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#### **REVIEW**



# Amelioration of the stability of polyunsaturated fatty acids and bioactive enriched vegetable oil: blending, encapsulation, and its application

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#### **ABSTRACT**

Lipid oxidation in vegetable oils is the primary concern for food technologists. Modification of oils like hydrogenation, fractionation, inter-esterification, and blending are followed to improve nutritional quality. Blending non-conventional/conventional vegetable oils to obtain a synergistic oil mixture is commonly practiced in the food industry to enhance the nutritional characteristics and stability of oil at an affordable price. Microencapsulation of these oils provides a functional barrier of core and coating material from the adverse environmental conditions, thereby enhancing the oxidative stability, thermo-stability, shelf-life, and biological activity of oils. Microencapsulation of oils has been conducted and commercialized by employing different conventional methods including emulsification, spray-drying, freeze-drying, coacervation, and melt-extrusion compared with new, improved methods like microwave drying, spray chilling, and co-extrusion. The microencapsulated oil emulsion can be either dried to easy-to-handle solids/microcapsules, converted into soft solids, or enclosed in a gel-like matrix, increasing the shelf-life of the liquid oil. The omegarich microcapsules have a wide application in confectionery, dairy, ice-cream, and pharmaceutical industries. This review summarizes recent developments in blending and microencapsulation technologies in improving the stability and nutritional value of edible oils.

#### **KEYWORDS**

Application; blending; encapsulation; microcapsules; oxidative stability

### Introduction

Vegetable oils extracted from various plant seeds are rich in polyunsaturated fatty acids (PUFAs) and monounsaturated fatty acids (MUFAs). These oils, due to their specific chemical and physical properties, are limited in technological application. Hence, modification like hydrogenation, fractionation, interesterification, and blending is mostly followed (Hashempour-Baltork et al. 2016). Hydrogenation utilizes catalysts such as hydrogen and nickel gas that saturates the unsaturated double bond and converts cis to a trans-state. Trans fatty acids are supposed to have toxic effects on human health (Iqbal 2014). Interesterification is an elective procedure for hydrogenation. Unsaturated fats are redistributed in the triacylglycerol structure amid this procedure, and no immersion or isomerization happens. In any case, this procedure needs uncommon and is costlier (Dijkstra 2015). Fractionation is a procedure in which a few fats/oils are isolated into two divisions with various dissolving and textural properties (Kellens et al. 2007). Fractionation can be utilized as a pretreatment preceding hydrogenation, interesterification, or mixing (Shahidi 2005). Therefore, blending is the simplest method of mixing diverse fats/oils with unique physical and compound attributes. Mixing vegetable fats/oils with various properties is one of the least complex techniques to make new explicit items with wanted nutritional and oxidative properties. The blended oil is rich in MUFA/ PUFA, which makes it chemically unstable and susceptible to oxidation. The exposure of oxygen to these oils creates cleavage in the unsaturated bonds, thereby elevating the oil rancidity. The unpleasant odor has a negative impact on the sensory attributes and lowers the overall acceptability. Thus, microencapsulation innovation could be a reasonable alternative to maintaining its textural, sensory, and oxidative characteristics.

Microencapsulation is the process of enveloping one substance termed as core material into another called wall/coating materials, improving stability and functional properties. Microencapsulation and controlled release of flavors have too reformed the nourishment, enhancing the flavor, stability, nutritive value, and appearance of their products (Pattnaik et al. 2021). In these regions, the conversion of liquid to easy-to-handle dry powders, gels, or beads were the inspirations for the utilization of microcapsules. The different types of microcapsules and microspheres are produced from a wide range of wall materials like carbohydrates, proteins, gums, etc. There are several different microencapsulation processes, such as spray-drying, coaxial electrospray system, freeze-drying, coacervation, in situ polymerization, melt-extrusion, etc. (Albert, Vatai, and Koris 2017; Bakry, Abbas, et al. 2016; Adelmann, Binks, and Mezzenga 2012).

There have been numerous review papers describing the blending of vegetable oils and their effect on their physicochemical properties, the methodology of microencapsulation of different core materials like volatile flavors, probiotics, essential oils but none of them have focused on vegetable oil encapsulation and its related problems (Jurić et al. 2020; Bakry, Abbas, et al. 2016; Kaushik et al. 2015b). Hence, this review paper will provide a concise reading to the researchers covering all the aspects starting from vegetable oil blending to oil powder/gel through microencapsulation. The paper focuses on the impact of vegetable oil blending on the nutritional, compositional, thermal, oxidative, and physical properties of the edible oils; providing a clear understanding of the selection of wall materials for encapsulation of vegetable oil, and the advantages and disadvantages of various conventional and emerging encapsulation techniques.

# Blending of vegetable oils

Formulation of an admix of edible oil to improve the health and nutritional aspect of edible oil is seeking little attention. To be counted as a healthy oil, the blending of vegetable oils has been a recent trend followed by most industries. "Healthy oil" by definition is the edible cooking oil that satisfies the fatty acid compositions recommended by World Health Organization (WHO) to prevent various diseases like diabetes, chronic heart disease, obesity (WHO 2008). Fats are an essential part of a healthy balanced diet; there is a shred of evidence to show that limiting saturated and trans fat intake is important since it contributes to the lessening of the build-up of fatty material (plaque) inside the blood vessels (arteries). This process is called atherosclerosis and is a major cause of heart disease. Saturated fatty acid (SFA) and trans fats increase low-density lipoprotein (LDL) cholesterol in the blood, which leads to plaque formation (Ference et al. 2017). PUFAs and MUFAs reduce LDL cholesterol and lipoprotein cholesterol increase high-density (HDL) (Manchanda and Passi 2016). MUFAs are beneficial in that they increase esterification of cholesterol in the liver, thereby reducing the free cholesterol pool and increasing receptormediated uptake of LDL cholesterol, resulting in a decrease in blood cholesterol levels as reported by the Dietary Guidelines Advisory Committee on the Dietary Guidelines for Americans, 2010. It can be seen from the studies that even higher consumption of PUFA has an adverse impact on the health by weakening the capability of the antioxidants in the human body to tackle free radicles, thereby increasing the risk of aging, cardio-related issues, diabetes, and cancer (Choudhary and Grover 2013; WHO 2008; Vani, Laxmi, and Sesikeran 2002). According to WHO, the total fat intake should be 30%-35% Total energy, SFA <10%, MUFA 10%-14%, PUFA 6%-11% Total energy (WHO 2008). However, to maintain good heart health,  $\omega$ -6 and  $\omega$ -3 must vary between 1:1 and 4:1 (Mishra and Manchanda 2012). Henceforth, a balanced ratio of MUFA, PUFA as well as essential fatty acids like  $\omega$ -6 and  $\omega$ -3 is essential to maintain a modulated lipid profile in the human body. A balanced fatty acid composition can be achieved by adopting vegetable oil blending practice, which will eliminate the need for hydrogenation or inter-esterification of oils.

# Effect of blending on the physical, chemical, and thermal properties

Lipid oxidation of edible oil has resulted in the development of off-flavors and rancidity, drastically lowering the stability of these oils, thereby imposing a pronounced negative effect on human health (Adbel-Razek et al. 2011). Therefore, the mixing of various edible oils is an economical way of enhancing oxidative stability by strengthening their antioxidant potentials (Table 1). For instance, flaxseed seed oil (FSO) and olive oil (OO) are rich in unsaturated fatty acids in terms of  $\omega$ -3 and  $\omega$ -9, respectively, making it prone to oxidative and hydrolytic cleavage of the double bonds. However, FSO, when combined with rice bran oil (RBO) in the ratio 2:1, exhibited a lower peroxide value, p-anisidine value, and acid value (Ghosh, Srivastava, et al. 2019). Furthermore, a ternary blend of FSO, RBO, and OO in the ratio 2:1:1 showed an exceptionally lower value of peroxide, p-anisidine, and free fatty acid (Ghosh, Srivastava, et al. 2019). RBO is a non-conventional oil; it has oryzanol, tocotrienols, and tocopherols in abundance along with squalene and phytosterol. The presence of oryzanol and trienols in RBO might have slowed down the formation of obnoxious compounds like aldehyde, ketones, and free radicles (Choudhary and Grover 2013; Reddy et al. 2013). Similarly, on blending RBO in different concentrations with OO, it showed the least peroxide value (0.53 and 0.33 mEq/kg), and highest when blended with mustard oil (MO), i.e., 1.73 and 1.33 mEq/kg. The possible reason for such disparity may be due to the variation in MUFA and PUFA in the respective oil blends. The blend containing a higher amount of MUFA or oleic acid than PUFA ought to slow down the degradation during shelf-life and (RBO + OO: 42% oleic acid, 36.9% PUFA and RBO + MO: 32.3% oleic acid and 50.8% PUFA) (Choudhary, Grover, and Kaur 2015). Although walnut oil (WO) and grape seed oil (GSO) has the same MUFA and PUFA content, the presence of a lower quantity of  $\omega$ -3 in GSO doubles its oxidative stability. Given the MUFA content, both RBO and toasted sesame oil (TSO) possess a similar amount. However, due to the prevalence of potent antioxidant components in RBO, it has a slightly higher induction period than TSO and 4 times higher than WO (18.7 h vs. 18 h vs. 4.2 h). The antioxidants activity of TSO in terms of tocopherol, sesamol a potent antioxidant formed from sesamolin lies on the higher side than the refined sesame oil (Kochhar and Henry 2009). Similar results were reported by Pattnaik and Mishra (2021), the addition of RBO (above 70%) to groundnut oil (GO) and FSO provided substantially higher oxidative stability. Consequently, owing to the pre-dominant bio-active components, RBO is labeled as "heart oil" and also considered to satisfy the fatty acid composition recommended by WHO (Choudhary, Grover, and Kaur 2015). Sometimes, coldpressed oil can be a great option to improve stability. The cold-pressed oils are more endowed with nutritive properties

Table 1. Major findings of physicachamical proporties and health benefits of various vegetable oil blands

