**Review article**

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# Halal edible biopolymers used in food encapsulation: a review

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**Abstract**

**Background and objective**: Encapsulation is extensively used in food science and technology for protection of bioactive compounds by using several materials originated from different animals and plants. However, application of some of them is limited due to their suspicious halal origin. In this regard, gelatin is of concern for industrial application since it can be obtained from pork. At this review, we tried to investigate the halal biopolymers frequently used in food preparations.

**Results and conclusion:** Biopolymers have incredible impact in the formula as gelling, stabilizing, viscosifying, and coating agent. Other than the favorable roles of biopolymers, their origin is important by some countries to find out they are derived in accordance to halal concepts. Therefore, those derived from halal animals through the process which does not threat the halal status are accepted by Islamic countries. In such cases, alternative biopolymers such as agar, pectin, and carrageenan by similar features for the specific purpose should be used instead of gelatin derived from pork.

*Keywords*: Biopolymer, coating materials, encapsulation, halal

## 1. Introduction

Muslims consume halal foods in accordance to the Islamic dietary law. These people concern about content of their foods including the raw materials and the additives [1]. Materials of concern are included to blood and muscle of haram animals and also alcoholic beverages. Today, there are several methods for production, processing, and development of functional foods; among them use of encapsulation is increasing. Encapsulation is an ideal method for protection of bioactive compounds against harmful conditions. During the process, selection of safe materials as shell, capsule or coating is important [2]. Proteins and hydrocolloids have been widely used in this regard. Among hydrocolloids, gelatin is of common materials which is derived from various sources and its halal status should be investigated before use [3-6]. Gelatin is derived from collagen that is a translucent and flavorless animal protein. Other than its role as gelling agent in food, it is used in medicine and other biological systems [5,7].

According to the report Dinar Standard in 2019-2020, global market of halal food costed 1.37 Trillion USD in 2018 and are expected to reach 1.97 Trillion USD in 2024 [8]. The number of gelatin industries in the Middle East is too

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limited, while European and American countries, which mainly used pork gelatin, are larger sources of production. The halal food market will account more than 17.4% of the world's food expenditure in the future. Therefore, such hydrocolloids can make a great opportunities in business of the halal market through halal capsules are producing for food and pharmacy industries and utilizing alternatives as natural resource or halal animals for halal food production in encapsulation methods instead of non-halal animals [5].

Cheating and adding mixed gelatin, cross-contamination, and mislabeling of halal food products with non-halal substances are of challenges in halal food market [6]. Therefore, investigation of different origins and use of halal-based gelatin sources for production of halal-based food and drug products is crucial to assure halal status of the final product. Malaysia is an outstanding halal food producers in Asia and follows the Malaysian Halal Food Standard [9] as a regional and global halal standard. Gelatin is also of concern whose halal status can be determined by this standard. Importantly, if we are not able to identify source of the materials, it can be done by analytical instruments including HPLC, GC-MS, PCR, and biosensors [10].

The aim of this review is investigation of suspended materials such as gelatin used in encapsulation of halal foods and medicines. In addition, halal assurance principles to make sure of their halal status are addressed.

## 2. Encapsulation

Encapsulation processes have been developed about three decades. The main application is coating or entrapment of a sensitive material to protect of its contents from some destructive factors in environment which these materials usually can be a liquid, solid or gas. Another names of them are internal phase and core material. There are some names for coating material including wall material, carrier or shell. Core material can include probiotics, antioxidants, flavors, aromas, pigments, drugs, detoxicants, etc. (Figure 1) [11,12]. Also, coating materials can be carbohydrates, proteins, hydrocolloids, lipids, etc. This method has many forms and some of them and some of encapsulation methods were shown in Figure 2.

