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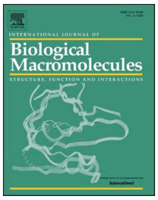

Review

Enhanced properties of non-starch polysaccharide and protein hydrocolloids through plasma treatment: A review

Shahriyar Sahraeian [a], Ali Rashidinejad [b,*], Mehrdad Niakousari [a]

[a] Department of Food Science and Technology, College of Agriculture, Shiraz University, Shiraz, Iran
[b] Riddet Institute, Massey University, Private Bag 11222, Palmerston North 4442, New Zealand

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ABSTRACT

Hydrocolloids are important ingredients in food formulations and their modification can lead to novel ingredients with unique functionalities beyond their nutritional value. Cold plasma is a promising technology for the modification of food biopolymers due to its non-toxic and eco-friendly nature. This review discusses the recent published studies on the effects of cold plasma treatment on non-starch hydrocolloids and their derivatives. It covers the common phenomena that occur during plasma treatment, including ionization, etching effect, surface modification, and ashing effect, and how they contribute to various changes in food biopolymers. The effects of plasma treatment on important properties such as color, crystallinity, chemical structure, rheological behavior, and thermal properties of non-starch hydrocolloids and their derivatives are also discussed. In addition, this review highlights the potential of cold plasma treatment to enhance the functionality of food biopolymers and improve the quality of food products. The mechanisms underlying the effects of plasma treatment on food biopolymers, which can be useful for future research in this area, are also discussed. Overall, this review paper presents a comprehensive overview of the current knowledge in the field of cold plasma treatment of non-starch hydrocolloids and their derivatives and highlights the areas that require further investigation.

1. Introduction

Hydrocolloids are widely used in the food industry due to their ability to provide various functionalities such as viscosity, gelation, emulsification, and stabilization of dispersions [1,2]. They are also used to develop functional foods that are high in fiber, low in fat, and provide various health benefits to consumers. However, to achieve specific functionalities, hydrocolloids may require modification [3–7]. For example, modification of hydrocolloids can improve their processing properties, enhance thickening, facilitate emulsion preparation, increase shelf life, and create new materials for food packaging [8–12]. On the one hand, starch is known as a copious polysaccharide used in food formulation as an emulsifier, thickener, and binder [13]. However, the application of this hydrocolloid is limited due to its solubility and instability under harsh pH conditions and shear rates. Therefore, we use the phrase "non-starch hydrocolloids" to introduce other types of hydrocolloids other than starch such as gum, mucilage, non-starch polysaccharides, and some proteins such as gelatin, collagen, whey, and their derivatives [14,15]. The reason why various methods are exploited to modify hydrocolloids may arise from their relatively low cost, minimum undesirable changes, controllability, and rapid result [16,17].

One of the emerging technologies for the modification of hydrocolloids is plasma treatment. Plasma is known as the fourth state of matter, which consists of a mixture of active species, including ions, free radicals, excited and ground-state atoms, and neutral molecules [18]. Plasma can be generated by applying thermal energy, electric field energy, radiation, or beams such as lasers, UV photons, electrons, and protons. Plasma can be categorized into two subcategories: thermal plasma and non-thermal plasma. Thermal plasma operates at high power and high temperatures, leading to the decomposition of chemical compounds. In contrast, non-thermal or cold plasma (cold plasma) operates at relatively low gas temperatures and is initiated by the high temperature of free electrons (Te $\geq$ 104 K). This type of plasma generates hot electrons, energetic ions, low-temperature excited species, free atoms, and radicals in the active zone [19]. Cold plasma can be further divided into two types: dielectric barrier discharge plasma and jet plasma, which are used for various purposes in different industries [20]. There are several novel non-thermal approaches to modify

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* Corresponding author.
E-mail address: A.Rashidinejad@massey.ac.nz (A. Rashidinejad).

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hydrocolloids such as plasma treatment, ultrasonication, homogenization, ozone treatment, and gamma and UV radiation [21–25].

This review aims to present a comprehensive overview of the recent advances in the plasma treatment of hydrocolloids and proteins, with a focus on non-starch hydrocolloids and their derivatives. The main objective of this review is to highlight the structural and physicochemical changes induced by plasma treatment, such as color, crystallinity, chemical structure, rheological behavior, and thermal properties. This review will also discuss the common phenomena that occur during plasma treatment, including ionization, etching effect, surface modification, and ashing effect. Finally, this review will present and discuss the results published recently on the influence of plasma treatment on non-starch hydrocolloids. Since the sensory properties of plasma-treated dairy products are overlooked, and to reveal the industrial potential of cold plasma processing, a section is also dedicated to discussing how plasma treatment enhances various sensory properties of dairy products. In the following sections, we will provide an in-depth discussion of the cold plasma generators, the characteristics of plasma, and the mechanisms involved in the plasma treatment of the abovementioned hydrocolloids. Chemical and physical changes resulting from plasma treatment of non-starch hydrocolloids and their derivatives include the majority of discussions in this paper. Several publications provide extensive information about plasma and plasma generation systems [20,26,27], and this review aims to summarize and synthesize the most recent findings in this field.

2. Cold plasma: instruments and environment chemistry

2.1. Plasma generation instruments

Plasma is the fourth state of matter that is created by applying additional energy to gases, resulting in the formation of an environment that consists of positive and negative ions, excited species, radicals, free electrons, and photons. This environment is electrically neutral and is known as a quasi-neutral environment. Plasma is widely found in nature, such as in the ionosphere, flames, and the sun, and can emit additional energy in the form of radiation, such as ultraviolet light.

Cold or non-thermal plasma is generated when the kinetic energy of electrons is higher than other species, resulting in a low-temperature plasma environment suitable for the treatment of thermosensitive materials. Plasma generators are used to create plasma and can use various high-energy sources, including radioactive (gamma radiation), thermal, optical (UV light), electrical, and electromagnetic radiation. The two most common sources used for cold plasma generation in food processing applications are electrical and electromagnetic radiation [28].