Oil blend	MUFA:PUFA	n6:n3	Findings	Health benefits	References
Flaxseed + tomato seed oil; Flaxseed + tomato seed oil + rice bran oil	_	_	Highest antioxidant activity and phytochemical content with excellent oxidative stability	_	Ghosh, Srivastava, et al. (2019)
Sunflower oil + sesame oil	0.70	_	Increased stability of sunflower oil	_	Ghosh, Upadhyay, et al. (2019)
Olive oil + sunflower oil + cress oil	1.5	4.8	Improved the functional characteristics, thermal and oxidative stability of individual oils	Balanced MUFA, PUFA & essential fatty acids have beneficial effects on cardiovascular health	Nehdi et al. (2019)
Rice $bran + sesame$ oil	1.19	88.9	Rich source of antioxidants and unsaturated fatty acids	Antihypertensive and lipid- lowering action	Devarajan et al. (2016)
Canola oil $+$ palm oil	1.62	2.92	Enhanced oxidative stability	Improvement in biochemical parameters and serum fatty acids	Adeyemi et al. (2016)
Rice bran oil $+$ partially hydrogenated oil	2.42	21.8	Increased the antioxidants majorly oryzanol content	Lowering of adverse effects and pro-inflammatory effects of pure partially hydrogenated oil	Rao, Kumar, and Lokesh (2016)
Olive oil + sunflower oil; olive oil + soybean oil	3.33;4.14	13.43;12.24	Higher in dietary MUFA content	Cardioprotective activity through lipid-lowering and plasma cholesterol reduction causing hypolipidemia	Jan et al. (2016)
Canola oil $+$ palm oil/ sunflower oil	2.62;1.10	12.5; 4.76	Frying stability of the blended oils	Improved lipid profile of dietary rats	El-Reffaei et al. (2016)
Soybean oil $+$ camellia oil	1.17	6.4	Thermal and frying stability of soybean oil due to increased phenols and MUFA in camellia oil	_ ′	Wang et al. (2016)
Rice bran $\operatorname{oil} + \operatorname{peanut} \operatorname{oil}$	0.75	_	Thermally stable to high cooking and frying conditions	_	Choudhary, Grover, and Kaur (2015)
Rice bran + garden cress oil; Sesame oil + garden cress oil	1; 0.93	2.2; 2.4	Increase the antioxidant activity of oils	No significant change in serum and liver peroxide content; deceased total cholesterol and regulated lipid profile	Umesha and Naidu (2015)
Sunflower oil + garden cress oil	0.56	2.3	Balanced essential fatty acids	Enhanced radical scavenging activity and decreased total cholesterol	Umesha and Naidu (2015)
Canola oil $+$ olive oil $+$ palm oil	2.66	3.74	Increased stability by modifying the fatty acid composition	_	Roiaini, Ardiannie, and Norhayati (2015)
Soybean + sesame oil	0.55	_	Better oxidative stability at high temperatures	Prevention of chronic diseases associated with oxidative stress, such as in cancer and coronary artery disease	Li et al. (2014)
Rice $\operatorname{bran} + \operatorname{olive}$ oil	1.42	2.14	High smoke point and frying temperature with good retention of antioxidants, lower acid value, and least peroxide formation	Favorable effects on cholesterol regulation and LDL cholesterol oxidation	Choudhary and Grover (2013)
Rice bran + flaxseed oil	1.1	4.0	Possessed good oxidative stability over the storage time	Functional and health- promoting oil blend with an ideal balance of fatty acids	Reddy et al. (2013)
Palm oil $+$ olive oil	4.4	34.8	Better oxidative stability of the blend with 20% olive oil comparable to palm oil	Comparable health benefits linked to cholesterol, LDL, HDL, and triglycerides	De Leonardis and Macciola (2012)

as they do not undergo any chemical or heat treatment, keeping the antioxidants or antioxidant precursors intact. Cold-pressed OO is the most desirable substitute for conventional oils because of its natural antioxidants, mainly phenols, and tocopherols. Thus, on mixing 20 and 40% of OO with sunflower oil (SFO) and soybean oil (SBO), it increases the radical scavenging activity of the individual oils from 55% to nearly 78% and the total phenolic content of SBO to 10.5 to 51 mg/100g, SFO 20.5 to 69.5 mg/100g (Abdel-Razek et al. 2011). Another such blend between nonconventional RBO and traditional OO (70:30) showed a better oxidative and antioxidant stability even after 28 days of

storage, possessing 2525 mg/kg of total natural antioxidants and 67.7% radical scavenging activity (Choudhary and Grover 2013). Several non-conventional oils like black cumin oil, garden cress oil (GCO), moringa oleifera oil, MO, and camellia oil have a substantial amount of antioxidative potentiality, hence, can be reviewed for judicious blending (Umesha and Naidu 2015; Wang et al. 2016; Mohamed, Elsanhoty, and Hassanien 2014; Anwar et al. 2007).

Color and viscosity are the prime attributes associated with the deep frying of oils. Higher viscosity denotes the formation of polymers or primary and secondary oxidation products. Low viscosity is indicative of unsaturation and

average fatty acid carbon length. Similarly, the color of individual oils changes after blending and almost darken after repeated frying at high temperature because of the accumulation of oxidation compounds. The blend of RBO and GO shows an appreciable smoke and frying temperature ranging between 160 and 182 °C (Choudhary, Grover, and Kaur 2015). As RBO is related to the high 4-monomethylsterols content with an ethylidene side chain, it may have contributed to its high smoke point oxidative stability. Besides, oryzanol is a combination of ester compounds that aid in stabilizing oils at frying temperatures. To meet the standards of frying oils, the frying temperature of vegetable oil should not exceed 180 °C as high temperature accelerates oxidation, polymerization, and free fatty acid formation of oils. When an oil blend of 50% RBO and 50% SFO was heated at 180 °C, it caused a 43% reduction in polymer triacylglycerol formation in the blend than SFO (Mezouari and Eichner 2007). A blend of canola oil and palm oil (1:1), when used for frying at 180 °C, was acceptable up to 3 frying cycles with respect to its change in physical and chemical properties (Enríquez-Fernández, Álvarez de la Cadena y Yañez, and Sosa-Morales 2011). Palm oil admixed with 20% OO led to a 32% reduction in the formation of short-chain fatty acids on heating at 130 °C due to the saturated fats present in palm oil, which favored less PUFA loss in the blend (De Leonardis and Macciola 2012). Henceforth, mixing OO more than 20% might lower the stability due to the excessive PUFA content. SBO might be considered as one of the superior vegetable oil, but it is regarded to be inferior in thermal stability at high temperatures. A binary mixture of SBO with sesame oil (SEO) (80:20 v/v) might enhance the lipid oxidative stability of fried products when fried at 160 °C (Li et al. 2014). Therefore, a better choice of vegetable oils and careful oil blending will govern the change in fatty acid composition and antioxidants affecting the overall quality of oils.

# Effect of blending on the nutritional value

According to the World Health Organization, the most important criteria for the nutritional evaluation of oils are: (i) ratio of saturated, mono, and polyunsaturated fatty acids, (ii) ratio of  $\Box$  -6 and  $\Box$  -3, and (iii) presence of antioxidants. As previously mentioned, the optimum ratio of saturated, mono, polyunsaturated fatty acids and 0-6, 0-3 to maintain a healthy heart is 1:1.5:1, 1:1-4:1, respectively.

□ -6 and □ -3 are the essential fatty acids to regulate the body's functioning. Due to the lack of omega-3 desaturase, a converting enzyme in human cells, they can neither convert □ -6 to □ -3 nor can produce it. Hence, it needs to be provided externally through dietary intake. Linoleic acid (LA) and eicosapentaenoic acid (EPA) are the parent  $\omega$  fatty acids 6 and 3, respectively, which produce eicosanoids responsible for the physiological changes in the human body. The eicosanoids produced from both the  $\omega$  fatty acids have opposite properties. The larger quantities of are a common problem in Western diets. It increases the eicosanoid metabolic products from LA, particularly hydroxy fatty acids,

thromboxanes, lipoxins, prostaglandins, and leukotrienes than EPA. The eicosanoid produced from LA should be in lower quantities to become biologically active. On the contrary, the larger quantities of eicosanoids formed from LA contribute to the formation of thrombus and atheromas with high blood viscosity, few allergic and inflammatory disorders leading to the proliferation of cells in most vulnerable people (Simopoulos 2016). A careful blending of oils will maintain a balanced fatty acid composition. Devarajan et al. (2016) claimed that a blend of 20% cold-pressed, unrefined SEO with 80% RBO showed excellent results in lowering the blood pressure and modulating lipid profiles such as an increase in HDL and LDL decrease after treating hypertensive patients. Blending of a -3 rich GCO with a -6 rich SFO, SEO, and RBO significantly reduced the radical scavenging activity (IC50), improved the antioxidants, lowered the " -6: " -3 ratio in Wister rats. It is noteworthy that it also increased the Glutathione peroxidase and catalase activity, protecting against oxidative damage (Umesha and Naidu 2015).

A similar reduction of LDL cholesterol and triglycerides in rats was observed by Sharma and Lokesh (2013) on mixing -3 rich FSO with GNO. A significant reduction in serum cholesterol by 27%-29% in hamsters was achieved when fed with blended oils of OO with SFO and SBO, but higher intake of soybean oil beyond 20% can lead to excessive weight gain, hyper-sensitive, higher blood glucose level, increased risks of the tumor (Jan et al. 2016; Li et al. 2014). Thence, OO has -3 in abundance and can be a potential oil for blending with other " -6 rich oils such as SEO, RBO, or SFO. Apart from essential fatty acids, OO is a source of a wide variety of antioxidants, predominantly hydroxytyrosol, tocopherols, and oleuropein, which are primarily responsible for lowering LDL cholesterol, exhibit anti-inflammatory, anti-hypersensitive, and antithrombotic like health benefits (Choudhary and Grover 2013). Tocopherols, γ-oryzanol, and phytosterol-rich vegetable oils possess a strong inhibition of oxygen radicles, risk of cancer, and anti-atherogenic effect. This  $\gamma$ -oryzanol particularly is associated with a strong inhibiting power against ADP and collagen-related platelet aggregation (Cicero and Derosa 2005). An effective blending of oils consisting of health-promoting antioxidants, several bioactive compounds, and -3, -6 incorrect amount will lessen mortality and facilitate more flexibility in contributing nutritional properties.

### **Encapsulation strategies for oils**

Encapsulation of vegetable oils means trapping liquid oil into a matrix to obtain the desired effect. Vegetable oils are encapsulated to mask its aroma, flavor, color, increase its oxidative stability, control its release, and increase its bioavailability (Sagiri, Anis, and Pal 2016; Adelmann, Binks, and Mezzenga 2012; Tonon, Grosso, and Hubinger 2011). Hence, depending upon the desirability of the product, the liquid oil is either converted into gels, beads, or powder.

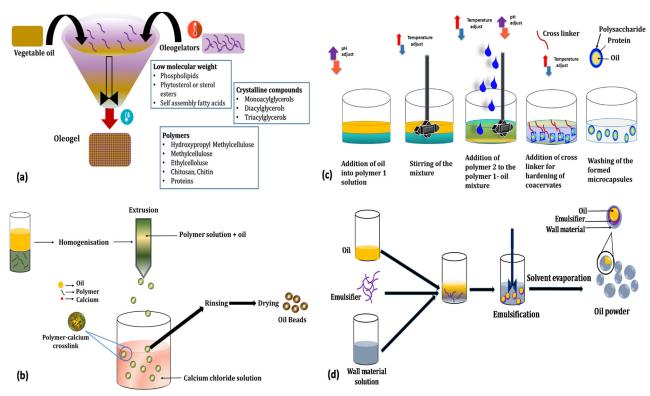


Figure 1. Schematic illustration of different strategies for encapsulation processes (a) oleogel, (b) oil beads, (c) oil capsules by coacervation, and (d) oil powder.

