the buildup of epalrestat in the various eye tissues. [138]

5. *Texture analysis*

The texture of pharmaceutical formulations, whether given topically or orally, has a considerable impact on their ease of use. Texture analysis allows for the optimization of formulation parameters to achieve the required application qualities, which promotes patient adherence to treatment regimens. Texture variations can indicate melting, crystallization, or gelation phases in oleogel compositions. [135] Overall, edible oleogels' textural qualities can be customized by carefully selecting the type and quantity of structural agents, as well as adjusting the processing conditions, to obtain the necessary functionality and sensory features. Texture analyzers and X-ray diffraction are two types of texture equipment that can be used to determine texture qualities. For qualitative examination, microscopes are employed. [88]

6. *Rheology of oleogels*

An Anton Paar MCR 301 rheometer (Anton Paar, St. Laurent, QC, Canada) was used to measure viscosity with a serrated plate geometry (PP 25/P2) with a diameter of 25 mm. [159] Rheology and tribology have been used to better understand the relationships between structural elements during multiscale deformation. On the other hand, rheo-tribological data can be used to replicate fat processing patterns in the mouth. Typical W/O emulsions demonstrated shear-thinning behavior and primarily elastic features, with surfactant type and content having a significant impact on rheological parameters. [76] It should be observed that when a shear rate was applied to all samples, the initial shear stress was similar. It is

feasible to see that the apparent viscosity of all samples reduced with increasing shear rates, indicating shear-thinning behavior, which is typical of non-Newtonian fluids. [86]

7. *Polarized Light Microscopy*

The internal structure of the oil gel was analyzed using both polarized light microscopy and bright field sample views. [32] Images were processed using Motic Image Advanced Software after seeing samples at × 200 magnification. [140] Polarized light microscopy is the most often utilized approach for examining the oleogel microstructure due to the fat crystals' birefringence. Although it is a widely used method for identifying network formation in oleogels, the trouble stems from micrometric measurements. [88] Polarized light microscopy was used to analyze the microstructure of oleogels. A drop of prepared oleogels was gently placed on a hot slide and carefully covered with a glass cover slip. After cooling at ambient temperature for 40 minutes, the samples were stored at -20°C, 5°C, and 25°C for further testing. [2]

8. *X- ray Defraction*

The crystalline structure of oleogel samples, CW, and AA powders was studied using an x-ray diffractometer (D5000; Siemens, Munich, Germany) with a Cu Kα source (λ=1.54 Å) at 40 kV and 30 mA. The X-ray diffraction patterns were examined using the X'pert High Score Plus software.[21] XRD is a technique that provides detailed information about the packing arrangement and crystallite size of wax crystals in the non-polar oil phase. [87]

9. *Fourier Transformed Infrared Spectroscopy*

Oleogles' molecular structure was studied using FTIR (Thermo Nicolet, USA) with a wave number range of 4000-1000 cm−1, a notional resolution of 1 cm−1, and 64 scans. [150] To investigate the changes in crystalline structure caused by hydrogen bonding in 15% MGs oleogels, the samples' infrared spectra were acquired at 4000-400 (1/cm) wave numbers with

an FTIR spectrometer. Samples (5 mm × 5 mm x 5 mm) were put on the 3-reflection diamond ATR plate and squeezed with a swivel tip. All spectra were taken against an air background prior to the MGs oleogels investigation, and 5 FTIR spectra were collected for each sample.[2]

10. *Diffirential Scanning Colorimetry*

The melting behavior of oleogels was studied using differential scanning calorimetry on a PL DSC-gold calorimeter. [2] Differential Scanning Calorimetry was utilized to study the thermal behavior of oleogels. Samples weighing 5-10 mg were cooked in a sealed aluminum pot at temperatures ranging from 0 to 90 ◦C using a nitrogen flow rate of 50 mL/min at 5 ◦C/min. An empty sealed aluminum pan served as a reference. [150]To evaluate thermal behavior, 5-10 mg samples were placed in a differential scanning calorimeter cell, with an empty pan serving as a reference. The oleogel samples were heated from 0 to 120 °C, and the margarine samples from 0 to 80 °C, with a temperature increase of 5 °C per minute. The melting temperature was calculated using the DSC thermogram. [37]