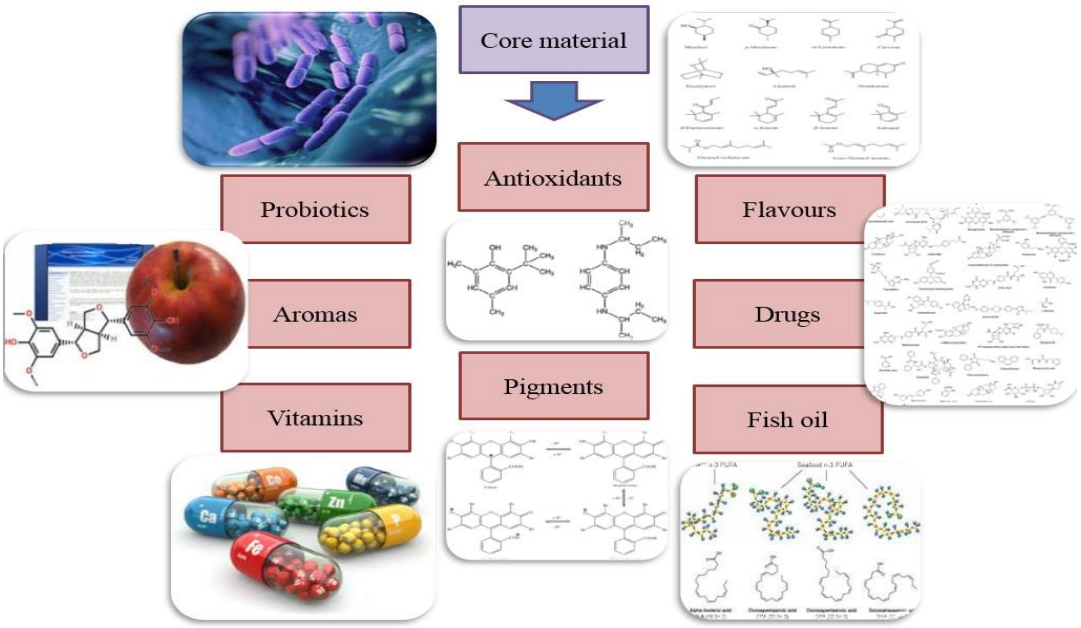


Figure 1- Some of bioactive core materials frequently used in encapsulation techniques