# **Oleogels**

One of the encapsulation methods of reducing the use of saturated fats in food applications is the formulation of oleo gels. Oleogels are mostly the structured oils that are preferred to prevent fat migration or the fat bloom during the melting of chocolates or similar products (Hughes et al. 2009). These are prepared by gelation of vegetable oil with oleogelators like waxes, alcohols, phospholipids, mono-diglycerides, and phytosterols. Oleogelators effectively trap and immobilize the liquid oil within a 3-D structure converting the resultant mixture from liquid to a hard gel-like structure (Figure 1a). Firstly, the oleogelators are heated at 50–170 °C until it melts, then it is added to oil where the solution is homogenously mixed before cooling to set it down into gels. There are three proposed methods for the formation of oleogel, namely; 1) self-assembled structures of polymers, 2) self-assembled structures of low molecular weight compounds, 3) other miscellaneous structuring materials (Patel et al. 2015; Co and Marangoni 2012). The first method involves gelation by hydrophobic polymers such as ethylcellulose, polysaccharides, or proteins as a gelling agent (Patel et al. 2015). In the second method, gelation is done by using low molecular weight phytosterols or entrapping oil within crystalline strands of monoacylglycerols (MAGs), diacylglycerols (DAGs), or triacylglycerols (TAGs) particles (Pehlivanoğlu et al. 2018; Patel et al. 2015; Sahoo et al. 2011). Lastly, some other inorganic gelators like fumed silica are used for producing gelling networks in SFO (Patel et al. 2015).

Oleogelators play a vital role in entrapping oil into 3-D structures. The food-grade oleogelators must be GRAS and possess certain properties viz. thermoreversibility, surface

activity, nontoxic, and lipophilic (Doan et al. 2015; Patel et al. 2013) (Table 2). There are several oleogelators available based on the gelling type namely; (i) low molecular weight compounds, e.g., phospholipids (lecithin), phytosterol or sterol esters (sorbitan monostearate, y-oryzanol, sorbitan tristearate, sitosterol), and fatty acids forming self-assembly fibrous network (sphingolipids, hydroxy stearic acid); (ii) polymers cellulose [EC]), hydroxypropyl methylcellulose [HPMC], methylcellulose, chitosan, chitin, proteins [zein proteins, gelatin, b-lactoglobulins]); (iii) crystalline compounds (plant or natural waxes, MAG, DAG, cholesterol, policosanol) (Patel and Dewettinck 2016). Several studies suggest that waxes are very efficient in binding oils within crystal networks at low concentrations (<10%) due to long-chain, high melting point, and low polarity (Patel et al. 2013). Identically, a thermos-reversible EC, when used as a gelator, results in hard oleogels because of a strong interaction between the oil phase and the gel phase through bonds. The increased gel strength of the oleogels with high setting temperature was associated with the strong network of polymer-polymer and hydrogen-bonding (Davidovich-Pinhas, Marangoni 2015a). On the other hand, certain surfactants can be used in combination or alone based on their plasticizing properties. The addition of surfactants can considerably affect the sol-gel transition temperature and interactions, thereby changing the quality of the gels (Davidovich-Pinhas, Barbut, and Marangoni 2015b). In some cases, proteins have been used as surfactants through an emulsion-template approach to entrap a good amount of oil content in their high internal phase emulsions. Tavernier et al. (2017) prepared oleogels through a similar approach by forming a protein-polysaccharnetwork using soy protein isolate (SPI) and

Table 2. Different coating materials used for encapsulating PUFA rich edible oil.

Coating materials	Preparation and properties	Limitations	Benefits	Application	References
Maltodextrin (MD)	Acid- or enzyme-catalyzed starch hydrolysate with Mw <4000 g/mol, white hygroscopic polysaccharide, easily digestible, soluble in water	Slightly sweet, increase the viscosity	Forms a conjugate with protein, helps in emulsifying when used in combination, film-forming ability, thickener	Emulsification, microcapsules	Nurhadi, Roos, and Maidannyk (2016)
Methyl cellulose	Produced by heating cellulose with caustic solution and treating it with methyl chloride. Dissolves in cold water forming a clear, viscous solution, gel formation on heating above 50 °C, gels are reversible on cooling	Insoluble in hot water and other solvents, high viscosity at low concentration	Stabilizes emulsion & foam, modify texture, act as thickener, adhesive, filmforming ability	Gelation, emulsification	Nasatto et al. (2015)
Ethylcellulose (EC)	Prepared by mixing the alkali cellulose with ethyl chloride in the presence of alkali at about 60° for 12 h under pressure. Solubility depends on degree of substitution (DoS):  1.0 < DoS < 1.5 soluble in water; 2.4 < DoS < 2.5 soluble in organic solvent	To entrap oil, EC needs to be heated above glass transition temperature and eventually cooled down	Forms elastic gels with surfactants, higher gel strength in the presence of oleic acid, acts as a stabilizer and film- forming agent, sustained release	Gelation, microcapsules	Wasilewska and Winnicka (2019); Singh, Auzanneau, and Roger (2017)
НРМС	Off-white/white nonionic cellulose powder prepared by etherification in alkaline condition, forms a non-flowable and semi-flexible mass when heated to critical temperature	Forms colloids on dissolving in cold water, soluble in polar organic but insoluble in diethyl ether, acetone, and anhydrous alcohol	Excellent film forming, act as stabilizer and surface tension enhancer, thickener, adhesive properties, water retention capacity	Thermal gelation, emulsification	Ding, Zhang, and Li (2015); Novak et al. (2012)
Gum Arabic (GA)	A mixed salt of a polysaccharidic acid (Arabic acid) with Ca <sup>2+</sup> , Mg <sup>2+,</sup> and K. Readily soluble in water, low viscosity	_	Emulsifying agent, high water-holding capacity, binding agent, degrades oxidation	Used as a thickener in oil gels and emulsions	Naeli et al. (2020); Mariod (2018)
Galactomannans	β-(1-4)-D-mannan (M) backbone with single D- galactose (G) branches linked α-(1-6). Water solubility increases with an increase in galactose	Film-forming properties vary with M/G ratio	Thickeners, excellent stiffeners and act as an emulsion stabilizer	Used with other polymers for film coating, or gels	Dos Santos et al. (2015); Silveira and Bresolin (2011)
Pectin	A methylated ester of polygalacturonic acid $\alpha$ -(1->4)-linked D-galacturonic acid; LMP (<50% esterified), HMP (>50% esterified), Dissolves in water	Forms clumps during dispersion; HMP- forms gels with sugar and acid, LMP forms gels with divalent cation	Gelling agent, thickener, water binder, and colloidal stabilizer	Beads or capsules	Sundar Raj et al. (2012)
Carrageenan	The number and position of ester sulfate groups influence structure; Kappa: 25%–30% ester sulfate groups; lota: 28%–35% ester sulfate groups; Lambda: 32%–39% ester sulfate groups. In reaction with water, it forms a gel, whereas when added to milk it reacts with proteins and stabilizes	KAPPA – rigid and brittle gel, thermo- reversible, high gel strength, shows syneresis. IOTA – elastic gel, thermo-reversible, no syneresis, thixotropic. LAMBDA – cold soluble, non-gelling, high viscosity.	Emulsification, foam stabilization, thickening, gelling, and suspending agent in water and milk systems	Gelation	Kariduraganavar, Kittur, and Kamble (2014)
Alginate	The anionic polymer obtained from seaweed. Sodium and potassium alginate dissolves in hot and cold water with agitation	lonic cross-linked gels by divalent ions are less stable but can be cross-linked by cell or covalent reagents	Acts as stabilizing, viscosifying, and gelling agent	Gelation	Lee and Mooney (2012)
Xanthan gum (XG)	High-molecular weight extracellular heteropolysaccharide produced by fermentation of <i>Xanthomonas campestris</i> . Soluble in cold and hot	High viscosity at low concentration	Acts as thickener and stabiles emulsion, suspension and foams, have a synergistic effect with other gums	Microcapsules	Bascuas et al. (2020); Cai et al. (2019)

(continued)

Coating materials	Preparation and properties	Limitations	Benefits	Application	References
	water, dissolves in most				
Chitosan	acids and bases The deacetylation of chitin derives a linear polysaccharide. The weak base having solubility < pH 6.5	Poor encapsulating properties, insoluble in water or organic solvents	Act as an emulsifying and stabilizing agent in combination with proteins	Used in conjunction with proteins	Kumar et al. (2020)
Milk protein concentrate or isolate (MPI/MPC)	MPI contains high protein >90%; MPC has protein <90% dry matter, Off white powders with sweet and milky aroma produced by spray drying of ultra or diafiltrated skim milk	MPC/MPI with high protein content has poor solubility	Emulsifying and oil binding capacity provides heat stability	Emulsification	Meena, Singh, Arora, et al. (2017); Meena, Singh, Panjagari, et al. (2017)
Sodium/ potassium caseinate	White or pale yellow, odorless powder formed by the reaction of casein with alkali. Soluble in boiling water, insoluble in ethanol	Slowly disperses in water with turbidity, Poor acid stability	Emulsifying properties, heat, acid, foam and freeze stability, water binding capacity, high surface activity, water- soluble emulsifier	Emulsification	Meena, Singh, Panjagari et al. (2017)
Whey protein isolate/ concentrate (WPI/WPC)	Pale yellow or white mixture of proteins obtained from spray drying of whey, soluble in water	Poor heat and freeze stability and poor water binding capacity	High surface activity, good emulsifying & foaming agent, excellent emulsion and acid stability	Emulsification when used in a composite blend	Meena, Singh, Panjagari et al. (2017)
Soy protein isolate/ concentrate (SPI/SPC)	Light brown powder prepared from defatted soy flour or by immobilization of soy globulin proteins; SPI: >90% proteins dry basis, SPC ~ 70% proteins dry basis	Low gel intensity and porous structure; solubility varying with pH	Good emulsifying and water retention capacity, provides colloidal and foam stability, shows high viscosity, plasticity and elasticity properties, gelation in the presence of salts	Emulsification, soft gelation	Wang et al. (2019); Tang (2017); Xu and Liu (2016)
Gelatin	An admixture of peptides and proteins produced by partial hydrolysis of collagen. Soluble in the water at temperatures above 35–40 °C, sets when cooled	Swells at low temperature absorbing more water	Acts as a stabilizer, thickener, and texturizer	Gelation	Kanwate and Kudre (2017); Haddar et al. (2011)
Lentil protein	Prepared by extraction from lentils and other pulses. Solubility varies with the type of lentil and pH	Weak gelling, foaming, and emulsifying capacity when used alone	Excellent nutritional factors, high digestibility, good oil binding capacity, act as an extender	Used for emulsification in conjunction with other proteins or polysaccharides	Jarpa-Parra (2018); Chang, Varankovich, and Nickerson (2016)
Cyclodextrin	Non-reducing carbohydrate obtained from enzymatically modified starches consisting of $\alpha$ -1,4-linked glucose monomers. Water solubility varies as $\gamma > \alpha > \beta$ —cyclodextrin, and solubility increases with temperature	Insoluble in organic solvents	Forms self-assembled aggregates, enhances penetration through a biological membrane	Encapsulation by complexation	Rakmai et al. (2018); Jansook, Ogawa, and Loftsson (2018); Marques (2010)
Carboxymethyl cellulose (CMC)	White to slight yellow modified cellulose powder produced by carboxymethylation, Soluble at any temperature	Low surface activity and non-foaming	Excellent film-forming capacity, good water retention but pH- dependent, good binding, stability, and emulsifying ability	Emulsification, coacervation, microcapsules, or micro/nanobeads	Bakry, Fang, et al. (2016); Ngamakeue and Chitprasert (2016); Devi and Maji (2011)