Several types of cold plasma generators are used, including radiofrequency discharge, glow discharge, dielectric barrier discharge (DBD), and corona discharge. Radiofrequency discharge systems generate plasma by applying electrical pulses to an electrically conductive coil [20]. The dielectric barrier discharge (DBD) system uses a highly electrically resistant material between two electrodes connected to a high-voltage source to generate plasma. The corona discharge system applies gases at atmospheric pressure to the sharp edges of one or two electrical conductors with a high electric field, and a sharp spark then appears. However, continuous corona discharge systems have the limitation of spark generation, which can be eliminated by applying periodic-pulsed currents. DBD systems use high-power pulses, which alter >10 Hz [29]. Glow discharge systems require direct voltages of over 100 V and apply plasma in larger volumes, resulting in uniform discharge at lower temperatures compared to other plasma generators [30]. In summary, cold plasma is generated by applying additional energy to gases and is suitable for the treatment of thermosensitive materials. Plasma generators can use various high-energy sources, and several types of cold plasma generators are used in food processing applications, including radiofrequency discharge, glow discharge, dielectric barrier discharge, and corona discharge systems. For more information about plasma generator systems, please refer to the following references: [19, 31–33].

2.2. Pure oxygen or air plasma environment chemistry

Plasma treatment involves the generation of a plasma environment where ionization of atoms and electrons occurs, resulting in ionization as the primary reaction observed. Cations and hot electrons are responsible for subsequent reactions through the plasma treatment. The presence of electronegative gases such as oxygen, sulfur hexafluoride, chlorine, and uranium hexafluoride in a plasma environment produces anions due to their high electron affinity [34].

During plasma treatment, the production of ozone, radicals, and emission of UV radiation should not be underestimated. Ozone is one of the most common reactive species generated during plasma treatment, which is formed through the electron bombardment of oxygen. The gross reaction of ozone synthesis is typically represented as $[3O_2 + e^- \rightarrow 2O_3 + e^{-\,-}]$ [35]. Other reactive species such as atomic oxygen, singlet oxygen, hydroxyl radicals, and superoxide are also generated as primary species, while hydrogen peroxide, peroxynitrite, nitric oxide, nitrates, and nitrite ions are formed as secondary species in plasma-activated water [36,37]. The presence of water in the plasma treatment environment can result in the production of reactive oxygen (ROS) and reactive nitrogen (in the case of the presence of nitrogen) species (RNS). These species can be utilized for surface modification of biopolymer films to functionalize and activate polymer surfaces. For example, plasma treatment of polyurethane and steel with He/N2 and He/Ethylene, respectively, has shown good adhesive properties as a surface modification [38].

Plasma etching is another process that can occur during the plasma treatment of a polymer. Excited species produced during plasma treatment can react with the surface polymer, resulting in superficial fragmentation and/or small molecules on top of the polymer surface [39]. Fig. 1 illustrates the scheme of etching during the plasma treatment of a typical film. Furthermore, plasma treatment can induce an “ashing effect” where singlet oxygen produced in a plasma environment can oxidize and transform organic compounds such as proteins into CO_2. Disinfection and reduction in microbial load are common applications of cold plasma treatment in food processing [40–43]. Overall, plasma treatment offers a versatile and promising approach for modifying material surfaces with various applications, including food processing and biomedical engineering.

In addition to air and oxygen as working gases, other gases such as nitrogen, argon, and gaseous hydrocarbons can be used in plasma generation systems. In order to use input gases other than ambient air, the gas is introduced into a vacuum chamber, where it is ionized through an electric field. Expectedly, applying pure gases such as nitrogen, argon, or helium results in their specific ions and reactive species. For example, it is reported that thirteen reactions including N^+, N, and N_2 radicals as main RNS are expected in the plasma environment of pure nitrogen. It is also reported that pure nitrogen results in a more uniform reaction condition [44]. This can be due to the thermal instability caused by the presence of oxygen in the plasma environment [32]. On the other hand, methane, hydrogen, and carbon radicals, as well as methane ions, are generated in the plasma environment when methane is the working gas [45].

3. Non-starch hydrocolloids and plasma treatment

3.1. Non-starch hydrocolloids: definition and types

As mentioned earlier, non-starch hydrocolloids are assigned to those hydrocolloids which are not starch or derived from starch and include non-starch polysaccharides and proteins. Historically, such categorization stems from the extraction and isolation method used to obtain hydrocolloids. For example, the remainder of the alkali extraction of cell

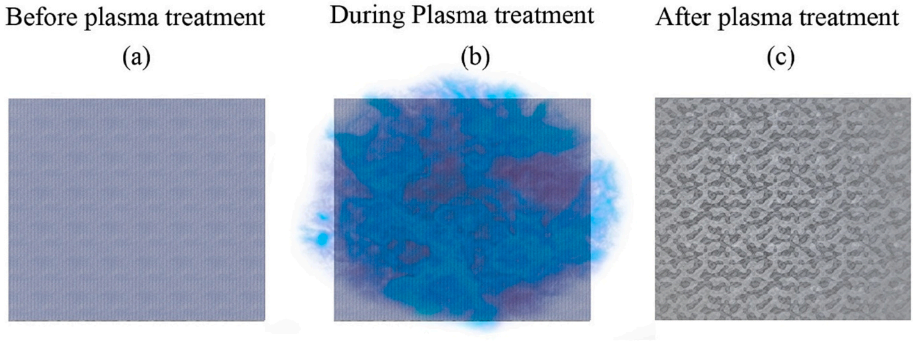

Fig. 1. Visualization of plasma treatment and etching effect on a surface of a uniform film; (a) prior to exposure to cold plasma, (b) during the plasma treatment, and (c) the plasma-treated film surface.

