

## Bigels: Paving the Way for Healthier and More Sustainable Food Innovations

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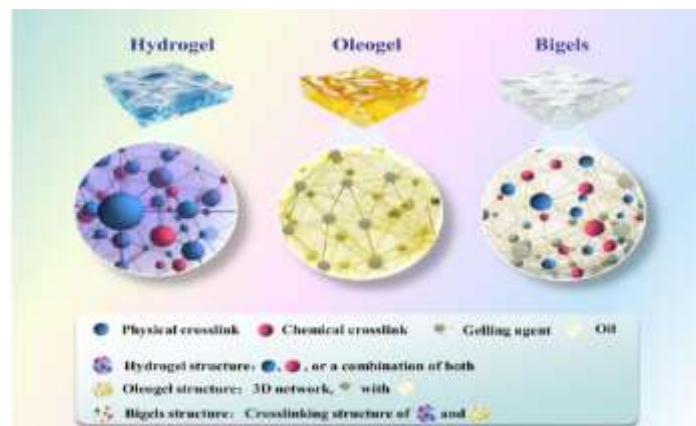
Growing awareness of food safety and the shifting attitudes of consumers have driven numerous innovations in the food system. One such emerging innovation is the development of bigels. Bigels, a novel class of biphasic gel systems, have gained increasing attention for their versatile applications in the food industry. These structured materials, formed by combining hydrogel and oleogel networks, offer unique physicochemical properties such as enhanced stability, controlled release of bioactive compounds and improved textural attributes. Their potential extends to diverse food applications, including fat replacement, encapsulation of nutrients and the development of functional foods with tailored sensory and health benefits. This exploration highlights the primary role of bigels in advancing food formulation, their advantages in improving product functionality, nutritional value and consumer appeal. Gels are classically defined as soft matter, typically composed of at least one component with low molecular mobility, known as structuring agents and another predominant compound usually a solvent. Gels can be categorized depending on the gelation of either polar or non polar liquid phases, resulting in hydrogels and oleogels, the latter being a subcategory of organogels.

Hydrogels, primarily consists of water and their network is formed through the crosslinking of biopolymer chains *via* chemical crosslinking with covalent bonds, physical crosslinking through non-covalent interactions, or a combination of both. Advantage of hydrogels it will respond to environmental stimuli, such as changes in temperature and pH and as ingredients to improve various food properties, including elasticity and stability during food product manufacturing. Disadvantage is nevertheless, hydrogels are not suitable for delivering hydrophobic compounds.

Oleogels, are defined as semi-solid mixtures containing an organic liquid phase trapped within a 3D network, achieved by adding a small amount of a gelling agent to liquid vegetable oil. Indeed, while oleogels offer several advantages, such as the ability to

restrict the flow of liquid fat and provide thermal reversibility, they do come with certain disadvantages such as softer texture and lower plasticity compared to traditional solid fats.

Bigels are also called as “Hybrid gels/biphasic gels” was first suggested in 2008 by Almeida and Co-workers. Bigels are innovative biphasic systems consisting of two gelled phases, typically formed through mechanical mixing under specific temperature and gel-setting conditions. Fabrication of bigels offers the advantages of both hydrogels and oleogels, including the capacity to deliver both hydrophilic and hydrophobic components. Different systems can be obtained by dispersing these two gel phases in different proportions, such as bi-continuous, hydrogel-in-oleogel, oleogel in hydrogel as similar to emulsion.



**Fig. 1. Schematic representation of hydrogels, oleogels and bigels**

Key properties of bigels encompass unique thermodynamic behaviour, texture, sensory properties, improved stability and versatility in preparation method. Main advantage of bigels is improved stability. Both phases in the gel network makes bigel stable for 6 -12 months, making it significantly different from emulsion gel. Besides, their versatility and unique structural properties bigels are having diverse application in the food system as fat replacer, intelligent food packaging, innovative 3D printing food production and controlled release of bioactive compounds.