 $\kappa$ -Carrageenan (charged polysaccharide) in the ratio 15:1 that showed a long-term emulsion (with 60% oil) stability. The dried oleogel had a unique honeycomb-like structure without any oil leakage for several months of storage. Similarly, Tavernier et al. (2018) demonstrated better oil retention in oleogels formed by SPI (2.5 wt%)—candelilla wax (1-5 wt%) than oleogels prepared by only candelilla wax due to the combination of both internal crystalline network and protein

stabilized compartments. Patel et al. (2015) reported high oleogel strength that contained >97 wt% oil using biopolymers like gelatin and xanthan gum (non-surface-active). They did not observe any coalescence of the oil droplets, additionally, there was tight packing of oil droplets within the polymer.

emulsion-template approach, solvent exchange method is another procedure to disperse proteins

in oil via strong protein networks in a hydrophobic environment. In this technique, the protein is initially denatured to form disulfide bridges and hydrophobic interactions between proteins. Thereafter, the polarity is reduced first by replacing water with intermediate solvents (acetone, oxolane) then further by oil (Scholten 2019). De Vries et al. (2015) reported successful oleogel formation using solvent exchange method with heat-set whey protein isolate (WPI) and SFO. They reported stiffer oleogels with proper networks conferring good oil holding capacity (up to 91%). Similar studies were conducted by De Vries et al. (2017) using the same approach on denatured WPI and SFO (1:10). They claimed that the WPI aggregates showed gel-like properties and formed a gel network with the oil even at low protein concentration ( $\sim$ 3%).

The type and concentration of oil and oleogelators influence the textural, thermal, and rheological properties of the oleogels (Patel et al. 2015). The gelling ability of various waxes is affected by the vegetable oils' fatty acid composition and acyl chain length (Demirkesen and Mert 2020). For instance, with the increase in SFA content, the gelling concentration gradually decreases due to the high melting rate of TAG, ultimately strengthens the oleogel structure. A lower gelling concentration of bee wax was observed for RBO because of high saturation than rapeseed oil. Similarly, keeping the bee wax concentration the same, a higher crystalline mass of wax was formed in RBO with the high latent heat of crystallization (Patel et al. 2015). Therefore, oleogels containing high melting TAG and SFAs produce rigid gels with high viscosity. Studies show that in comparison to heterogenous waxes, homogenous waxes having less minor components and higher ester concentration possess lower critical gelling concentration. Due to this lower critical gelling concentration, it requires a minimum amount of oleogelator to form oleogel that provides good oil binding capacities (Demirkesen and Mert 2020). Consequently, from an economic point, a lower concentration of oleogelators (0.5-7% w/w) can be utilized to prepare firm oleogels with vegetable oils consisting of high oleic content such as RBO, SFO, and OO. Oleogels can be used for developing functional foods by entrapping bioactive compounds without requiring a large amount of saturated fat because of their distinctive solid-like-fat attributes that confer better protection and higher stability.

#### Oil beads

Oil beads are typically like hydrogel beads wherein the core material is a liquid oil. The basic preparation of such beads involves the dispersion of vegetable oil into the solution of wall materials, after that thoroughly mixed to form a homogenous solution (Figure 1b). The oil load differs and can reach up to 50% of the final product or beyond depending upon the oil-to-wall weight ratios. The oil-to-wall ratios normally vary from 0.1 to 1.0; however, 0.2 to 0.5 are more common (Chan 2011). There is a large variety of proteins, carbohydrates, and gums used as wall materials (Table 2). Gums have an excellent stabilizing property but exhibit poor

encapsulating ability (Mahdavi et al. 2016; de Oliveira, Paula, and de Paula 2014; Ramı rez et al. 2002). A cyclic polymer of six alpha-1,4-linked glucopyranosyl units known as  $\alpha$ -Cyclodextrin ( $\alpha$ -CD) is utilized as an encapsulating medium due to its amphiphilic behavior. It self-aggregates to form oil beads on continuous shaking at low temperature (25-37 °C) with the triglycerides. The absence of any chemical solvent, low temperature, and good encapsulation efficiency (80%-87%) make this polymer suitable for the development of soft beads (Bochot et al. 2007). However, minimal knowledge about its toxicity and the dosage limits its use in food products. Sodium alginate has been a classic example of gelling and thickening agents for a longer time. It is a copolymer with two monomeric units of D-mannuronic acid and α-L-guluronic acid. The ability to reduce the interfacial tension between the oil and water phase makes it an efficient wall material for bead preparation. They form thermo-irreversible and water-insoluble gel beads on chemically reacting with calcium ions. The calcium cations cross-link to the guluronic sequences to form gel networks; therefore, high guluronic acid content (M/G ratio = 0.59, G type), have better encapsulation efficiency than high mannuronic acid content (M/G ratio = 1.56, M type) (Chan 2011). Pectin extracted from citrus fruits is associated with similar gelling properties, which can be used alone or in combination with alginate to form gel beads. Depending upon the methyl esterification of the galacturonic acid in the chains, pectin can be classified into low-methoxy (LMP) or high-methoxy (HMP). HMP is conventionally used as a thickening agent, and the formation of gel occurs only by hydrogen-bonding and hydrophobic interactions in the presence of acids and a high amount of sugar. On the contrary, LMP forms thermo-reversible gel beads with calcium ions and does not require high sugar content. LMP has a less demarked dimerization step than alginates, due to the random distribution of ester and amide groups along the pectin chain. Additionally, the gel strength is influenced by crosslinks and concentration of calcium divalent ions while reduces with an increase in temperature and acidity (Yang et al. 2018; Capel et al. 2006). The calcium-pectin oil gel beads have gained popularity for their sustained release and targeted drug delivery.

Various studies have been conducted in recent years on the oil beads prepared from different wall materials. To improve the encapsulation efficiency, polysaccharides are used as structural strengthening agents, while proteins act as emulsifiers (Corstens et al. 2017). For instance, Morales et al. (2017) observed an encapsulation efficiency of 98.7% in oil beads prepared by a protein-polysaccharide complex of sodium alginate-shellac and SFO in the ratio of 80:20. They also observed a smooth and non-aggregated oil bead surface with an encapsulation efficiency of 98.7% (oil load: 38.6% w/w), which showed swelling properties under basic conditions (pH 7). Lin et al. (2020) investigated the interaction of SPI and alginate with varying oil content (10%-40%). They found that emulsion stability decreased on increasing the alginate concentration to 1.5%, oil to 40%, and limiting SPI to 1%, due to high viscosity that hindered

the movement of SPI to the oil-water interface. However, more SPI concentration led to better absorption to the oilwater surface but favored some flocculation of oil droplets. In another study, Lin et al. (2021) reported that the presence of WPI established stronger interactions with alginate than SPI during gel beads formation via hydrogen bonds between polar amino acids and alginate molecules that helped in preventing water loss and SFO loss from the beads. To obtain oil beads with high oil loads some researchers have suggested the addition of emulsifiers or adoption of emulsification technique in combination with ionic gelation. For example, e Silva et al. (2019) demonstrated that 1% Tween 20 exhibited stable emulsion through alginate interaction. They also observed high encapsulation efficiency (>99.5%) of the microparticle for oil loads up to 75%. On the other hand, Piornos et al. (2017) obtained beads with high oil load (66.37% linseed oil) using Lupin protein isolate (LPI) (56 g/ L) and alginate beads (47 g/L) by allowing gelation for 30 min. They further observed a high encapsulation efficiency (98.30%), due to the emulsifying properties of LPI. Nonetheless, the bead size, water/oil content, and mechanical properties (shrinkage, compressibility, elasticity) of the oil beads can be controlled by maintaining the gelation time, optimizing the formulation, regulating the process technologies and preparation methods (Lin et al. 2020).

# Oil capsules by coacervation

Coacervation in the colloidal solution is defined as the phase separation between two liquids caused by a change in pH, temperature, ionic strength, and the carrier medium's solubility. When the coacervation takes place or is completed, a visible separation of two phases occurs. One phase is called coacervate, and another is known as the equilibrium phase. When the coacervation is conducted using one polymer solution, it is a single coacervation, whereas the presence of two oppositely charged polymer solutions (preferably proteins and polysaccharides) is termed as complex coacervation (Timilsena et al. 2017; Schmitt and Turgeon 2011). The main driving force for complex coacervation is the electrostatic interaction between two charged particles, along with hydrophobic interactions and Van der Walls forces (Timilsena et al. 2017). The microencapsulation of oils by complex coacervation is carried out by three basic steps (Figure 1c) (Ruiz, Ortiz, and Segura 2017). Firstly, a protein-based polymer solution is dispersed into an aqueous medium by adjusting its pH beyond the isoelectric point and temperature above the gelling point. Secondly, the homogeneous oil-in-water emulsion is prepared by homogenizing oil in the prepared protein solution. Thirdly, another polymer solution is prepared by dispersing polysaccharides in an aqueous solution, which is followed by blending with the above O/W emulsion. The coacervates instantaneously form a coating around oil particles if there is adequate opposite charge density. In case of insufficient charge density, the polymer solution's temperature or pH is adjusted to a satisfactory charge level for the induction of coacervates. Gelatin is the most commonly used shell material for the

formation of complex coacervates. Flaxseed oil capsules formed from gelatin-gum Arabic (GA) matrix had an efficiency of 84% and transitioned from spherical mononuclear to irregular multinuclear when the rpm of the homogenizer was changed (Liu, Low, and Nickerson 2010). However, to increase the stability of the coacervates, either mild heat is applied, or some cross-linkers are added. The degree of solidification depends on the concentration of cross-linkers used in the process. Several cross-linkers include tannic acid, transglutaminase, glutaraldehyde, Gallic acid, or formaldehyde. Despite the strong linkage ability, the toxicity of the cross-linkers should be studied before its application. Timilsena et al. (2016) prepared complex coacervates from chia seed gum-chia seed protein isolates complex (wall material) and chia seed oil (core) using transglutaminase as a cross-linking agent. He observed the highest encapsulation efficiency (>93.9%) and longer storage stability almost 6 times than unencapsulated oil when the core: wall ratio was kept at 1:2. Kaushik et al. (2015a) studied the effect of pH alteration over a range of 8 to 1.5 and claimed that at pH 3.1, flaxseed protein isolate configured its helix structure, thus, provided a stable and strong linkage with flaxseed gum. It can be concluded that the strength of coacervates depends on the process parameters, charge density, concentration of the polymers, pH, and temperature. In comparison to other microencapsulation techniques, this process can take a high payload up to 90% for single nuclear and 60% for multi-core by producing micro particles of the wide particle size range (1-1000 μm) (Kaushik et al. 2015b). Conversely, it is a batch process that wastes time and the ionic charge and pH of the material govern microcapsules' stability. Hence, a limited variety of wall materials and cross-linkers can be utilized for this process.