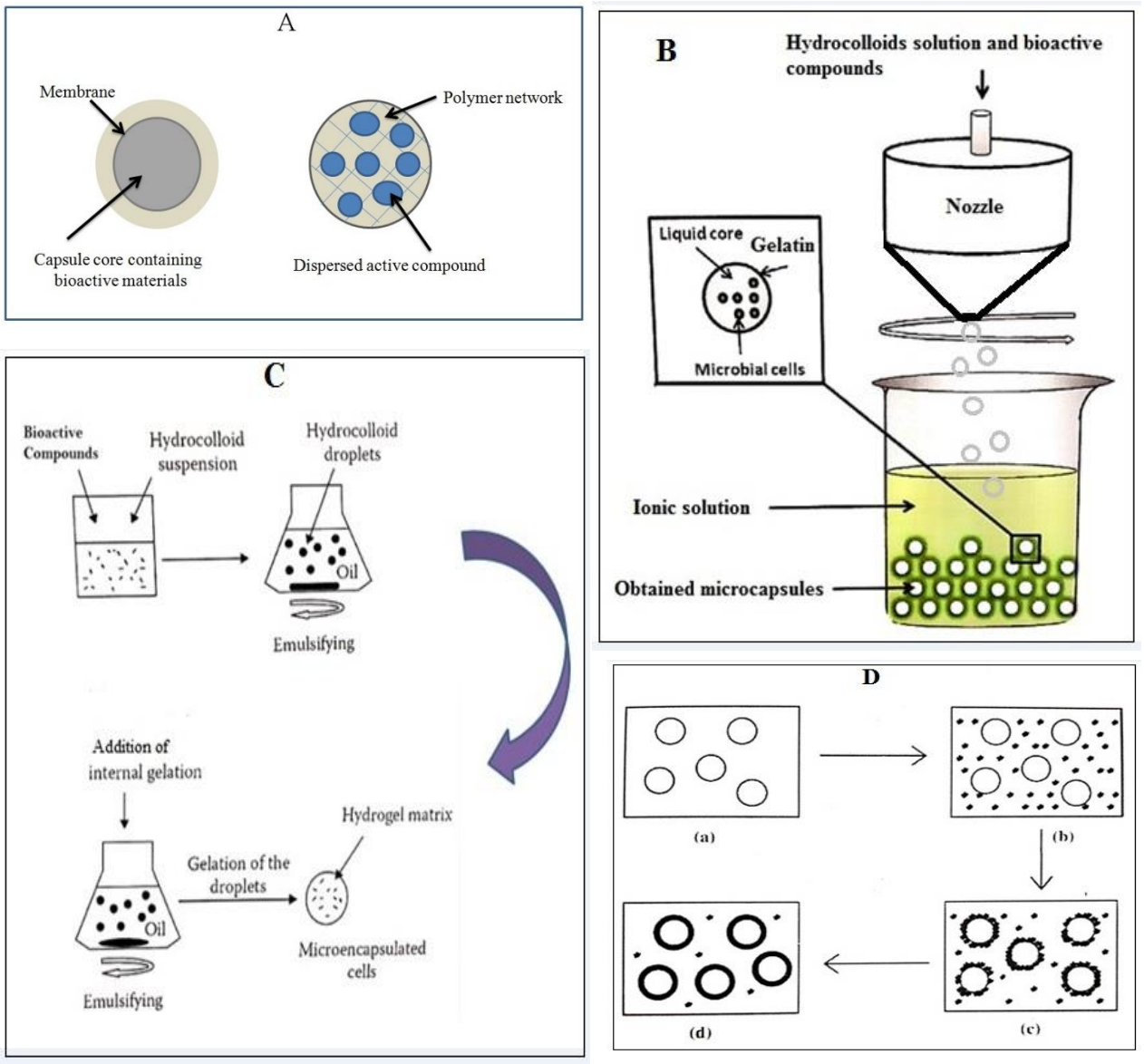


Figure 2- A) Two forms of encapsulates; Three methods of gelatin encapsulation: B) extrusion, C) emulsification, and D) coacervation including a: dispersion of core materials in a polymer coating, b: separation of coacervates from the solution, c: cover of the core materials with small droplets of the coacervates, d: accumulation of the coacervates to form a continuous coating around the cores.

Encapsulation technologies have been used in pharmaceutical industry for many years and now they are developed in other industries such as foods, chemicals, and cosmetics to enhance stability of the materials [11-13]. There are various methods for gelatin encapsulation including spray drying, spray chilling, fluidized bed coating, emulsion, extrusion, freeze drying, coacervation, liposomal encapsulation, etc. Jafarpour et al. optimized nano-encapsulation of omega-3 fatty acids by using coacervation method with response surface methodology (RSM). They produced nanoparticles containing omega-3 made by rainbow trout gelatin and Arabic gum using complex coacervation method [14]. In a study, encapsulation of curcumin was done with gelatin by electro-hydrodynamic atomization to improve antioxidant and antimicrobial properties of curcumin [15]. In another study, encapsulation of oregano essential oil was done by gelatin and chia mucilage. The spray dried powders had encapsulation efficiency more than 80% [16].

## 3. Halal materials

As stated in Quran, Halal is defined as permissible or lawful to use for Muslim people. There should be an optimum for halal food in life for every Muslim consumer that can influence on health, spiritual equilibrium and emotional sense. The global community in accurately labeling the origin of species used in halal foods has made food supply chain management beyond reliable and without validation support [17].

The subject of eating halal food is important for Muslim because God in the Quran commands human being eating halal and lawful foods. These materials and products include halal foods that are permitted under Islamic law and must be free of any prohibited ingredients. So followers of some religions, including Islam and Jewry, are forbidden from using the non-halal and non-Kosher products [18]. God mentioned in the Quran, Surah Baqareh, verse 168: "People! Eat everything that is halal and clean in the ground and do not obey devil who is the obvious enemy".