**Table 1: Differences between hydrogels, oleogels and Bigels**

Property	Hydrogels	Oleogels	Bigels
<b>Definition</b>	Gels formed by water-based networks, often stabilized by polymers	Structured gels containing an oil phase trapped within a gel network	Hybrid gel system combining both oleogels and hydrogels
<b>Composition</b>	Water-based networks using protein, hydrocolloids or polysaccharides.	Oils structured using gelling agents (waxes, proteins, fatty acids, <i>etc.</i> )	Combination of oleogel (lipid-based) and hydrogel (water-based).
<b>Nutritional Benefits</b>	Low in fat, high in fiber or proteins	Typically high in fats, but healthier versions exist with unsaturated fats	Can reduce fat content while maintaining desirable texture and functionality
<b>Texture &amp; Rheology</b>	Gel-like, elastic, can be brittle or soft	Soft, spreadable, or firm depending on the structuring agent	Balanced properties of both oleo gels and hydrogels, with tunable firmness.
<b>Thermal Stability</b>	Can lose structure upon heating due to water evaporation	Highly stable under thermal conditions	Generally, more stable than hydrogels due to the presence of oleo gel.
<b>Application in Food</b>	Used in beverages, dairy products, gels and low-fat formulations	Used in spreads, chocolates, meat substitutes and cream-based products	Used for fat replacement, texture modification and stability improvement in various products, 3D printing, intelligent packaging <i>etc.</i>
<b>Advantages</b>	Provides hydration, dietary fiber and a light texture	Provides a fat-like texture without trans fats, can be made with healthy oils	Combines the benefits of both hydrogels and oleo gels, improving texture, stability and mouthfeel
<b>Disadvantages</b>	Can be fragile, lose moisture and have limited fat-like properties	Can be too greasy, less hydrating, and sometimes lacks structure	More complex to formulate and manufacture than oleo gels or hydrogels alone.

### Fabrication Process and Categories of Bigels

Preparation and types of bigels, which are structured systems combining hydrogel and organogel components (Fig. 2). Here's a breakdown of the key elements:

**1. Hydrogel Formation (Top Beaker - Blue):** Various biopolymers such as carrageenan, xanthan gum, alginate, hydroxypropyl methylcellulose, agar-gelatin blends, *etc.* Water as the dispersing medium. These components form a hydrogel, which is water-rich and gel-like in texture.

### 2. Organogel Formation (Bottom Beaker - Yellow):

Low molecular weight crystalline materials such as waxes, stearic acid,  $\gamma$ -oryzanol/phytosterol, glycerol stearate, sorbitan monostearate *etc.* Oil as the dispersing medium. These form an organogel, which is oil-based and gel-like.

**3. Bigel Formation Process:** Hydrogel and organogel are mixed using a high-speed homogenizer or mechanical stirrer. The resulting bigel's structure and properties depend on: ratio between oleogel and hydrogel, mixing temperature, stirring speed and gels proportion.

#### 4. Types of Bigels

- **Hydrogel-in-oleogel bigels** – Water-based droplets dispersed within an oil-gel network.
- **Oleogel-in-hydrogel bigels** – Oil-based droplets dispersed in a water-gel matrix.
- **Bi-continuous bigels** – A networked system where both hydrogel and oleogel phases are interconnected.



**Fig. 2. Fabrication process of bigel**

#### Classification of Bigels

In general, bigels can be categorized as Oleogel-in-hydrogel, Hydrogel-in-oleogel or Bicontinuous bigel (Fig. 3).

**1. Oleogel in hydrogel bigel :** Oleogel-in-hydrogel systems, where the hydrogel is the continuous phase and the oleogel is the dispersed phase, are possibly the most extensively researched bigel systems.