### Oil powder

Vegetable oils rich in PUFA are prone to oxidative degradation, which can deliberately shorten its shelf-life. Encapsulation of such oils can prevent untimely lipid oxidation and preserve the quality of the oils. It serves several benefits, such as protection from environmental conditions, increasing the stability of the oils, and the controlled release of omega fatty acids into the food product (Kaushik et al. 2015b). The encapsulated liquid oil can be converted into stable powders by following few steps viz. as shown in Figure 1d: (i) Emulsification is the process by which the core or active material (oil) is dispersed in the solution of wall materials and then thoroughly homogenized by a shear blender or homogenizer to form an emulsion, (ii) Solvent evaporation is used to remove the solvents used in the solution to dissolve the wall materials. Different drying methods can be employed for the conversion of an emulsion into powder such as spray drying, microwave drying, freeze-drying, fluidized bed drying, etc. which will be discussed in Bulk encapsulation of oil. Consequently, it produces microspheres of oil powder enveloped in the matrix of wall materials. Depending on the physico-chemical properties of the core, the wall composition, and the used microencapsulation

technique, different types of particles can be obtained such as a simple sphere surrounded by a coating of uniform thickness, a particle containing an irregular shape core, several core particles embedded in a continuous matrix of wall material, several distinct cores within the same capsule, and multi-walled microcapsules. The selection of wall materials is based on the emulsifying activity, stability, solubility, properties of the core, and desired final product. They actively influence the physico-chemical properties of the microcapsule viz. encapsulation efficiency, particle size, powder morphology, and lipid oxidation. The commonly used wall materials can be broadly classified into carbohydrates, proteins, gums, and wax (Table 2). Polysaccharides provide stability to emulsion by forming a network in the continuous phase (Kumar et al. 2020). The polysaccharides like GA and gelatin have emulsifying capacity due to interfacial properties but possess low encapsulation efficiency (Mahdavi et al. 2016). Modified starches, like maltodextrin, have poor emulsifying activity and low oil retention. Conversely, sodium caseinate has excellent emulsifying activity and stabilizes the emulsion but lacks in entrapping liquid oil. However, a combination of maltodextrin with protein or gums has been observed to show better results (Pattnaik and Mishra 2020; Mahdavi et al. 2016). Furthermore, the addition of a surfactant like a lecithin or a caseinate to the above solution might provide a better result (Salminen et al. 2014). Recent studies suggest that microcapsules with better encapsulation efficiency and stability can be achieved by forming a conjugate between protein-polysaccharide via Maillard reaction. Li et al. (2017) reported great emulsion stability at pH 11 and efficiency of about 95% with high oil loading up to 80% on using sodium caseinate-lactose conjugate formed through Maillard reaction. The use of low molecular weight carbohydrates in encapsulation is linked with caking and re-crystallization problems on storage (Gharsallaoui et al. 2007). Milk-based proteins should often be used as encapsulating material as they show good functional and film-forming properties (Gharsallaoui et al. 2007). Aberkane, Roudaut, and Saurel (2014) reported that pea protein could also be considered as a good coating material for encapsulation. The use of lentil protein as effective wall material is also supported by findings reported by Chang, Varankovich, and Nickerson (2016). In this study, the solution of the lentil protein, maltodextrin, and sodium alginate produced rigid microparticles with enhanced oxidative stability and entrapment efficiency (88%). Gomes and Kurozawa (2020) claimed that rice protein after enzymatic hydrolysis by flavourzyme protein hydrolysate demonstrated excellent emulsion stability and had a maximum encapsulation efficiency of 89.5% when used for microencapsulation of linseed oil. Nonetheless, certain protein-based entrapping agents have allergens, precipitate at long-term storage, and denature at high drying temperature depending on drying technique (Haque and Adhikari 2015; Zhao et al. 2013).

Besides the wall materials, the particle size of the microcapsules also governs the oxidation process. Sanchez, Cuvelier, and Turchiuli (2016) affirmed that surface to volume ratio is the main factor influencing the oxidation, because of exchange surface area. Linke, Hinrichs, and Kohlus (2020) observed higher oxidation in smaller particles than larger ones, as the surface to volume ratio was smaller that caused a higher particle-air interface. The larger particle-air interface exposed the surface to oxygen and led to more oxygen diffusion resulting in detrimental changes by reacting with the oil droplets. However, by modifying the encapsulation procedure, the particle properties (porosity, density, size) can be changed which might affect the oxidative stability of oil powders.

# Bulk encapsulation of oil

Traditionally, there are several drying methods practiced for the encapsulation of PUFA rich edible oil. However, due to the inherent unsaturation of the oils, some techniques failed to provide efficient protection against lipid oxidation. The primary reason for the reduced stability is the lower encapsulation efficiency, resulting in the exudation of un-encapsulated oil to the surface. Furthermore, the drying temperature of certain methods caused antioxidant depletion, followed by shelf-life deterioration. In recent years, many novel techniques and technological advancements have emerged to eliminate existing issues (Table 3). This section deals with the working principles, problems, and their related solution of novel technologies along with the most widely used encapsulation methods.

# Spray drying

Spray drying (SD) is the most commonly used drying technique for emulsions. They are widely used by food and pharmaceutical industries for encapsulating flavors, essential oils, fats/omega-rich oil, etc. The ease of scale-up, flexibility of the process, good powder quality, and cost-effectiveness of this method make it a popular one (Kaushik et al. 2015b). The basic steps of spray drying involve dispersion of core material into the wall material solution, formation of a homogenous emulsion, transferring into the feeding pump if the spray dryer, atomization of spray through pressure nozzles, collection of the dried powder microcapsules (Figure 2a) (Albert, Vatai, and Koris 2017). The processing conditions of the spray dryer, such as inlet temperature, feed flow rate, composition, and homogeneity of feed solution influences the size of the dried microcapsules varying from 0.1 to 100 μm fine particles to 2-3 mm coarse ones (Nedovic et al. 2011; Gharsallaoui et al. 2007). Viscosity of the emulsion has a direct effect on the powder particle size. High viscosity emulsions not only interfere in the atomization process but also cause air inclusion inside particles leading to larger sizes (Bakry, Abbas, et al. 2016). The effectiveness of the process depends on the encapsulation efficiency (%EE) of the microcapsules and their related operating conditions. Ghosh, Srivastava, et al. (2019) reported a decrease in %EE when the feed rate and the inlet air temperature of the spray dryer were increased beyond 35 mL/min and 175 °C. The increased feed rate and the high inlet temperature might have encountered insufficient air volume for solvent evaporation and

 Table 3. Different techniques for encapsulation of PUFA rich edible oil and their operating conditions.

Sesame oil 2.4 135 Rice bran 1; — 188 120 Freeze drying Sunflower Coacervation PUFA rich oil blend Microwave power (W): 180  Spray drying Sesame oil 2.4 138  Soybean 0.2–0.3; 1.8 120  Tailored PUFA rich 0.33 150  Sunflower — 136  Freezing temperature Pre (°C,) Time (h) 0.11  Sunflower — 32, 24 — 11  Echium — 40, 2 0.1  Haxseed — 20, —; —70, 24 0.1  Flaxseed 9.0; 3.1 70	Inlet temperature (°C) 135 180; 155 120–160; 150 150, 180 130; 110 150 Iture Pressure (mbar)	Outlet temperature (°C)				
Sesame oil Rice bran Flaxseed Tailored PUFA rich Soybean Sunflower Rice bran Sunflower Echium Flaxseed Flaxseed	ture	temperature (°C)				
Sesame oil Rice bran Flaxseed Tailored PUFA rich Soybean Sunflower Rice bran Sunflower Echium Flaxseed Flaxseed	ture					
Flaxseed Tailored PUFA rich Soybean Sunflower Rice bran Sunflower Echium Flaxseed Flaxseed	ture	80 90; 92–96	1:1, 2:1 1:4; 1:2–4	Tamarind seed mucilage MD, GA, WPC; Pea	91, 81 78; 74	Alpizar-Reyes et al. (2020) Atta et al. (2020); Benito-
Flaxseed Tailored PUFA rich Soybean Sunflower Rice bran Sunflower Echium Flaxseed Flaxseed Flaxseed	ture			protein, MD		Román, Sanz, and Beltrán (2020)
Tailored PUFA rich Soybean Sunflower Rice bran Sunflower Echium Flaxseed Flaxseed	ture	60–80; 75	1:4; 1:0.8	Polysaccharide gums (PSG)	90.78; 99.7	Shahid et al.(2020); Domian
Soybean Sunflower Rice bran Sunflower Echium Flaxseed PUFA rich oil blend	ture	80, 98	4:1	WPC, pectin, MD,	89–93	Vélez-Erazo, Consoli, and
Sunflower Rice bran Sunflower Echium Flaxseed Ying PUFA rich oil blend	ture	I	1:4; 1:2.3	MD, modified starch;	95; 40.15	da Silva James et al. (2019);
Rice bran Sunflower Echium Flaxseed ying PUFA rich oil blend	ture	09	1.22	WPI, sodium caseinate	66-96	Domian et al. (2014)
Rice bran Sunflower Echium Flaxseed ying PUFA rich oil blend Flaxseed		Drying time (h)				
Sunflower Echium Flaxseed ying PUFA rich oil blend Flaxseed	0.15	48	1:2–4	Pea protein, MD	96	Benito-Román, Sanz, and Beltrán (2020)
Echium Flaxseed ying PUFA rich oil blend Flaxseed	I	48	1:2	Sodium caseinate, lactose;	61.2; 77.1	Holgado et al. (2020)
Flaxseed ying PUFA rich oil blend Flaxseed	-	24	1:2, 1:3	GA, sinapic acid	83.27	Comunian et al. (2019)
ying PUFA rich oil blend Flaxseed	1 0.1; —	36; —	0.47; 1:1.5	Sodium octenyl succinate	95.7; 95.4	Domian et al. (2018);
ying PUFA rich oil blend Flaxseed				starch, trehalose; WPI, MD, sodium alginate		Fioramonti, Rubiolo, and Santiago (2017)
Flaxseed	r (W): 180 Temperature (°C)	0.2 Rpm	1:1.85	MD, sodium caseinate, MPI	68	Pattnaik and Mishra (2020)
	40: 50	- :008	1:2: 1:4	Flaxseed protein isolate.	95.4: 87	Pham et al. (2020): Kaushik
				flaxseed gum, transglutaminase; Flaxseed protein isolate, flaxseed gum,		et al. (2016)
Pequi 4.5	40	I	1:4	Cashew gum, gelatin, tannic acid	70.98	Alexandre et al. (2019)
Echium — Linseed 3.5	15 30	600 200	1:2, 1:3 50% oil load	GA, sinapic acid Gelatin-flaxseed mucilage	83.27 >95	Comunian et al. (2019) Mohseni and Goli (2019)
-	;			(FM)-oxidized tannic acid		
Chia seed 2.7	40	I	<u></u>	Chia seed protein, chia seed gum, transglutaminase	93.9	limilsena et al. (2016)
Spray cooling/chilling Feed (°C)	Inlet & outlet air	Feed flow rate (L/h)				
Ascorbic acid 80		9.0	1:4	Palm oil, fully hydrogenated palm oil	93.5	dos Santos Carvalho et al. (2019)
Co-extrusion Core flow rate (L/h)		Hardening time (min)	Ċ	OMIL STORY	0.23	(2000) month bas 11004)
Canola 0.03; 0.03	0.42 0.2; 0.2	120; 10	<u>.</u> [	Alginate, nivir Alginate, quercetin; alginate, HMP, quercetin	07.3 78.3; 68	Waterhouse, Wang, and Sun-Waterhouse (2014);
						Wang, Waterhouse, and Sun-Waterhouse (2013)
Olive —	I	10	0.05	Alginate, caffeic acid	9.09	Sun-Waterhouse et al. (2011)