About 80% of edible gelatins in Europe are made from pig skin, but there are also gelatins derived from vegetables, seaweeds, fish bones and non-pig sources that are halal [19-21]. Instrumental approach can be effective in determining fraud and compounds in foodstuffs. In studies by FT-IR spectra, has been shown sharper peaks of pig gelatin type compared to bovine type [1,22]. Also, in the HPLC method along with mass spectrometry, high sensitivity was observed in the discrimination of beef and pork [23]. A sensitive biosensor has been developed to detect the pig type from beef type in food products [24,25]. Gold nanoparticles (GNP) are embedded in products as a powerful sensor for separating a variety of pig compounds from beef. This method based on color changes that are visually recognizable [26,27].

Since the middle of this century, the world's population has been increasing and doubling, while demand from some consumers, especially in industrialized countries, has led to the production of food products through the use of healthy and nutritious foods. Therefore, the use of modern technology in the food industry is leading [12].

One of these techniques is the encapsulation of bioactive compounds with nutritional value. This technique is used to achieve the highest stability of these compounds in food products. The sensitivity of some bioactive compounds such as proteins, antioxidants, vitamins, minerals, enzymes, probiotics and nutrients to environmental conditions and maintaining their effective activity are important [6,11]. This technique is attractive in the technical foods industries and is promoting due to growing numbers of opportunities, so greater demands are being made on nutrient capsules in a wide field. The origin of compounds used in a novel technology should be verified. Therefore, all coating materials should be investigated whether they are halal or originated from halal source [11].

Consideration is given to the nature of the compounds used for wall and core can be halal-based and non-halal-based. When it comes to the extraction of compounds from different sources, it is important to consider their origins from the Islamic point of view.

Different materials can be used at this technology including hydrocolloids, proteins, lipids, and carbohydrates. Among them, some agents are suspicious in term of halal origin. Therefore, they should be investigated when are used for coatings purposes. Encapsulation has been used in various studies. Santos et al. [28] investigated microencapsulation of vitamin $D_3$ using carboxymethyl tara gum and gelatin and showed results stability and prolonged release during digestion. Nguyen Le et al. [29] surveyed complex coacervates of hydrolyzed karaya gum with gelatin to protect soybean oil and curcumin which had effective outcomes. Also, Belščak-Cvitanović et al. 2015 studied to improved caffeine controlled release formulation in alginate hydrogel beads with pectin, carrageenan, chitosan and psyllium. The results showed that high concentrations of sodium alginate for hydrogel production in combination with pistachio or chitosan coatings showed the best carrier systems for inactivating caffeine [30]. These illustrate the use of hydrocolloids in this technique. So the coating material is approved by Malaysian Halal Food Standard as regional and global Halal standards [9].

## 4. Common halal encapsulating agents

Among all the shell materials, proteins and polysaccharides are mostly used (Table 1) [31].

Table 1- Different polymeric materials used in encapsulation techniques

| Carbohydrates | Proteins | Fats and Wax | Hydrocolloids |
|---|---|---|---|
| Chitosan | Gelatin | Lecithin | Carrageenan |
| Dextran | Whey protein | Liposome | Alginate |
| Cellulose | Soy protein | Wax | Arabic gum |
| Modified starch | Pea protein | Glyceride | Aar |

### 4.1. Proteins

Today, food hydrocolloids especially food proteins are widely used for protecting sensitive cores in capsules. They have different chemical functional such as ability to self-associate, amphiphilic properties and flexibility of molecular chain so that these proteins have excellent functional properties [32]. Solubility, viscosity, gelling and film forming properties are some of them and would be useful to use in encapsulation. The most commonly protein used for encapsulation is gelatin (Figure 3). It has good film forming and coating properties and is effective as well as other ideal chemical and physicochemical properties in the encapsulation process. Proteins and polysaccharides are among the essential pragmatic compounds in the food industry that can be used as complexes in encapsulation. A variety of proteins can be a good alternative to expensive biopolymers and provide desirable properties. External factors such as temperature, pH and pressure also affect the formation and stability of capsules. Also, the most important effective bonds are electrostatic bonds [5].