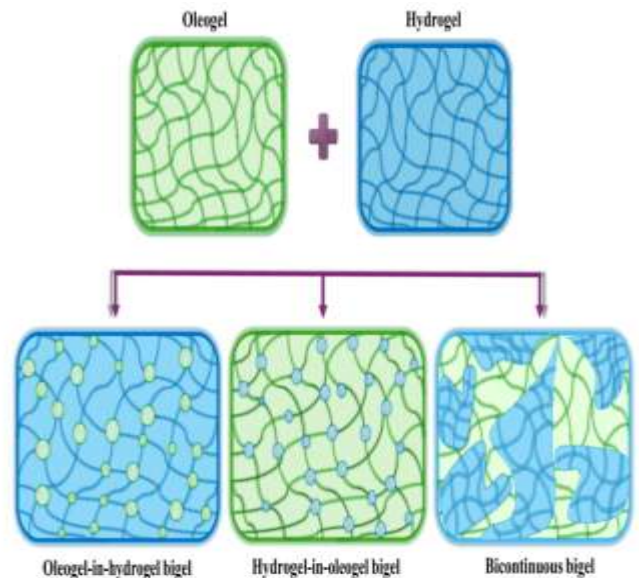
**2. Hydrogel in olegel bigel :** Second type of bigel, known as hydrogel-in-oleogel, comprises a system in which the hydrogel phase is dispersed throughout the continuous oleogel matrix.

**3. Bicontinuous bigel :** Bicontinuous-type bigels are considered as more complex metastable structures, where there is no discrete continuous or dispersed phase due to the co-existence of the two structured phases in the bigel matrix.

#### Factors Effecting on the Main Properties of Bigel

Bigels offer a wide range of properties and functionalities, which can be classified into different categories based on their characteristics. It is important to highlight that bigels produce an effective reduction of saturated fat and the modification of the lipid fraction of meat products. This is due to these gels being composed of a high-water amount (>50 %, m/m). Moreover, bigels improve oil and water

retention, preventing their release during cooking and thus, improve the texture and juiciness of the food product. Using bigels, manufacturers can create a more uniform texture while also improving the nutritional value of the final product.

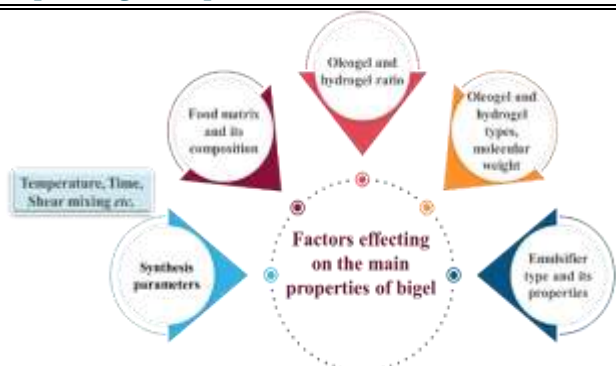


**Fig. 3. Classification of Bigels**

Factors affecting the mechanical properties of bigels mainly includes intrinsic characteristics, such as formulation adjustments and material combinations (*i.e.*, the proportion between oleogel and hydrogel, type and several gelling agents, gelling method, additives/emulsifiers, pH, temperature). Manipulation of phases (hydrogel/oleogel ratio, types of organogelators and hydrogelators) allows to affect the final properties. Furthermore, bigels with higher oleogel content have a lower shrinking and swelling rate. Additionally, sensory evaluations indicated high acceptability.

Oil holding capacity of bigel is another important physicochemical characteristic that is affected by protein levels of bigels. At low protein levels, bigels exhibit poor stability because they fail to form a sufficient network capable of retaining oil droplets. Moreover, the type of hydrogel that was used had an impact on the peroxide value and binding ability of bigels. These changes may be attributed to formation of more interconnected and denser network, more elastic response of bigels. Increased presence of fat crystals in bigels may result from the crystallization of monoglycerides within the oleogel phase. This increase, in turn, enhances the mechanical strength of the bigels.





**Fig. 4. Factors effecting on the main properties of bigel**

### Application of Bigels in Food System

Application of bigels in the food system is vast and diverse, encompassing various aspects of food production and innovation from serving as fat replacers to 3D printing (Fig. 5).