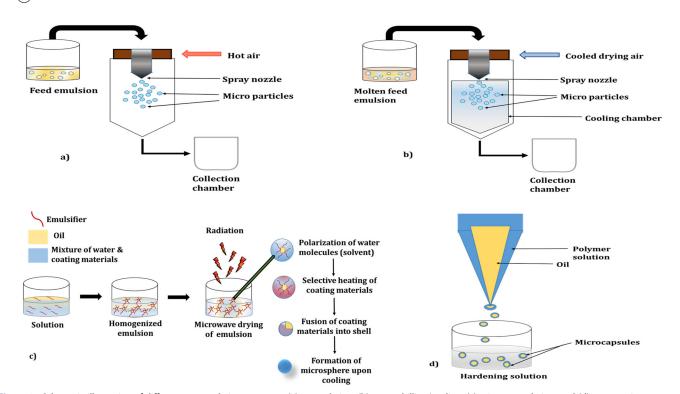


Figure 2. Schematic illustration of different encapsulation processes (a) spray drying, (b) spray chilling/cooling, (c) microwave drying, and (d) co-extrusion.

protein denaturation, respectively. Encina et al. (2016) reported that microcapsules produced with low outlet temperature has high moisture content and a rubbery state while high outlet temperature results in dents and cracks that aids oxidative deterioration along with excess oil release. The wall material composition also governs the quality of the powders obtained from spray drying. Drusch et al. (2006) observed the crystallization of the low molecular weight carbohydrates used as wall materials when the inlet temperature was raised beyond the glass transition temperature, thereby exposing the core substance out of the matrix. However, the crystallization can be avoided by the optimum mixing of low molecular weight carbohydrates with proteins due to their high glass transition temperature (Adhikari et al. 2009). Islam, Edrisi, and Langrish (2013) found an increase of %EE from 20 to 60 at high humid conditions on adding whey protein isolate to lactose solution. The presence of protein film reduces the particle-air also particle-particle interaction due to its surface activity and low diffusivity of the molecules. Additionally, the emulsification process also determines the droplet size, which has a noticeable impact on the %EE. The processes resulting in smaller oil droplet sizes are responsible for high %EE due to the perfect enclosing of the core within the polymer matrix. Few researchers have investigated the effect of mechanical homogenization (MH; rotor-stator), micro fluidization (MF), membrane emulsification (ME), and ultra-sonication (US) on the surface oil content and its %EE. They have claimed smaller and monodisperse particle size with low surface oil and high %EE in the order: MF > US > ME > MH (Albert, Vatai, and Koris 2017; Ramakrishnan et al. 2014; Jafari et al. 2008). Many studies show that an increase in the homogenization pressure (>80 MPa) and homogenization cycles (>5) cause a rise in temperature and decrease in particle size which leads to the aggregation of particles along with the formation of primary oxidation products (Kuhn and Cunha 2012). Additionally, El-Messery et al. (2020) compared spray-dried (inlet, outlet temperature—130 °C, 71%-75 °C, respectively) and freeze-dried (−50 °C condenser temperature, 0.04 Mbar vacuum pressure) microcapsules produced from maltodextrin, gum Arabic, and krik oil. Their findings suggest that the spherical-shaped spray-dried particles (62.2%-78.8%) had better encapsulation efficiency than irregular-shaped porous freeze-dried particles (51.9%-58.2%) owing to lower gas permeability and provided extra protection to the core.

However, spray drying has few limitations, such as high product temperature accelerates powder porosity and lipid oxidation. Increased oil loading beyond 20%-25% is associated with an increase in wall materials concentration which leads to high viscosity, higher viscosity emulsion arises problem during atomization through nozzles, crystallization of sugars, and sticking of powder on to the dryer chamber reduces the yield (Kaushik et al. 2015b; Ramakrishnan et al. 2014). Several researchers have prepared multilayered emulsions to promote better lipid protection that also contributed to high oil loading (Vélez-Erazo, Consoli, and Hubinger 2020; Jiménez-Martín et al. 2015; Carvalho, Silva, and Hubinger 2014). Fioramonti et al. (2019) claimed that spraydried flaxseed oil powder prepared by layer-by-layer deposition technology of double emulsion contained a high oil load (up to 66%), whey protein concentrate (WPC), and sodium alginate that had an encapsulation efficiency of  $\sim$ 84%. The prepared particles showed good oxidative stability than the liquid oil for up to 6 months at -18 & 4 °C storage temperature. Similarly, Chang, Varankovich, and Nickerson (2016) entrapped an extra 20% canola oil by adding sodium



alginate in conjunction with lentil protein isolate (LPI) and maltodextrin. Despite high emulsion viscosity and larger droplet size, they observed a high encapsulation efficiency ( $\sim$  88%) due to a strong electrostatic complex between LPI and sodium alginate. Moreover, the drying of microcapsules through cross-linking and different techniques can be utilized to eliminate the problems of aggregation and adherence of the microparticles on the walls of the spray dryer.

# Spray chilling/cooling

The basic process of spray chilling is similar to spray drying. The steps consist of dispersion of core into encapsulating matrix, preparation of homogenous emulsion, feeding the emulsion through nozzles, atomization of the solution, solidifying the matrix, and collecting through the cyclone and filter bags. The main difference lies in the drying chamber; here, the atomized particles through the nozzle fall into the cooling chamber where the particles are gelled or solidified (Figure 2b). In spray drying, energy is applied for solvent evaporation, whereas, spray chilling involves energy removal for solidification. The solidification process is the function of cooling chamber and its material properties. The temperature in the cooling chamber must be regulated below the melting point or gelling point of the solid. Moreover, the matrix materials in the solution must quickly solidify in the chamber before reaching the collection chamber. The cooling capacity and chamber size depend on the size and surface area of the atomized particle (Oxley 2012). The lipid particles produced by the spray cooling process had a wrinkled but spherical surface, possessing a high entrapment efficiency of 92%-96% (Ribeiro, Arellano, and Grosso 2012). This process can also be utilized for encapsulating temperature-sensitive materials in a lipid matrix as it does not require organic solvents, for instance, Matos et al. (2015) prepared solid lipid particles loaded with ascorbic acid by spray cooling technique. He claimed increased storage stability of the loaded material and better encapsulation efficiency up to 84%. Similarly, Xiao et al. (2020) investigated docosahexaenoic acid (DHA) microcapsules enclosed within dodecenyl succinic anhydride-esterified agarose (DSAG) having a good entrapment efficiency (65%-85%), They claimed that 0.03 MPa atomization pressure is optimum for producing larger and uniform sized microcapsules, on contrary, microparticles tend to aggregate and they are difficult to prepare with a spray pressure beyond 0.04 MPa. Few studies have shown that the modulation of the atomization process in the spray cooling technique the polymeric forms of hydrogenated fats could be changed to  $\alpha$ -form, hence this could potentially be used in confectionery or as enhancers for the crystallization process (Lopes et al. 2015).

The spray chilling process has been effectively used to encapsulate heat-sensitive PUFA rich oils to delay oxidation, in pharmaceutical industries, or for enclosing probiotics and enzymes for minimal thermal loss. The absence of organic solvents makes this process inexpensive. In comparison to spray drying, it has a higher production rate and reduced exposure to elevated temperature (Oxley 2012). Moreover,

the process yield of spray chilled microcapsules was found to be higher than spray dried microcapsules (60% vs. 45%) without the occurrence of any agglomeration in the microparticles (Fadini et al. 2018). However, the process has a limitation of the coating matrix with respect to viscosity and solid concentration. The solid concentration in the slurry should be optimized to minimize the viscosity for the production of small atomized droplets, and this will enhance the production rate and cooling capacity of the chamber.