wall materials was named cellulose and the solubilized portion is called hemicellulose [46]. Based on Choct (1997) [47] and Bailey (1973) [48], non-starch hydrocolloids can be categorized as cellulose and non-cellulosic polymers (Fig. 2). It is reported that wheat, rye, barley, oat, rice, and sorghum are found to possess arabinoxylans. These polysaccharides are in small amounts in comparison with other polysaccharides. However, they constitute a predominant portion of cell walls that surround starch in the endosperm and the aleurone layer [49]. In the above-mentioned cereals, these polysaccharides cannot be solubilized in water due to a strong linkage to the cell wall. On the other hand, the portion of arabinoxylans which can be solubilized in water can absorb a large amount of water (1:10 arabinoxylan to water) and form a viscose solution. Structurally, β-glucans are glucose units with β(1→4) and β(1→3) bonds which make them similar to cellulose. However, the presence of β(1→3) bonds results in a looser structure in comparison with cellulose and this in turn can bring about amorph, soluble, and flexible matrice [50,51]. Therefore, it can be stated that the application of β-glucans can be extensive. For example, these hydrocolloids are found to be responsible for the reduction of cholesterol and glucose level. Furthermore, these macromolecules can form gels in certain conditions and be exploited as thickening agents in various food formulations [52,53]. D-Galacturonic is the main sugar unit of pectin molecules' backbones which are connected by α(1→4) bonds. Arabinans, arabinogalactans, and galactans together are considered pectic materials. The former consists of (1→4) α-L-arabinose unites and the latter is formed by β(1→4) galactan backbone. Arabinogalactans also consist of the same units, while they possess 3- or 5-bonded arabinose units. These molecules are classified as type I and II. Type I arabinogalactans are copious in grain legumes [54]. Finally, it can be stated that the predominant portion of non-starch hydrocolloids is as discussed above, and now, it is time to define gum and mucilages. When special plants are exposed to disintegration, a gummy substance is oozed which is called the "gummosis" process. Gums are members of two main families including *Leguminosae* and *Sterculiaceae* and other families such as

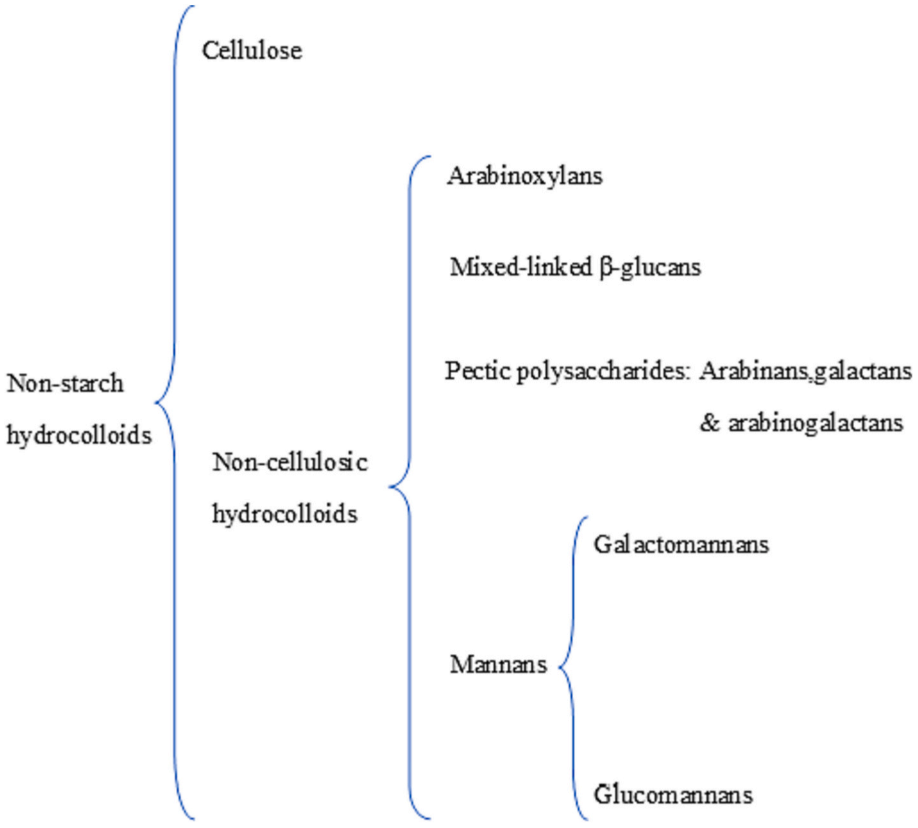

Fig. 2. Classification of non-starch food hydrocolloids.

Anacardiaceae, *Combretaceae*, *Meliaceae*, *Rosaceae*, and *Rutaceae* are capable of gum production. These hydrocolloids can be obtained from plants, animals, microbial and marine sources [55] and can be classified based on their charge, source, chemical structure, and shape. For example, based on their charge, they are divided into anionic, cationic, nonionic, amphoteric, and hydrophobic. Former include alginic acid, pectin, xanthan gum, hyaluronic acid, chondroitin sulfate, gum Arabic, gum karaya, gum tragacanth which are natural and Carboxymethyl chitin and cellulose which are modifed anionic gums [56]. Chitosan, on the other hand, is considered cationic natural hydrocolloid and cationic guar gum and hydroxyethylcellulose are modified cationic gums [57]. Regarding nonionic hydrocolloids, starch, dextrins, guar gum are natural and cellulose ethers such as hydroxyethyl cellulose, methylcellulose, and nitrocellulose are modified ones [56]. Furthermore, carboxymethylchitosan, N-hydroxyl-dicarboxyethylchitosan, modified potato starch are included as amphoteric gums and cetylhydroxyethylcellulose, polyquaternium are hydrophobic hydrocolloids [58]. In another classification which is based on the monomeric units, gums are regarded as homoglycans (amylose, arabinans, cellulose), diheteroglycans (algins, carrageenans, galactomannans), tri-heteroglycans (arabinoxylans, gellan, xanthan), tetra-heteroglycans (gum Arabic, psyllium seed gum), and penta-heteroglycans (ghatti gum, tragacanth). Further classifications are depicted in Fig. 3 [59].