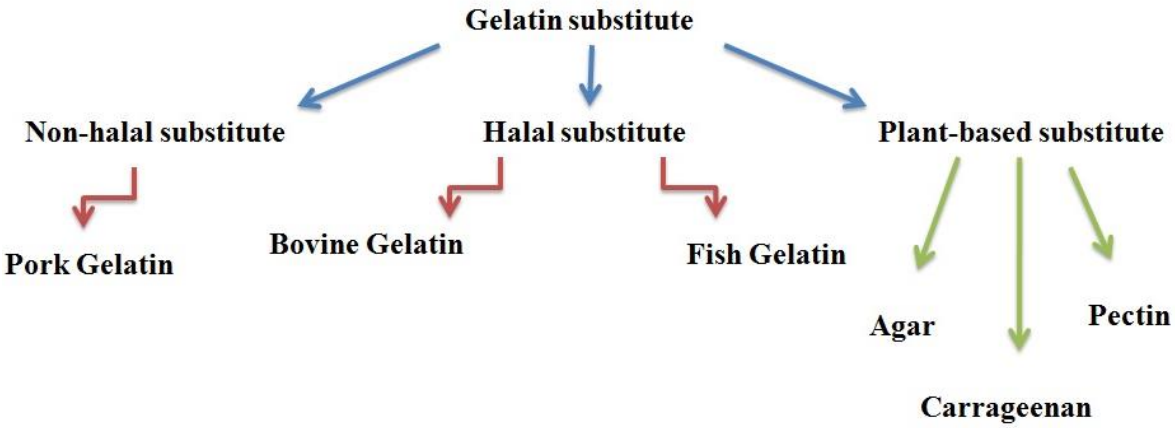


Figure 3- Different sources of gelatin

Protein-based materials such as gelatin are able to form stable coating with sensitive bioactive compounds [33]. Gelatin is derived from collagen and obtained by destruction of cross-linkages between polypeptide chains of collagen. It is used in different methods of encapsulation [34] and the special hydrocolloid widely are used in the food, drug, and cosmetic industries. Molecular gelatin is mainly composed of multiple repeated structure of Gly-Pro-Hyp. Gelatin has molecular weight of 30 kDa and more [35] and it is able to form walls when a mixture of volatile compounds is used. These gelatin capsules can be used to protect a wide range of core materials such as sensitive bioactive, nutraceutical and seasonings for use in foods and other applications. In research for protecting sensitive compound, microcapsules have been made from coacervates of two hydrocolloids, gelatin and CMC. Furthermore, addition of gelatin 1% to the mixture of maltodextrin and Arabic gum increased the retention of ethyl butyrate during spray drying and developed a

controlled release during time. So gelatin causes to promote the making crust on the surface of the droplet [36]. In other researches gelatin and carrageenan are able to form thermo-revisable hydrogels through some conformational transitions. Furthermore, it has been used for encapsulation of drugs in pharmaceutical industry for many years [37].

#### 4.1.1. Gelatin resources

Gelatin is made from the collagen as animal protein. One of the major sources of gelatin production in the world is pig. This causes restrictions on the consumption of its products for Muslims**.** If the gelatin is derived from the slaughtered halal-meat animals (such as cows or poultry), it is halal. Also, the compounds are derived from animals on the condition of Islamic slaughter are halal. If they are extracted from animals on the condition of non-Islamic slaughter are subject to doubt and if they are derived from the taboo-meat animals are taboo [5]. Almost 50% of the world's gelatin production is obtained from pigskin and rest of them comes from animals that have not been lawful and religiously slaughtered. Therefore, an important disadvantage of gelatin is the challenges with regard to its kosher and halal status. It should be noted that fish gelatin has a small share among animal sources and also, plant resources have a small share but in addition to research work, they can be widely used in industry in the future (Table 2) [38].