# Freeze drying

Freeze drying (FD) is a simple and less complicated process usually preferred for encapsulation of heat-sensitive materials and essential oils. The PUFA rich oil emulsion is first frozen at a low temperature between -90 and -40 °C, then the frozen mixture is sublimed from solid to a gaseous state at reduced pressure. Although it is a batch process, yet it has high nutrient retention than the spray drying process. Therefore, it is suitably used for or -rich oils like fish, linseed, virgin olive, and WO. A good encapsulation efficiency of about 99 and 84% was observed for extra-virgin oil and flaxseed oil, respectively, with protein as wall constituents (Calvo et al. 2012; Karaca, Nickerson, and Low 2013). The microcapsules formed by the freeze-drying process show an exceptionally high protective effect against lipid oxidation because of the antioxidants retention during the storage period at ambient temperature (Karaca, Nickerson, and Low 2013). The combination of various wall materials (maltodextrin: whey protein: arrow root; maltodextrin: whey protein; whey protein: arrow root) was investigated and compared by Charles et al. (2021). They claimed that the microparticles produced from the combination of maltodextrin, arrowroot, and whey protein showed better oxidative stability with improved entrapment efficiency (>80%) of fish oil for 90 days at 25 °C storage temperature due to the cryoprotective behavior of arrowroot which aided stabilization and protection of air-sensitive fish oil. On the other hand, Perrechil et al. (2021) experimented with rice protein concentrate (RPC) and modified starch for producing freezedried flaxseed oil microcapsules. They discerned that RPC had a poor emulsifying ability (encapsulation efficiency, EE <1%), while on increasing the content of modified starch, the EE improved from 12.9 to 90.6%. Besides displaying good oxidation stability, few studies show low microencapsulation efficiency due to the presence of a porous, irregular flaky structure (Anwar and Kunz 2011; Velasco, Dobarganes, and Márquez-Ruiz 2003). Similar porous structures were observed by Rodriguez et al. (2019) on encapsulating chia seed oil in sodium caseinate and lactose (core:wall—1:2) possessing an excellent encapsulation efficiency (84%). The porous structure of such oil powders favor easy ingress to oxygen, thereby accelerates the oxidation process. To include higher oil content, Fioramonti, Rubiolo, and Santiago (2017) prepared multi-emulsion by ultrasonication (75% Amplitude, 150 s, 20 kHz) using maltodextrin, sodium alginate, and WPI as wall materials. Although, they observed a significant increase in

encapsulation efficiency from 27% to 95% on increasing the maltodextrin content (0 to 20%), yet, 10% maltodextrin concentration showed excellent oxidative stability. The presence of thicker interfacial layers at high maltodextrin concentration might inhibit the interaction between the continuous phase and liquid oil, additionally, emulsification by ultrasonication triggers lipid oxidation due to high local intensities caused by cavitation.

In a study conducted by González et al. (2016), the EE did not show any significant difference regardless of the drying methods, i.e., spray and freeze-drying, mainly because of the high solids concentration (wall materials: oil—2:1) that prevented the migration of oil particles on to the surface. Conversely, Domian et al. (2018) reported a higher EE in spray-dried linseed oil powder (EE >95.88%) than freezedried microcapsules (EE <95.73%). The major drawback of this process, in contrast to other processes, is the long processing time, high energy use, and production cost (Prosapio, Norton, and De Marco 2017). Howbeit, the use of low temperature in freeze-drying process would be beneficial for microencapsulation of unsaturated fatty acids by overcoming the porosity issue with certain technical advancements like temperature-regulated nucleation during freezing. The spray and freeze-drying can be combined to obtain fine unagglomerated powder without any heat damage. The spray-freeze drying (SFD) is relatively a new technique that involves rapid freezing of atomized droplets in cryogenic gas or liquid, later the frozen water is sublimed to produce final dry microparticles (Ishwarya, Anandharamakrishnan, and Stapley 2015). Pang et al. (2017) compared spray drying, freeze-drying, and spray-freeze drying of fish oil microcapsules developed using acacia gum, sodium alginate, and Tween-80 (3:1:0.1). From their study, they concluded that freeze-drying takes a longer time (36h) and high overall cost, while spray drying has a shorter drying time (2h) and poor powder quality affecting its yield. Contrarily, SFD produced uniform-sized microparticles with larger surface area, excellent powder quality, and better entrapment efficiency (EE of SD, FD, SFD-72.64%, 49.7%, 90.8%, respectively).

# Microwave drying

Microwaves are electromagnetic radiation produced by magnetron in the presence of both electric and magnetic fields. Microwave drying of agricultural food products is a familiar technique often employed for heat-sensitive food products because of its low product temperature, shorter drying time, energy efficiency, and higher drying rate (Chandrasekaran, Ramanathan, and Basak 2013). Theoretically, microwave drying is a volumetric direct heating method that produces heat by the rapid movement of polar molecules. The polar molecules re-orient themselves in the direction of an electric field. However, due to high electric field frequency, these polar molecules align and realign themselves rapidly about million times per second, generating heat by internal friction, thus resulting in volumetric heating (Chandrasekaran, Ramanathan, and Basak 2013). This phenomenon builds up a vapor pressure gradient between the external and internal surrounding, which up thrusts the moisture out, hence, accelerates the drying rate and shortens the drying time.

The application of microwave for drying of microencapsulated oil emulsion is a relatively novel idea. Limited literature suggests that microwave drying of microencapsulated emulsions is less explored. Hassan and Muhamad (2017) attempted drying of oil emulsion containing perah seed oil with a high amount of 0 -3 fatty acid by combining freezedrying with microwave technique. He reported that the produced microencapsulated oil powder had an irregular porous surface; however, they effectively protected the oil inside the solid matrix due to lower gas permeability. Similarly, Pattnaik and Mishra (2020) claimed improved physicochemical properties of microencapsulated oil powder by using only the microwave technique to dry oil-in-water emulsion. They have demonstrated a comparatively excellent entrapment efficiency (nearly 90%) despite porous structure with improved antioxidants retention and powder flowability with other conventional drying methods. The selective heating of coating and core materials in the emulsion causes significant entrapment of active ingredients within the microcapsule (Figure 2c). For successful encapsulation of oil, the wall and core materials' dielectric constants and dissipation factors must be substantially different. This differential dielectric factor causes the heating of wall materials alone, which upon cooling diffuses around the core to form a hard shell (Pattnaik and Mishra 2020). Besides the numerous advantages, microwave drying produces a porous powder structure, and at higher microwave power levels, there might be an occurrence of some non-enzymatic browning or protein denaturation. Nevertheless, the above issue can be easily controlled by fine-tuning the optimum operating conditions.

#### Extrusion

Extrusion has been potentially used to produce high-density microencapsulated products like omega-rich oils, flavor, essential oils, and enzymes. This process involves the mixing of molten wall materials with the core materials followed by solidification. The dispersed core in the hot melt is extruded through a single or twin-screw system at high pressure (Akoh 2017). This process increases the storage stability of the less porous microparticles due to their glassy state, but it requires a lower oil load during encapsulation. Moreover, it is an expensive technique when compared to spray drying, and the high shearing caused by high pressure during extrusion might affect the stability of sensitive materials like unsaturated oil (Kaushik et al. 2015b; Saerens et al. 2011).

Co-extrusion is an alternative extrusion technology which applies a jet atomizer equipped with concentric nozzles to form emulsion droplets (Figure 2d). The matrix solution is extruded through the outer tube, and the core material is extruded out of the inner tube, then the formed droplets are passed into a carrier fluid for hardening of the microcapsules (Whelehan 2011). Chew and Nyam (2016) extruded emulsion containing alginate and kenaf seed oil through vibrating nozzles into calcium chloride solution for gel hardening. They reported stable microspheres with 0.2 water

activity (a<sub>w</sub>) and 76.62% encapsulation efficiency. The use of concentric nozzles can induce higher oil loading in the microcapsules ( $\sim$  90%). Alginate is commonly used for coextrusion since it has lower toxicity, chemical stability, and good cross-linking capacity in the presence of calcium ions. The entrapment efficiency is influenced by the extent of cross-linking at the surface of the extruded droplet and the emulsion stability (Dolçà et al. 2015). The experimentation conducted by Chan (2011) demonstrated a high encapsulation efficiency and good emulsion stability when the oil-towall weight ratio was kept up to 15 g/g. However, the particle size of the microcapsules produced by the gravitational force is reported to range between 2 and 7 mm, which might affect the food product's mouthfeel (Martins, Poncelet, Rodrigues, et al. 2017). Besides, the nozzle diameter and feed flow rate can be regulated to obtain the desired size particles. In some cases, electrostatic dripping is adopted to produce smaller uniform-sized microcapsules where the oil emulsion is extruded by placing the nozzle under an electrical potential difference. The electrostatic forces accelerate the droplet fall rate that is comparatively faster than other dripping processes (i.e., under gravitational force) (Martins, Poncelet, Rodrigues, et al. 2017). Droplets fallen are collected in a bath where it is hardened through the cross-linking method. Martins et al. (2015) stated the influence of curing time and alginate-calcium chloride concentration on the membrane thickness and diameter of the oil microcapsules. They noted the formation of a thicker membrane after 20 min of curing with the maximum amount of calcium ions released from the oil core. This thicker membrane is beneficial for providing excellent protection to the core material (Abang, Chan, and Poncelet 2012). However, a longer curing time results in a weaker membrane structure due to the migration of calcium ions from the membrane to the solution (Martins, Poncelet, Marquis, et al. 2017). Lower alginate content (<15 g/L) did not affect membrane thickness, similarly, higher calcium chloride concentration (<4.6 g/L) produced well-structured microcapsules (Martins et al. 2015).

Extruded microcapsules can be further dried by various drying methods to improve the integrity of the structured microparticles. For example, Menin et al. (2018) prepared flaxseed oil microcapsules using low methoxyl pectin (15% oil/pectin) by vibrational extrusion technology, followed by active drying (fluidized bed drying) and passive air drying. The fluidized-bed dried micro particles possessed higher surface oil owing to the porous structure, while both the drying techniques displayed good encapsulation efficiency (>96%). The major limitation of this encapsulation technique is the frequent clogging of the nozzles because of the viscous polymer solution, thus interferes in the drop generation.

# Application of microencapsulated oils

# **Food industry**

The growing awareness about the benefits of consuming PUFA enriched food products has urged the food industries to incorporate them into the food product. Marine animals like fish are endowed with omega fatty acids, but the concern of the vegan race limits its use in the food product. Therefore, most food technologists have switched over to vegetable-based omega-rich oils. However, the underlying problem with the inclusion of PUFA enriched vegetable oil their susceptibility to oxidation during storage. Henceforth, microencapsulated oil in the form of gel or powder has been successfully utilized in food products.