4. Plasma treatment of non-starch hydrocolloids

4.1. Overview

It is known that proteins are sensitive to extrinsic undesirable factors such as harsh pH conditions, ions, UV, and oxidation and almost all these factors exist simultaneously in the plasma treatment environment. However, scientists believe that ROS and RNS are responsible for the final conformational changes in proteins. These species are capable of breaking down peptide bonds and other covalent linkages, triggering oxidation, and affecting the conformation of proteins. On the other hand, some other intramolecular or intermolecular linkages may be formed during the plasma treatment of proteins which causes aggregation and polymerization as well as the unfolding of protein which is said to be progressive [60,61]. It is significant to monitor the changes in the secondary structure of proteins because most of the functionality of

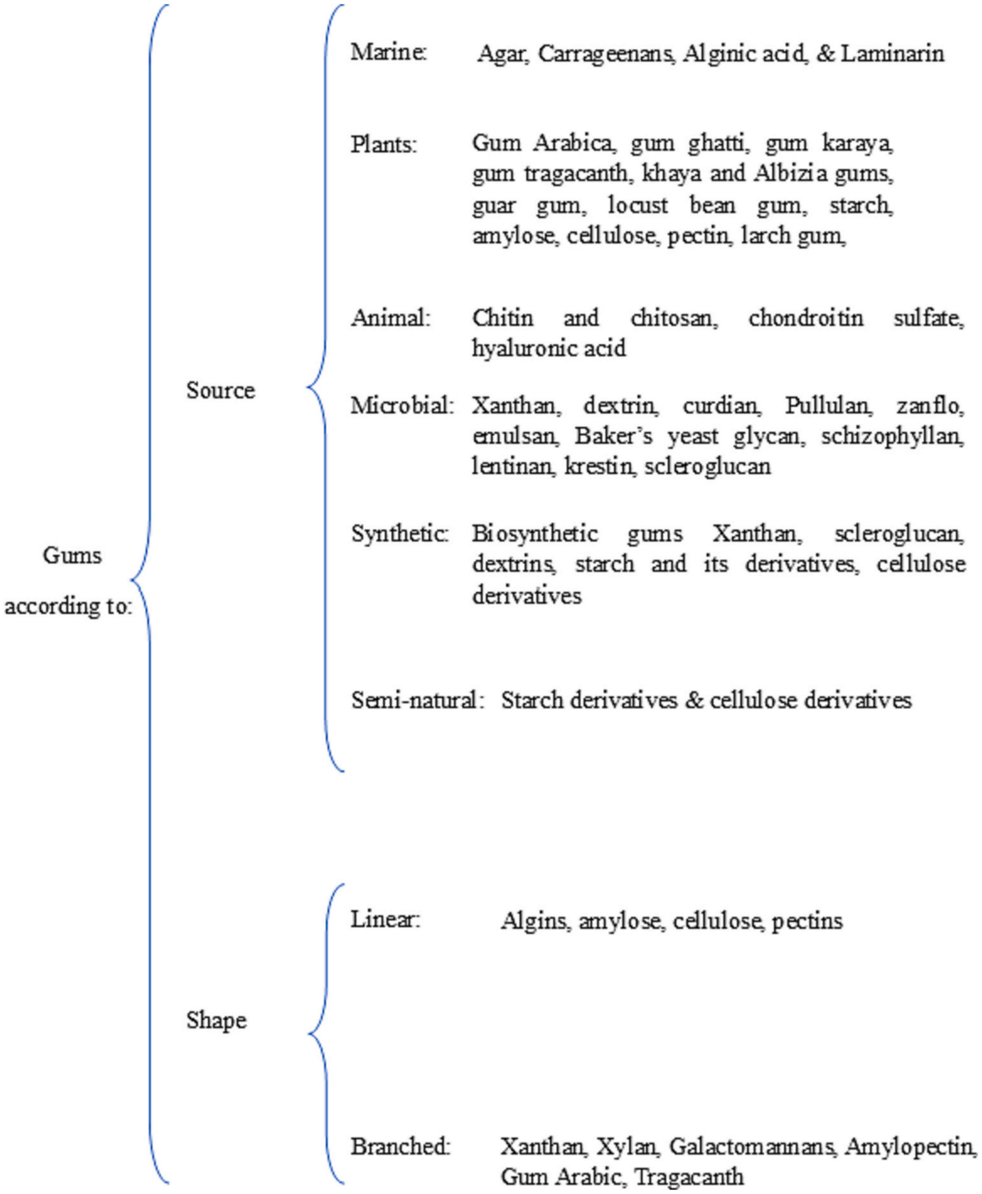

Fig. 3. Different classifications of various gums.

proteins arises from the secondary and tertiary structures. For example, it is reported that β-sheets decreased and α-helices and β-turns increased after plasma treatment of wheat proteins [7]. In addition, increasing the thiol groups was observed in the case of plasma treatment of zein powder [62]. Dynamic light scattering (DLS) and high-performance liquid chromatography (HPLC) can help scientist to monitor the unfolding changes in proteins [63]. A significant unfolding was observed when whey proteins were subjected to the plasma treatment and no significant changes in α-helices were observed in the case of sodium caseinate films Zein film: Effects of dielectric barrier discharge atmospheric cold plasma [64].

On the other hand, since the nature of non-starch polysaccharides differs from proteins, plasma treatment of such macromolecules causes different results, and the mechanism of reactions is different. Scientists have exploited the surface-effective attribute of plasma treatment to produce hydrocolloid-based pollutant absorbents. In this technique, plasma treatment causes surface functionalization through oxidation, amination, sulfuration, and coating [65–67]. Other hydrocolloids such as chitosan have been subjected to the experiment more than cellulose. For example, chitosan was gelled in situ using plasma treatment which was called “liquid phase gelation”. Breakage of β(1→4) bonds due to the oxidation and simultaneously, formation of aldehyde groups caused crosslinked structures with amine groups of this polysaccharide [68]. Generally, many polysaccharides were reported to be more soluble in water and other solvents such as ammonium hydroxide after plasma treatment [69,70]. This may stem from free radical reactions and exposure of more hydrophilic groups to polysaccharides. More thermal stability of hydrocolloids is also reported which is rationalized by the formation of more stable intermediates which possess lower water content during heating [71]. Fig. 4 illustrates an overview of frequent changes in non-starch hydrocolloids and their derivatives during plasma treatment. Most of the physical and chemical changes during the plasma treatment of non-starch hydrocolloids will be discussed in the following sections in detail.