**Fig. 5. Application of bigels in food system**

#### 1. Bigels as a fat replacer

- Obesity, a global health issue, is often associated with high consumption of saturated fats.
- Bigels will highlights the need for healthier fat replacers which can be can be incorporated into different bakery products such as cream, burgers, cookies and chocolate to reduce saturated fat content.
- Bigels with higher ratio between oleogel and hydrogel were thermally more stable

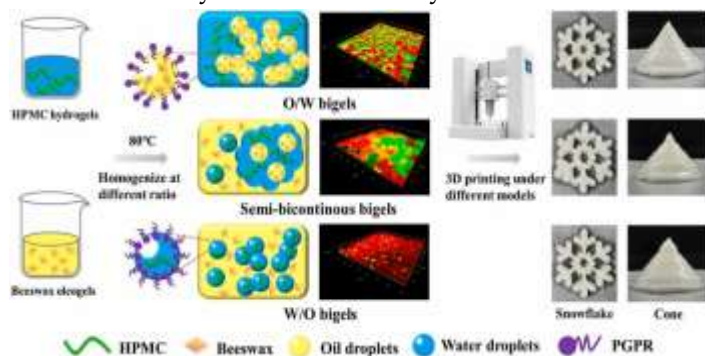
according to their greater viscoelastic behaviour.

#### 2. Bigels used in Intelligent food packaging

- Bigels have emerged as innovative materials in intelligent food packaging due to their ability to act as physical barriers, enhancing the stability of encapsulated bioactive compounds and also highlight the potential of bigels in intelligent food packaging applications for real-time freshness monitoring and improved food preservation.

#### 3. Bigels used in 3D printing

- Despite the instability and weak self-supporting properties of emulsions and emulsion gels in food 3D printing, integrating 3D printing technology with bigels presents a promising solution to enhance structural stability and functionality.



**Fig. 6. Bigels used in 3D printing**

#### Advantages

- Bigels dual-phase structure enables the encapsulation of both hydrophilic and oleophilic nutrients, enhancing nutritional diversity in food 3D printing.
- 3D printing technology has the potential to enhance food system by offering personalized food and nutritional choices, enabling customization of food products to meet individual preferences, dietary restrictions and nutritional needs.

#### 4. Bigels-based delivery systems for bioactive compounds

Co-delivery systems, capable of encapsulating two or more bioactive compounds simultaneously, emerged as valuable tools to improve health benefits and nutritional value of food products. Generally, bigels with higher gel strength could provide

enhanced protection for encapsulated bioactive compounds. Bigels with a higher ratio of oleogel/hydrogel possess higher gel strength, which can inhibit the penetration of gastric juices inside the bigels network. During the intestinal digestion stage, the oleogel network slowly collapsed, enabling a slow release of the encapsulated bioactive compounds.

Diagram illustrates the co-delivery of lipophilic and hydrophilic bioactive compounds, highlighting how different bioactive molecules interact with delivery systems to enhance their stability, release and bioavailability. Here's a more detailed breakdown of the key components:

### 1. Coenzyme Q10 (CoQ10)

- **Properties:** Adhesive, viscous, and highly lipophilic.
- **Benefit:** Exhibits the highest drug permeation, meaning it can efficiently pass through biological membranes.

### 2. Bioactive Fatty Acids

- Derived from avocado oil, coconut oil, and pomegranate oil.
- **Types:** Oleic acid, medium-chain fatty acids (MCFA), and conjugated linolenic acid (CLNA).
- **Behavior:** CLNA shows reduced stability in simulated gastric fluid (SGF), suggesting it may degrade under acidic conditions.

### 3. Probiotics

- Protected using **branched polysaccharides (starch, maltodextrin)**.
- **Benefit:** Increases bacterial **resistance against gastric pH**, ensuring probiotic survival in the stomach.