11. *NMR Spectroscopy*

NMR spectroscopy is a sophisticated method of characterisation that can determine a sample's molecular structure at the atomic level. NMR spectroscopy, which is widely used in pharmaceutical research and development, may detect a wide range of properties including phase changes, solubility, conformational and configurational changes, and diffusion potential.
[135] Oil binding is defined as a shift in the oil's molecular mobility—either in translational or rotational dynamics—as reflected in difusion and transverse NMR relaxation. NMR-transverse relaxation and difusion are notable for their ability to evaluate both intramolecular and translational mobility. [134]

12. *Liquid Chromatography – Mass Spectrometer*

LC-MS is a frequently used analytical technique that combines mass spectrometry (MS) and liquid chromatography (LC). Because of its increased sensitivity, contemporary MS has replaced many immunoassays with LC-MS. The increased sensitivity and specificity of LC-MS has improved the efficiency of drug discovery operations. [128] The drug's stability in chitosan nanoparticles created by spray drying was evaluated using Electrospray Ionization- Mass Spectroscopy (ESI-MS) techniques. [6]

13. *HPLC*

One of the most important analytical methods for measuring and identifying bioactive compounds is still HPLC. It is especially useful for evaluating the chemical makeup, stability, and quality of oleogels. [59]

14. *ULTRA VIOLET VISIBLE SPECTROSCOPY*

Oleogels and their component parts are frequently structurally analyzed using UV-visible spectroscopy. This non-destructive analytical method uses molecules' ability to absorb UV and visible light to reveal information on the stability, concentration, and chemical makeup of the materials that make up oleogels. UV-visible spectroscopy can be used to track the stability of oleogel over time. Possible oxidation, degradation, or other chemical changes in the oleogel composition can be indicated by changes in absorption spectra, such as variations in peak position or intensity.[135]

ADVANTAGES OF OLEOGELS

The use of edible oleogels is a unique and promising technology for decreasing saturated animal fat and increasing the nutritional profile of food products. Oleogelation has been proposed as an alternate approach for using liquid vegetable oils to create hardstock structures that mimic the properties of traditional fats. [2] Oleogels contain or fix liquid oil in a three- dimensional network structure, and the sol-gel process is thermally reversible. [3] Shellac wax (SW) has sparked widespread interest in the creation of oleogels for the food industry. [4] Oleogels can deliver hydrophobic and hydrophilic chemicals for the long-term therapy of a variety of ailments. [6] Oleogels have showed potential in culinary applications as replacements for traditional fats. [23] It must be food-grade, economically viable, efficient, adaptable, and compatible with the end product's physical qualities. [8]

DISADVANTAGES OF OLEOGEL

- Stability problems associated with oleogel –

Oxidation is a major source of quality loss in oil-based meals containing unsaturated fatty acids. [5] The capacity to bind oil, as well as oxidative and thermal stability, all contribute to oleogels' stability. Oil binding capability is related to network organization and gel mechanical resistance. [78] Oxidative stability is a challenge due to the large presence of unsaturated fatty acids, which promote oxidative reactions; nonetheless, there is evidence that the oleogel network can shield liquid oil from oxidation, delaying breakdown. [144]Oleogel stability was determined using a centrifugal approach based on the method of Oleogel stability (SO) was estimated using the following formula:

$$SO = \frac{(Mg - Mo)}{(Mz - Mo)} .100\%$$

in which:

- Mg—oleogel and test tube weight after centrifugation [g];
- Mo—tube weight [g];
- Mz—oleogel and test tube weight before centrifugation [g]. [82]

APPLICATIONS OF OLEOGELS

1. *Oleogel in delivery of neutraceuticals*

Though numerous researchers have employed oleogels to transport vitamins, polyphenols, and volatile aromatic compounds, little study has been conducted on the delivery of nutraceuticals utilizing oleogels. Recently, a few studies have shown that certain oleogels are effective at delivering lipid-soluble (or hydrophobic) nutraceuticals. [87] Phytosterols and ceramides can both be classified as nutraceuticals. It would be ideal to employ a nutraceutical organogellator, however this is not always possible or practical. [161] Protein-

polysaccharide complexes, including Maillard-type and electrostatic polysaccharide conjugates, have the potential to stabilize oleogel-based nanoemulsions for nutraceutical delivery. [75]