4.2. Health-beneficial effect

As mentioned earlier, plasma treatment of hydrocolloids revealed to have several advantages over conventional methods. Modification of such macromolecules through plasma treatment has been shown to be applicable for many purposes. For instance, many polysaccharides revealed higher antioxidant activity after plasma treatment [72–74]. In these cases, polysaccharides had intrinsic antioxidant activity, however, degradation and lower molecular weight after plasma treatment resulted in more movement and activity of such molecules. Fan et al. (2020) [75] studied the effect of plasma treatment on the immune activity of the polysaccharide from *Dendrobium nobile Lindl*. In addition to the higher water solubility and cross-linked structure of this polysaccharide, an enhancement in the phagocytosis ability of RAW264.7 and secretion of cytokines TNF-α, IL-6, and IL-1 were observed. They deduced that the higher solubility of this polysaccharide affected its immune activity.

4.3. Technological benefits

A considerable number of hydrocolloids are capable to stabilize the emulsion due to the possession of both hydrophilic and hydrophobic units in their structures (chitosan) or the presence of proteins in their structure (gum Arabic, pectin) [76]. For example, it is reported that nitrogen gas plasma treatment of chitosan resulted in a more structured network which led to a strong layer surrounding oil droplets and a more stable emulsion [77]. Misra et al. (2018) [78] reported that lower interfacial tension and higher viscosity of plasma-treated xanthan gum led to a more stable emulsion in comparison with native gum. Moreover, scientists tend to exploit plasma-modified hydrocolloids as packaging material, because plasma treatment has been proven to be an enhancement method to produce strong and suitable composite films. Plasma treatment of lily polysaccharide and alginate to produce composite films was done by Cui et al. (2022) [79]. They observed a more compact network due to more interactions between the abovementioned polysaccharides. More tensile strength, thermal stability, and higher antioxidant and antimicrobial activities were the final results of plasma-treated hydrocolloids. In another study, Chen et al. (2020) [4] applied the plasma treatment to chitosan-based complex coacervations which resulted in a higher encapsulation efficiency for resveratrol (82 %). They stated that more interaction between chitosan and zein led to a stronger complex and therefore, a more sustainable encapsulation system. Thus, it is not unexpected if scientists have a great tendency to apply the plasma treatment to different hydrocolloids to modify them for the food,

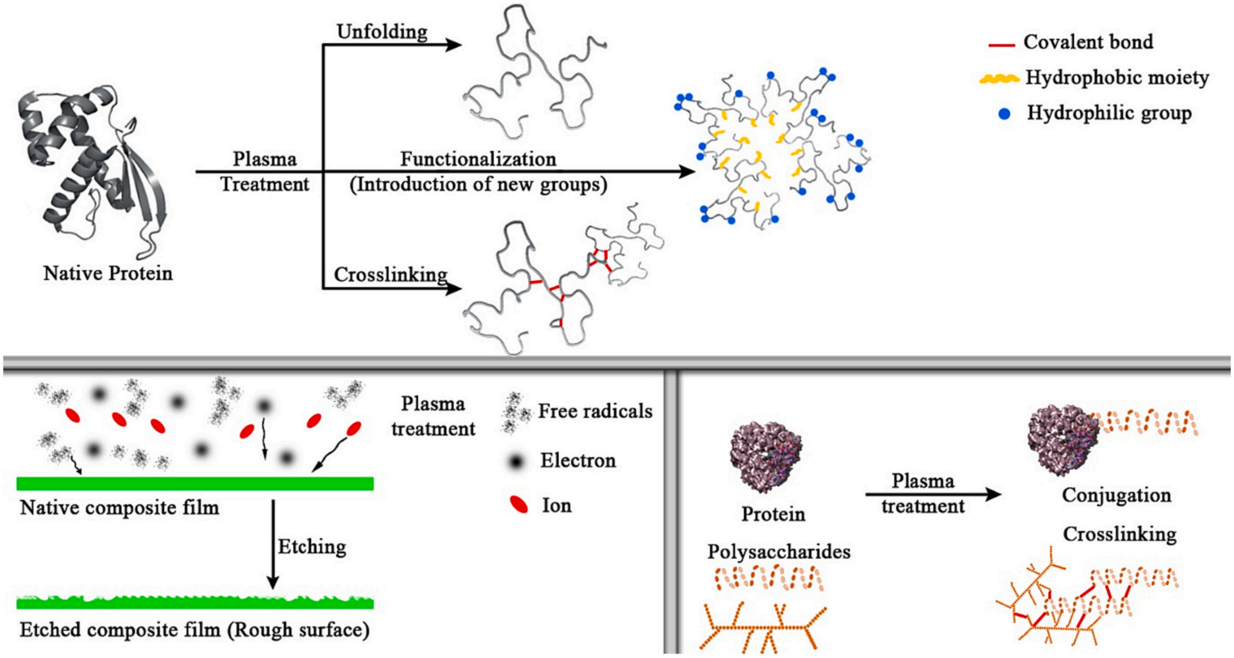

Fig. 4. An overview of frequent changes in non-starch hydrocolloids and their derivatives during plasma treatment.

pharmaceutical, and cosmetic industries. Further investigations on the application of plasma-treated non-starch hydrocolloids are summarized in Table 1 [3,62,64,77,80–91].

5. Chemical and physical changes in non-starch hydrocolloids

5.1. Chemical changes

The presence of various reactive species in the plasma environment can cause different chemical interactions with the samples. By reviewing previous studies and evaluating the factors mentioned earlier, it is possible to predict the chemical changes in plasma-treated hydrocolloids and their derivatives. For instance, Jampala et al. (2005) [6] studied the plasma modification of xanthan gum (XG) and reported the reaction of primary amine groups with ethylenediamine (ED) resulting in the grafting of ED in XG. Fourier-transform infrared spectroscopy (FTIR) analysis and labeling of the treated samples with fluorescamine confirmed this interaction. However, in some cases, there are no observable changes after plasma treatment. This might be due to the reaction time and intensity, the accessibility of reactive species to the interior parts of samples, etc. For example, Misra et al. (2018) [78] found that the FTIR results of control and treated XG powders were similar, and the main backbone chains remained intact.