The oleogels were associated with the replacement of trans and saturated fats in breakfast spreads, confectionery, dairy products, meat products, and sweets. Oleogels prepared with SFO and 2% shellac without any emulsifiers were stable for around 4 months, the crystallization of shellac wax aided in the stabilization of the oleogel (Patel et al. 2014). Similarly, spreads obtained from virgin OO with 7% monoglycerides (MG) had similar textural and thermal properties to commercial margarine with storage stability of about 3 months (Öğ ütcü and Yılmaz 2014). The replacement of shortening in muffins with foam oleogels of SFO with 4% HPMC up to 50% level displayed acceptable physical, textural and sensorial properties (Oh and Lee 2018). Furthermore, a full replacement of margarine in muffins with high oleic SFO and 4%, 7%, or 10% MG provided desired results of lowering saturated fats, acceptable properties, and lowered migration of oil up to 50% (Ergun, Thomson, and Huebner-Keese 2016). Zulim Botega et al. (2013) explored the replacement of dairy fat (4%, 8%, 15%) with rice bran wax and glycerol monooleate-based oleogels. He claimed that glycerol monooleate was responsible for fat networks forming during ice-cream preparation with favored desirable characteristics like overrun and melting. The restriction of fat migration on to the surface of halva was achieved by adding oleogelators made up of sunflower wax, bee wax, and shellac wax (Oğütcü, Arifoğlu, and Yılmaz 2017). Lim et al. (2017) deep-fried instant noodles in oleogels made up of soybean oil and carnauba wax instead of soybean or palm oil. He reported that there was no adverse change in the texture of the noodles and the noodles also absorbed less oil.

Fortification of food products with oil powder was also reported in the literature. For instance, Goyal et al. (2015) reported that the fortification of milk with flaxseed oil powder showed comparable sensorial aspects with storage for up to 5 days. He also stated that this flaxseed oil powder, when added to milk, met the nutritional necessity of  $\omega - 3$  fatty acids in non-fish eating or vegan eaters. The microspheres of linseed oil dried by spray drying when incorporated in soup formulation at 14% fulfilled 80% of the daily requirement of  $\alpha$ -linolenic acid. In addition to it, the oil powder showed excellent oxidative stability for 8.78 months (Rubilar et al. 2012). RBO powder, when added to yogurt, increased the acidity and water holding capacity during storage; however, the product was acceptable from sensorial aspects (Atta et al. 2020). Yogurt and bread fortified with palm oil powder using complex coacervation method and chitosan/xanthan gum and chitosan/pectin as wall materials show appreciable stability and release in the gastrointestinal fluids (Rutz et al. 2017).



# **Pharmaceutical industry**

The poor bioavailability and absorption of some compounds/drugs in the human digestive system have encouraged the need for a proper delivery system. The lipid-soluble compounds can be effectively structured into a matrix of crystalline sphere. While formulating any delivery system, one must consider the solubility and micellarization of the enveloping material and its effect on nutrient bioaccessibility and digestibility. Most of the lipid digestion and absorption occurs in the stomach and small intestines (Mei et al. 2006). Therefore, the micelles are formed when vegetable oil comes out of the enveloping matrix, and these micelles serve as the drug vehicle in the gastrointestinal fluid carrying the active component (Davidovich-Pinhas 2016).

The carotene from 12-hydroxystearic acid canola gel showed release of carotene from oil between 0 to 30 min during the digestion in intestine, whereas the same had a controlled release between 30 to 75 min when an oleogel was used (Stortz et al. 2012). A similar oleogel emulsion was prepared from zein protein and used for delivery of carotene demonstrated good color stability, protection, and retention of active compounds (Chen et al. 2016). An oleogel composed of canola, corn, or coconut oil with monostearin and Span 20 enclosing 2.6% curcuminoids showed high bioaccessibility than curcumin powder on dispersing in water and nearly 5 times higher during the fasted state. The storage stability improved, and there was no occurrence of precipitation (Yu et al. 2012). Almeida et al. (2008) formulated a bi-gel for topical application composed of oleogel and hydrogel, which showed good spreadability and stability for up to 6 months. Besides this, it provided enhanced cooling and moisturizing effect in the absence of tensoactives. Bochot et al. (2007) claimed an efficient delivery of Isotretinoin, a poorly soluble lipophilic molecule into a self-assembling lipid bead system consisting of soybean oil and α-cyclodextrin. The soybean oil beads could potentially be used in pharmaceutical applications (oral or topical) as well as in cosmetics. Microspheres of alginate-pectin and calcium-pectinate prepared from the emulsion-gelation method were capable of forming floating beads of Ranitidine hydrochloride or other oils for intragastric conditions (Jaiswal et al. 2009; Sriamornsak, Thirawong, and Puttipipatkhachorn 2004). The oil-loaded calcium-alginate beads produced from the emulsion-extrusion method had a positive antifungal effect against Aspergillus niger and Fusarium verticillioides fungi species (Soliman et al. 2013). Marefati et al. (2015) reported a promising Pickering double emulsion to prepare the oil powders from octenyl succinic anhydride (OSA) modified quinoa starch with high encapsulation efficiency and oil content of about 97% and 70 wt.% respectively for its effective use in pharmaceutical industries. There is also development of several non-chemical-based mosquito repellents and ointment containing encapsulated essential oil microcapsules encouraged as a promising alternative offering a longer duration of action with desirable characteristics (Solomon et al. 2012).

### Pesticides, fungicides, and insecticides

Pesticides and fungicides are used enormously to minimize post-harvest losses and food deterioration. However, due to the well-known adverse impact of these pesticides and fungicides on the environment and human health, there is an urgent need for alternative practices. The delay of postharvest decay can be achieved by applying encapsulated essential oils extracted from various plants which are environment friendly as well as biodegradable. For instance, encapsulated essential oils from Rosmarinus officinalis, Salvia mirzayanii, Artemisa persica, and Thymus vulgaris were applied to mango fruit for controlling its decay caused by A. niger, thereby enhancing the storage life and maintaining the internal quality of mango fruits (Javadpour et al. 2018). Many insect-resistant packaging for food products has attracted attention from the food industry, such as thyme oil and cinnamon oil used as microcapsules or as films containing microcapsules for effective repelling of insects and/or moth larvae by releasing an effective insecticide (e.g., cinnamaldehyde in cinnamon). These films have excellent tensile properties helpful in preventing the invasion of larvae about 90% into food products (Chung et al. 2013). Additionally, an improved anti-fungal activity by slow-release of Cuminum cyminumc essential oil encapsulated in chitosancaffeic acid nanogel was presented by Zhaveh et al. (2015). Kulkarni et al. (2000) prepared neem seed oil beads encapsulated in sodium alginate-glutaraldehyde polymers incorporated in liquid pesticide for controlled release in soil. A green approach involving environmentally friendly pesticides against the insect pest Myzus persicae was developed by encapsulating Pennyroyal (M. pulegium) essential oil into baker's yeast via diffusion through the cell membrane that showed efficient insecticidal activity for a period of 3 days (Kavetsou et al. 2019).

# Lubricants, textile, and personal care

Nowadays, mineral oil-based lubricants have created a negative impact on society owing to their toxicity and inhibition of plant growth. There is more demand for eco-friendly, bio-degradable, and pollution-free lubricants. Vegetable oils can be a promising alternative to mineral oils for boundary and hydrodynamic lubricants because of their high viscosity index, low volatility, biodegradability, and nontoxicity (Sharma, Adhvaryu, and Erhan 2009; Suzuki, Ulfiati, and Masuko 2009). Compared to traditional soap-based lubricating greases, the oil gels do not demand sophisticated manufacturing equipment or expertise. Martín-Alfonso and Valencia (2015), reported such potential oleogel for lubricating grease manufactured from ethylene-vinyl acetate copolymer (EVA) copolymer with 28% vinyl acetate (VAc) content, SFO, and high oleic SFO. EVA is a thickener agent possessing flexibility, adhesive, and fracture toughness properties. Linear viscoelasticity with a frequency of oleogels was similar to the commercially available lithium greases. The increase of linear elasticity with EVA concentration indicated a strong microstructural network. However, more related data about the tribological behavior of EVA with

vegetable oils are needed to be studied to find out its various applicability. Some aromatic oil encapsulated within the polymer matrix could be potentially used as a functional textile product at spa centers, personal cares, or for aromatherapy (Carvalho, Estevinho, and Santos 2016). Sarıışık, Okur, and Asma (2012) reported similar use of berry oil capsules encapsulated by  $\beta$ -cyclodextrin and later incorporated into 100% cotton towel fabric. Insect repellant cotton fabrics are manufacture by including microencapsulated biopesticides through impregnation or surface coating of the textiles. Similar experiments were conducted by Miro Specos et al. (2017) to contain microencapsulated citriodiol or citronella essential oil pads within cotton woven fabrics. These cotton fabrics exhibited extended durability and 100% repellency for more than 30 days.

# **Future scope**

Future research must be more focused on the technological advancement of existing methods like spray drying. The improvization of pressure nozzles to withhold high viscosity without creating lumps can be attempted. The modification of nozzles successively can support a higher oil load. Nonetheless, microwave drying favors high oil load; a future suggestion would be to model this process as a continuous one. Spray cooling is generally preferred for encapsulation of probiotics, essential oils, or bioactive compounds; hence, future research can be conducted on vegetable oils. In this way, a better powder product can be obtained without compromising the other properties. Despite the methods described above, the use of supercritical fluid (specifically carbon dioxide) for the production of atomized spray particles can be explored in the near future. This supercritical carbon dioxide will not only eliminate the solubility issue but also produce particles of low temperature. The authors also suggest the adoption of biopolymers prepared from industrial or agricultural waste products to be utilized as carrier material. In addition to their abundance availability, they are also biodegradable and nontoxic. The employment of such polymers will lessen the environmental pollution load to some extent.

#### **Conclusion**

The gradual shift of the population toward vegan products has urged the food industries to look for alternative vegetable oils rich in omega fatty acids than marine oils. Marine oils (mostly fish) have long-chain fatty acids such as EPA and DHA, while vegetable oils are endowed with short-chain fatty acids like linoleic and linolenic acid. Moreover, the fatty acid composition of vegetable oils varies based on origin and source. The vegetable oils have to be tailored for balancing their fatty acids without producing any unwanted or toxic compounds like trans-fats. Henceforth, blending is the most economical way of achieving the desired balanced fatty acids with improved nutritional, functional, and thermal properties. The use of ternary blending should be explored more than binary blending since they deliver better

characteristics than the latter. The susceptibility of such PUFA enriched vegetable oils to oxidative degradation has beseeched the technologists toward its preservation through encapsulation. The liquid oil can be effectively entrapped into a matrix of gel or powder in numerous ways. Spray drying and coacervation are considered to be the most commonly used techniques. Nonetheless, there has been an emergence of a few other novel methods like microwave, spray cooling, spray-freeze drying . The encapsulated oil microparticles in the form of gel or powder have a wide application in all the fields viz. food, pharmaceutical, and textile industries along with personal care products and agriculture because they are environment friendly, pollutionfree, biodegradable, and favor controlled release of core particles.

### **Author contributions**

Monalisha Pattnaik: conceptualization, co-developed the methodology, drafted the manuscript by analyzing the literature and heavily involved in curating the manuscript. Hari Niwas Mishra: Supervision, conceptualized the idea, codeveloped the methodology, involved in review & editing.

## **Conflicts of interest**

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