2. *Oleogel in cosmetics*

Oleogel systems provide a distinct hydrophobic environment that can be used for the delivery of hydrophobic bioactive compounds. These properties can be applied in a variety of applications, including pharmaceutical therapeutic treatments, dietary supplements, and bioactive delivery in cosmetics. [60] Oleogels have several advantages for cosmetic applications, including increased physical and chemical stability of active molecules, improved rheological properties, lower-cost composition and manufacturing preparation processes, and improved and versatile delivery of lipophilic compounds through the skin. Other particular applications include decorative cosmetics such as makeup, eyeshadows, and mascara, as well as supportive care for diabetic skin, perianal skin diseases, and decubitus. [28]

3. *Oleogel as pharmaceuticals*

Oleogels are used in a variety of drug delivery modalities, such as transdermal, parenteral, topical, and oral. [135] Oleogel systems provide a distinct hydrophobic environment that can be used for the delivery of hydrophobic bioactive compounds. These properties can be applied in a variety of applications, including pharmaceutical therapeutic treatments, dietary supplements, and bioactive delivery in cosmetics. [60] Edible oil gels (oleogels) have sparked interest in recent years because of their potential use in cosmetics, meals, and pharmaceuticals. [23] Although several studies have been conducted on oleogel as a pharmaceutical delivery system in recent years, its exploration as a nutraceutical delivery system for edible purposes remains a research gap. When investigating the potential of oleogels for delivering lipid soluble bioactives, b-carotene (BC) is frequently employed as an excellent nutraceutical model. [7]

4. *Topical and transdermal drug delivery*

Soybean oil-based oleogels are more flexible, have superior drug release capabilities, and have lesser thermal stability, whereas olive oil-based oleogels were generated at lower gelator and co-gelator concentrations. [98] Organogels have gained popularity due to their capacity to build a crystalline network and entrap oils at low concentrations (<10% wt). Organogels, in addition to being easy to prepare, can improve medication penetration into the stratum corneum due to their lipophilic composition. [7,131] Olmesartan medoxomil (OLM) has a low oral bioavailability (28.6%) due to poor water solubility and oral issues such as the substantial hepatic first-pass effect and efflux pumps in the gastrointestinal tract, both of which interfere with medication absorption. Thus, the transdermal drug delivery system (TDDS) is a recommended alternative strategy for avoiding the drawbacks of OLM oral delivery while increasing therapeutic efficacy and bioavailability. [124]

5. *Ocular drug delivery*

The oleogels were tolerant of ocular tissues and had no harmful effect on mammalian cells. Finally, oleogels made from palmitic acid and safflower oil can be used to deliver drugs to the eyes. [110] Oleogel rods are effective devices for extending the delivery of ophthalmic medicines. Drug loading and partition coefficient can be used to alter release duration. [19] Oleogels improved corneal and scleral permeability while minimizing ocular discomfort and increasing in vivo ability to reach interior eye tissues. [138]

CHALLENGES AND FUTURE PROSPECTS OF OLEOGELS

The future manufacture of oleogels has potential in bioprocessing fermentation technologies and the utilization of oleogelators derived from renewable sources such as food processing byproducts. [74] Oleogels, a structured liquid oil matrix made up of various gelling agents (known as oleogelators or organogelators), such as long-chain fatty acids, vegetable-based waxes, mono and di-glycerides, alcohols, and phospholipids, may be a potential solution to the problems mentioned earlier. [62] Long-acting injectables are being investigated in order to achieve prolonged medication release. Tenofovir-loaded chitosan nanoparticles were prepared and spray-dried. The NPs were integrated into the oleogel matrix and assessed for stability using non-Newtonian rheological characteristics. [6]Oleogels are used in a variety of therapeutic domains, including ophthalmology, dermatology, wound healing, and pain management, to successfully deliver drugs to target regions regardless of whether they are hydrophilic or lipophilic. [135]

CONCLUSION

Oleogels are a promising and diverse class of semi-solid dosage forms in pharmaceuticals, providing several benefits in drug administration. Their unique ability to encapsulate both lipophilic and hydrophilic medicines, together with their adaptable mechanical properties, makes them ideal for a variety of applications, including topical, transdermal, and controlled release systems. Oleogels have good stability, biocompatibility, and ease of formulation, making them a desirable alternative to standard dosage forms such as creams, ointments, and gels. Additionally, adding active pharmaceutical ingredients (APIs) into oleogels has the potential to improve drug bioavailability and therapeutic efficacy. Oleogels are set to become a key component in the future of pharmaceutical formulations, particularly in the context of personalised medicine and targeted delivery systems.

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