Although changes in the structure of samples are possible, they are not always evaluated in studies. Bulbul et al. (2019) [92] performed an ATR-IR analysis of treated XG and reported that no changes occurred in the main polysaccharide chains, although some peaks showed a slight difference in intensity. Amirabadi et al. (2020) [82] conducted an FTIR analysis of hydrophobically modified gum Arabic (GA) and found that plasma treatment of GA resulted in the formation of C═O, C—O, and C—H groups, and oxidation reactions. The reduction in uronic acid content was also observed from the decreased carboxylate anion corresponding peak (1600 cmˆ-1). These observations suggest that the addition of oxygen-containing molecules through plasma treatment of gum Arabic and other hydrocolloids is predictable. Pankaj et al. (2017) [88] prepared a modified chitosan film treated using cold plasma and found an increment of oxygen-containing peaks in the FT-IR analysis. This observation was further confirmed by X-ray photoelectron spectroscopy (XPS) analysis, which also revealed that increasing oxygen groups in the chitosan film did not affect amide bonds. The use of different feed gases can cause different results in plasma treatment due to the differences in the reactive species generated in the plasma environment. For instance, Padil et al. (2016) [93] prepared electrospun nanofibers based on kondagogu (KG), karaya (GK), and Arabic (GA) gums by mixing electrospinning solutions with polyvinyl alcohol (PVA) or polyethylene oxide (PEO) and compared the alteration in functionalities due to methane plasma treatment with untreated samples. The grafting of carboxyl (—COOH), carbonyl (—C═O), and hydroxylated (—OH) groups resulted in the development of new functional groups in plasma-treated samples.

Table 1
Recent investigations on the application of plasma-treated non-starch hydrocolloids (categorized by source).

Hydrocolloid	Improved functionality	Proposed application	Reference
Chitosan	Sustained release	Food packaging	[80]
Chitosan	Multiple functionalities	Food packaging	[82]
Chitosan	Mechanical Properties	Food packaging	[81]
Chitosan and zein	Multiple functionalities	Food packaging	[3]
Pectin	Multiple functionalities	Multiple purposes	[85]
Pectin	Multiple functionalities	Multiple purposes	[77]
Pectin	Rheological properties	Thickening agent	[83]
Pectin	Rheological properties	Thickening agent	[84]
Gum Arabic	Interfacial properties	Emulsions	[88]
Grass pea protein	Interfacial properties	Emulsions	[89]
Ovalbumin	Interfacial properties	Emulsions	[90]
Peanut protein	Multiple functionalities	Multiple purposes	[91]
Zein	Multiple functionalities	Multiple purposes	[62]
Zein	Multiple functionalities	Multiple purposes	[87]
Zein	Surface properties and cytocompatibility	Food packaging	[86]
Zein	Physical properties	Food packaging	[64]

Dalei et al. (2019) [94] prepared nitrogen and ammonia plasma-treated hydrogels based on carboxymethyl guar gum (CMGG) and polyvinyl alcohol (PVA) using tetraethyl orthosilicate as a cross-linker agent. They observed a relatively higher peak intensity of hydroxyl groups in the FTIR spectra of treated hydrogels. Although FTIR spectroscopy of untreated hydrogels was similar to treated ones, the peak at 1140 cmˆ-1, which was attributed to the semi-crystalline nature of PVA, was not elucidated in the FTIR spectroscopy of hydrogels because intermolecular mixing reduced the crystallinity. Similarly, Li et al. (2019) [95] reported that plasma modification of the chitosan film surface showed a considerably declined peak at differences between low-power (<600 W) and high-power (≥600 W) plasma treatment in the peak intensity of 1413 cmˆ-1, which is attributed to CH_2 bending vibrations [95].

Overall, plasma treatment can induce different chemical changes in hydrocolloids and their derivatives, depending on various factors such as the type of gas, plasma power, treatment time, and accessibility of reactive species to the interior parts of the samples. These changes can include the formation of new functional groups, such as carbonyl, carboxyl, and hydroxylated groups, and the modification of existing functional groups, such as primary amine groups. The addition of oxygen-containing molecules through plasma treatment of hydrocolloids is possible and predictable, which can be confirmed by various spectroscopic techniques such as FTIR, XPS, and attenuated total reflectance FTIR (ATR-FTIR). However, in some cases, no changes can be confirmed after plasma treatment due to the aforementioned factors. Therefore, a comprehensive understanding of the plasma treatment parameters is required to predict the chemical changes in hydrocolloids and their derivatives after plasma treatment.

5.2. Crystallographic change

It has been reported that reactive species generated in the plasma environment can have a substantial influence on the crystallinity of non-starch hydrocolloids. Many scientists believe that reactive species can penetrate crystalline and amorphous regions, react with the polymer and alter the crystalline structure of hydrocolloids [96–98]. Moreover, in some cases, researchers attribute the changes to the evaporation of water molecules from the matrix of such biopolymers [99,100]. In some studies, the condition of plasma treatment is such that reactive species attack the susceptible regions and break the linkages between polymer chains which leads to a decrease in crystallinity [97,101,102]. On the other hand, there are several reports which indicate that the plasma treatment can improve the crystallinity of hydrocolloids. For example, it is reported that the acidic molecules generated through plasma-treated water can degrade the amorphous regions and increase the crystalline portion in the hydrocolloid structure [103]. Furthermore, nitrogen gas-based plasma treatment is reported to lead to more linkages between molecular chains and a stronger network which increases the crystallinity of hydrocolloid-based edible films [104]. In order to study the structural changes and alterations in the crystalline state of hydrocolloid polymer matrices, scientists primarily tend to exploit X-Ray diffraction (XRD) graphs. Crystallography of native and treated gum Arabic is carried out by XRD patterns. Results showed that plasma treatment and duration of the process could not affect the amorphous nature of GA which may be a result of the surface-effective property of the plasma process. However, the intensity of peaks was higher in plasma-treated GA and became sharper with increasing treatment time. This means an increase in particle size during plasma treatment of GA [82]. This increase may be attributed to the ROS and RNS and the new cross-linked section in the GA structure. Similar results have been reported by Wu et al. (2018) [105] regarding the modification of banana starch treated with atmospheric pressure cold plasma. ROS and RNS generation, and subsequently cross-linking effects resulted in more crystallinity in treated granules samples.