Table 2- Main sources of gelatin production in the world

| Source | Amount |
|---|---|
| Cow and sheep skin | 28% |
| Bone | 27% |
| Pork skin | 44% |
| Others | 1% |

#### 4.1.2. Non-halal resource

The origin of chosen gelatin depends on the geographical location and most common are bovine, beef, fowl, porcine and fish skin. It is noted that pork is over used while this is non-halal or non-kosher, but gelatin made from fish, beef or cow can be halal or kosher [39,40]. Pork gelatin is a protein product produced by partial hydrolysis of pork skins rich of collagen. The gelatin molecule is made up of amino acids joined together by amide linkages in a long molecular chain. These amino acids perform an imperative function in the building of connective tissue in humans which shows its structural similarity with many other proteins [41].

#### 4.1.3. Halal resource

Halal and Kosher gelatin is usually made from fish as there are fewer concerns compared to beef. Fish gelatin has now been identified as a real alternative to mammalian gelatin [39].

##### 4.1.3.1. Fish gelatin

The traditional sources of gelatin such as pork gelatin cannot be used for religions as Judaism and Islam. All fish are acceptable in Islam while fish without scales are acceptable in Judaism with minimal restrictions. Recent studies have shown that fish skin can be an alternative source for gelatin [38]. Fish skin is a raw material and might be an alternative for non-halal gelatin production because it provides a relatively suitable quality gelatin and also can be useful for religious people [35,42]. Some fish gelatin is less characterized than pork and is commercial. The most important gelatin properties in food industry are gel strength, viscosity and melting point. Fish gelatin has a much lower melting temperature than beef or pork gelatin and is more consumable as a religious product like halal and kosher because it is a common ingredient in kosher and halal food products. Fish gelatin is a protein product produced by partial hydrolysis of fish skin rich of collagen. The gelatin molecule is made up of amino acids joined together by amide linkages in a long molecular chain. The waste produced by fish filleting can account for most than 70% of the total weight of catches, and processing causes yielding gelatin can help to avoid harmful environmental effects. Gel strength and heat stability are two quality factors of a fish gelatin is determined [37,43]. Recent studies showed that

certain fish gelatin might have similar quality characteristics compared to mammalian gelatin. It can depend on the processing conditions and the species fish gelatin. Therefore, more attention can be paid to its replacement with pork gelatin [37,38,44]. Physicochemical characterization of fish gelatin as alternative gelatin source has been studied by Ratnasari, 2016. The results showed that the gel strength, viscosity, gelling temperature, melting temperature were higher than some commercial gelatin. Therefore, it can be used as a new alternative to gelatin for stabilization, thickening and gel-forming [45].

#### 4.1.3.2. Bovine gelatin

Hydrolyzed bovine gelatin is a protein product produced by partial hydrolysis of collagen, a protein material, extracted from animal tissue such as skin and bone. Bovine gelatin has similar structure like fish and pork origin. Except porcine, bovine skin gelatin is widely utilized in food manufacturing because the sources are more available. Gelatin from bovine skin produced from alkaline treatment that named type B gelatin. It may be chosen by food manufacturer in order to different characteristics [35]. Bovine gelatin has been processed in accordance with religious lawful can be acceptable.

### 4.2. Plant sources for gelatin replacement

#### 4.2.1. Agar

Agar is a gel-forming polysaccharide extracted from red algae. It is obtained from the cell walls and is a gelling agent with unbranched chains. Agar can be used as gelatin substitute; a thickener for some foods and a clarifying agent [46]. It has a strong gel with low elasticity. The polysaccharides are usually linear polymers and agar contains mainly agarose but also agaro-pectin. It solubilizes at around boiling water temperature and decreasing the temperature causes to form a suitable gel. In a survey, properties of gels made by agar and fish gelatin by Somboon et al. 2014 have been studied. It demonstrated agar and fish gelatin are acceptable for gelling agents in halal food [47]. Also, extraction of Agar from seaweeds in Red Sea Region was done by Abraham et al. 2018. Conduction of several works for extraction and purification of agar from different sources has indicated its importance as safe and lawful resource for the people concerning about halal status [48].