### 4. Lutein

- **Mechanical Properties:** Increased hardness, gumminess, and acts as an "active filler" in the gel matrix.
- **Release:** Decreased release in both SGF and simulated intestinal fluid (SIF), indicating controlled digestion and absorption.

### 5. EGCG (Epigallocatechin gallate) & Curcumin (CUR)

- Stabilized using GMS (Glycerol Monostearate).

- **Properties:** Enhanced thermal stability and oil-holding capacity.
- **Effect on Release:** CUR release is delayed, improving controlled delivery.

### 6. Quercetin

- **Health Benefits:** Improves sperm quality, motility, DNA integrity, and testosterone levels in rats with non-alcoholic fatty liver disease (NAFLD).
- **Potential Use:** Supports reproductive health and metabolic function.

### 7. $\beta$ -Carotene

- **Structural Benefits:** Increased storage modulus, stiffness, and fracture stress, making it more stable in the delivery system.
- **Release Profile:**
  - **SGF:** Reduced release (slower digestion in stomach).
  - **SIF:** Controlled release in intestines.

### 8. Lycopene

- **Structural Benefits:** Increased storage modulus and hardness in the matrix.
- **Controlled Release:** Reduces the immediate release of lycopene, allowing sustained absorption over time.

### Key Takeaways

- Bigel-based or composite delivery systems influence the stability, mechanical properties, and bioavailability of bioactive compounds.
- These formulations ensure protection, controlled release, and enhanced absorption of nutrients and drugs in the digestive system.

### 5. Bigels-based dysphagia diet product

Dysphagia is a swallowing dysfunction, causing difficulty in transferring a liquid or food bolus from mouth to stomach. Traditional dysphagia diet products rely on hydrocolloid thickeners which often compromise sensory appeal and nutrient bioavailability. Bigels technology offers an innovative approach consists of an interconnected network of aqueous (hydrogel) and lipid (oleogel) phases, providing controlled texture modification, enhanced nutrient encapsulation and improved sensory perception. Bigel-based dysphagia diet products can be applied in: thickened beverages, soft-textured solid

foods, gel -based nutritional supplements and fat-enriched purees. This novel approach has the potential to improve quality of life for individuals with swallowing disorders.

### Challenges and Perspectives

1. Bigels have become the focus of research due to its dual advantages of oleogel and hydrogel. However, some issues closely related to food applications remain and need to be explored over time.
2. One significant gap in research lies in the limited investigation of multi-component interactions between bigels and food matrices.
3. Furthermore, a comprehensive understanding of the metabolic fate of bigels within the human body is imperative, necessitating an elucidation of the complete metabolic mechanism.
4. Regrettably, there remains a dearth of detailed information concerning the specific digestive fate and metabolic processes involving bigels.

### Future Trends

**1. Environmentally Responsive Bigels:** Bigels are a new and exciting idea for the food industry and can change their properties based on environmental factors like pH, temperature, salt levels or enzymes. This makes them useful for controlling how and when nutrients or bioactive compounds are released in food. However, this concept is still not widely studied in the

food industry and more research is needed to develop it further.

**2. Evaluating the bioavailability of bioactive compounds through *In-vivo* experiments:** Bigel-based nutritional delivery systems show promise for functional foods, but their metabolic fate remains unclear. Most studies are *in vitro*, highlighting the need for *In vivo* studies to assess bioavailability and long-term effects.

### Conclusion

Bigels represents an innovative and versatile structuring system with broad applications in the food industry, particularly in developing healthier and more sustainable food products. Their ability to function as fat replacers, stabilizers and delivery systems for bioactive compounds makes them highly valuable in improving food quality and nutritional content. Additionally, the compatibility of bigels with natural biopolymers aligns with the increasing consumer demand for clean-label and functional foods. Future research should focus on optimizing the formulation and processing of bigels to enhance their performance in different food matrices, explore their interactions with other food ingredients, and assess their long-term stability. The integration of bigels into large-scale food production could pave the way for novel food structures that contribute to improved health outcomes and sustainability in the food sector.

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