Analysis of XRD patterns of chitosan films treated with glow discharge plasma by Li et al. (2019) [95] was carried out. Results showed that an increase in plasma power treatment results in an increase in crystallinity. This change may be attributed to ionized atoms generated through the plasma discharge and their subsequent reaction with surface groups of chitosan films. Subsequently, the bombardment of surface molecules may cause an increase in interaction between superficial groups which leads to a more packed structure. Similar trends were observed when XRD patterns of plasma modification of high methoxy pectin (HMP) were obtained by Momeni et al. (2018) [77]. The formation of new hydrogen bonds in hydrophilic segments of various sites of pectin was reported as a reason for an increment in the crystallinity. In another study conducted by Pankaj et al. (2015) [106], it is reported that an increase in the crystallinity of starch films treated with atmospheric air plasma may be attributed to changes in the orientation of the double helices in the crystalline regions.

However, it is not surprising if plasma treatment does not affect the crystallinity of some derivatives as shown by Dalei et al. (2019) [94]. They studied the effect of cold plasma treatment on carboxymethyl guar gum/PVA hydrogels and they found inconsistent results in crystallography patterns obtained from an X-ray diffractometer. Untreated samples showed lower crystallinity, which may be due to hydrogen bonding through hydrogel preparation, resulting in a disorder in parent polymers. However, the crystallinity of treated hydrogels showed insignificant changes. It can be concluded that plasma treatment might not affect the organization of polymer chains in the hydrogel [94]. Finally, it can be stated that cold plasma treatment of gums and their derivatives can bring about more crystalline regions due to more interaction between molecular groups and subsequently, more oriented structures.

5.3. Color properties

Cold plasma treatment has been shown to have a significant effect on the color properties of non-starch hydrocolloids and their derivatives. These properties become a chief factor when the appearance of samples such as powders and biocomposite films is important. When plasma treatment is conducted at high temperatures (much higher than 50 °C) the possibility of pigment formation through reactions in the plasma environment increases and the change in the color of hydrocolloids will be obvious [82,107]. However, most investigations on the plasma treatment of hydrocolloids are done below these temperatures and the reason for the color change is assigned to other factors such as chemical changes and changes in porosity and light scattering. For example, it is reported that the oxidation reaction is the primary chemical factor of color change during the plasma treatment of hydrocolloids [63,78,82]. Moreover, the etching phenomenon and surface roughness are regarded as physical changes that affect the color properties during the plasma treatment of hydrocolloids and their derivatives [108]. Generally, there is no report to assert how the color properties of non-starch hydrocolloids and their derivative changed after cold plasma treatment and there are controversial reports on color changes after plasma which can be due to the condition of reaction and the nature of samples. For example, Misra et al. (2018) [78] investigated the effects of cold plasma at atmospheric pressure on the color, rheological, and emulsion stabilizing properties of xanthan gum (XG), and chemical changes were identified qualitatively by infrared spectroscopy. As xanthan gum is a white to tan colored powder, it was found that plasma treatment of XG increased the whiteness parameter and this difference became more obvious during storage. Although the reason for the color change was unknown, it is declared that plasma is a known source of ROS and this color change can be due to oxidative reactions [78,109]. However, this change in color properties was similar to the previous report by Misra et al. (2014) [41] for treated and untreated cold plasma strawberries [41]. The opposite trend of color change was found in another study conducted by Amirabadi et al. (2020) [82]. Cold plasma treatment did affect the whiteness (L* score) and redness (a* score) in negative mode and yellowness (b* score) in positive mode. However, prolonged time of treatment did not cause more changes in whiteness but yellowness and redness increased and decreased respectively. The reason for this color instability might be due to oxidation and other factors such as chemical reactions and altered physical properties (texture, surface roughness, etc.) [82].

Bulbul et al. (2019) [92] reported a marginal increase in yellowness after cold plasma treatment which might be due to the increase in porosity of XG powder due to the etching phenomenon. It has also been suggested that the change in color properties may be due to the formation of new chemical groups and the breakdown of existing groups during plasma treatment [110]. To the best of our knowledge, the results of color properties of plasma-treated gums and their derivatives are inconsistent due to the fact that this property is determined by various factors such as the method of treatment (instrument, time, and intensity), the composition of feed gases, and the physical states of samples (powder or film).

5.4. Rheological properties

The rheological properties of hydrocolloids can be modified by plasma treatment. For example, Jampala et al. (2005) [6] found that plasma treatment with dichlorosilane (DS) enhanced the rheological properties of xanthan gum (XG). Soft gels made from both untreated and plasma-treated XG samples showed more elastic modulus (G′) than viscous modulus (G″), but the treated XG had greater dynamic moduli than the untreated XG. This suggests that the network in the treated sample was stronger than in the untreated gels. In addition, functionalizing of XG in DS-plasma conditions and subsequent grafting of amine I enhanced the XG cross-linking. Cold plasma treatment of XG strengthened the network as measured by the dynamic storage modulus. Misra et al. (2018) [78] also investigated the rheological properties of plasma-treated XG and found that the treated samples showed a significant increase in viscosity followed by a decrease at low shear rates, while no significant increase was observed for the untreated sample. This is likely due to the formation of a more ordered network with higher hydrogen bonds through plasma treatment, as intermolecular interactions such as hydrogen bonds and interwoven polymer chains result in high viscosity in dispersed solutions at low shear rates. Extended plasma treatment time caused even more viscosity in low shear rates, which could be a function of treatment time.