#### 4.2.2. Pectin

Pectin is a structural hetero-polysaccharide and was first isolated and described in 1825. It is produced as a white to light brown powder and often considered the primary vegan gelatin alternative and sometimes used as a gelatin alternative. Pectin comes from fruits like apples and oranges and also acceptable for both Islam and Judaism [39,49]. Pectin has some characteristics such as form gels at a relatively high temperature, can remain solid at room temperature and can be bitter. High methoxy pectin requires sugar and an acid to set properly and the sugar is often used to help hiding its bitterness. When gelatin sets, it will have a soft and unsteady jelly consistency; pectin will be denser in addition to being more gummy and moist than gelatin. Pectin is a water-soluble fiber came from the cellular walls of fruit and the sources have high concentration of that are grapes, apples, cherries and citrus fruits. There are some powder or liquid types that known as commercial pectin. Eating products made with pectin may offer some health benefits, but more scientific studies is needed [42]. A comparative study on effect of pectin and other polysaccharides as replacement with animal gelatin in textural properties of non-fat yogurt was done. These researches draw more attention to alternative sources and their use in halal food products [50]. So it is successfully extracted valuable pectin from plant wastes, seaweed and fruits. Pectin can act as a protective layer of food ingredients and can be used for encapsulation of bioactive compounds and the materials sensitive to the harsh environmental conditions. The polymer is used alone or in combination with other biopolymers to achieve the required results [51-53].

#### 4.2.3. Carrageenan

The sources of carrageenan are algae or seaweed, and the important application is thickening agent and comes from red alga. It is a common ingre-

dient in many foods, including milk products [46]. Rising demand for non-mammalian gelatin for halal food markets has increased interest in gelatin alternatives from plant sources. Hence, the value of such biopolymers as a gelatin substitute as a gelling and stabilizing agent and a cheap source of gelatin substitution is considered. Carrageenan can provide some of the gelatinizing properties and can be used to replace it, either in its use in the complexes of both or in its application in encapsulation as a wall material. In study of Derkachin et al. 2014, the rheological and gel properties of gelatin containing carrageenan as an anionic polysaccharide (a wide range of low polysaccharide concentrations) has been studied. Since in the appropriate ratio of these two substances (gelatin and carrageenan), the performance of gels and their effective viscosity at specific yield stresses (rheological properties) are greatly increased.

The outcomes showed that the combined use of two biopolymers increases the strength of the gel. In this case, the gels yield stress and non-Newtonian flow are generated at higher shear stresses [54]. A study is done about extraction process of plant-based gelatin replacer by Jaswir et al which indicated increasing demand for plant gelatin for halal and kosher food markets [46]. In another study, the combined structure of gelatin and carrageenan was investigated for targeted release of bioactive compounds in the intestine. Significant electrostatic interactions between gelatin and *i*-carrageenan in aqueous solution, at pHs lower than the isoelectric point of the protein, resulted in the formation of a polyelectrolyte complex by complex coacervation, resulting in a more stable gelatin structure and greater control over compound release [37].

## 5. Conclusion

Numerous developments have been made in the field of food production. One of the novel technology is encapsulation which provides the specialized needs of the food and pharmaceutical market. In term of halal, the most important aspect is investigation the extraction origin of the hydrocolloids used as wall materials. Halal status of the compounds used at this technology should be investigated because of food consumers are from different religions, especially Islam and Judaism. Gelatin is one of the compounds whose halal status is important because the major source of its extract is from non-halal animals in western countries. So nowadays it has been tried to replace it with plant-based and halal animal sources. Various methods such as extrusion, emulsion, coacervation, etc. are used to cover bioactive compounds with gelatin, which before using gelatin in them, it is better to be examined by halal approval organizations. However, because of the population growth consuming halal foods in the world and the lack of sources producing halal gelatin, use of similar alternatives in technical points has been developed.

## 6. Conflict of interest

The authors declare no conflict of interest.

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