Bulbul et al. (2019) [92] studied the changes in rheological properties of XG due to plasma treatment at different power and exposure times. Both untreated and treated XG showed shear-thinning behavior, and increasing the XG concentration resulted in an increase in viscosity at a specific shear rate. At a specific time and power of plasma treatment, treated XG showed higher viscosity in comparison with untreated XG, likely due to cross-linking and higher intermolecular interaction in treated samples.

High methoxy pectin (HMP) solutions also exhibit a pseudoplastic trend, and Momeni et al. (2018) [77] found that plasma treatment of HMP resulted in shear-thinning behavior and a considerable increase in apparent viscosity from 85.23 to 92.65 MPa. Treated HMP gels had a shorter linear viscoelastic region, lower resistance to breaking, and higher elastic (G′) and viscous modulus (G″) than untreated HMP gels.

The higher resistance to breaking in treated HMP gels might be due to more effective inter-chain interaction and lower electrostatic repulsion at low pH values, resulting in a stronger matrix in the treated samples and higher storage modulus (G′) compared to the untreated samples. However, the new network structure in the treated samples required more time and caused a reduction in G′ through frequency sweep testing. Thus, plasma treatment of hydrocolloids can result in a stronger network with higher viscosity and storage modulus (G′), likely due to the formation of a more ordered structure with increased intermolecular interactions such as hydrogen bonds and cross-linking.

5.5. *Thermal properties*

The thermal properties of hydrocolloids have been investigated by several studies. Tang et al. (2016) [111] compared the differences between cross-linked sesbania (CLSG), dialdehyde cross-linked sesbania gum (DCLSG), and sesbania gum (SG) and attributed the differences to the new bonds of aldehyde groups and –(CH2)3. The molar mass distribution of CLSG was more limited than DCLSG and SG. The addition of aldehyde groups in SG resulted in more thermal stability than SG, and this may be due to the congestion of neighboring aldehyde groups in DCLSG [111]. This finding was consistent with the study reported by Momeni et al. (2018) [77] which showed that plasma treatment led to a lower weight loss in treated samples, indicating a stronger network with higher thermal properties that were developed during plasma treatment. However, Pankaj et al. (2017) [88] reported contrary results to Tang et al. (2016) [111], as they found that applying plasma treatment of chitosan films did not affect the thermal stability and the main phase of weight loss of films. Li et al. (2019) [95] observed that by increasing the power of plasma treatment of chitosan films, more thermal stability was observed, which could be attributed to the increase in crystallinity of chitosan films caused by increasing the power of treatment. On the other hand, another study conducted by Bulbul et al. (2019) [92] reported that plasma treatment of XG did not cause a significant effect on the thermal properties of XG. In comparing these results, it can be concluded that plasma treatment can enhance the thermal properties of hydrocolloids as long as it is able to develop new bonds and ordered structures. However, in some conditions, the plasma treatment may not be effective enough, and no significant number of bonds are created. Moreover, harsh conditions of plasma treatment, such as higher power or time, can cause cleavage in newly built bonds and natural bonds of hydrocolloids, which can lead to lower thermal stability compared to untreated samples.

6. Stereochemical changes in proteins and their derivatives

6.1. *Proteins*

Research on plasma treatment of proteins and their derivatives has been ongoing for the last decade. One novel application of cold plasma is the treatment of food products that contain protein-based allergens, as it is an effective way of deactivating these compounds [112]. Thermal processing can inhibit allergic effects by altering or destroying the conformational structure of epitopes caused by heat treatment, but it may have negative effects on the food and certain allergens, such as tropomyosin, are heat-resistant. Non-thermal processing techniques, such as cold plasma treatment, pulsed light, high-pressure processing, and gamma irradiation, are more practical alternatives. Cold plasma treatment can alter the conformation of certain proteins, reducing their allergenic effects, while the other components in food remain largely unchanged [113,114]. Some hypothesized mechanisms for the allergen-inhibitory effects of cold plasma treatment include conformational changes of epitopes during crosslinking, reactive species generated in plasma discharge, and oxidation reactions [112].

The conditions of plasma treatment, such as applied power, treatment time, and environment (dry or wet), may affect the results. For example, Tammineedi et al. (2013) [115] demonstrated that using non-thermal atmospheric plasma and ultrasound treatments had no significant effect on allergenicity reduction in a-casein and whey proteins in comparison with UV-C and high-intensity ultrasound technologies.

Zhang et al. (2021) [116] studied the changes in physicochemical, structural, and allergenic properties of soybean protein isolate (SPI) solution affected by non-thermal plasma treatment. They found that the color properties were not affected, but the pH of the SPI solution reduced slightly after plasma treatment, possibly due to hydrogen peroxide and nitrous acid production through the plasma treatment. The emulsification property of SPI improved from 56 % (for native SPI) to 168 % (for plasma-treated SPI), while the foam-forming ability increased from 60 % to 194 %. At the frequency of 120 Hz for 5 min treatment, immunoglobulin E (IgE) binding was reduced by up to 75 %. The researchers suggested that increasing the carbonyl group increases protein oxidation through plasma treatment [117]. Protein oxidation was found to be time-dependent, as reported in a previous study conducted by Segat et al. (2015) [63], in which the oxidation of whey protein was extended by increasing the treatment time. The thiol group levels in protein were reduced by extending the treatment time, which was consistent with the rise of carbonyl groups. Mild oxidation of protein through plasma treatment increases its hydrophilicity, while extended oxidation increases its hydrophobicity [118].

Stereochemical studies have shown that α-helix and random coil content slightly reduced and extended, respectively, with increasing treatment time. However, other studies have reported that plasma treatment results in a decrease in α-helix and β-sheet content of proteins extracted from whey, peanuts, wheat flour, and lactate hydrogenase enzyme [7,119–121]. It is concluded that the natural structure of proteins and the type of plasma treatment are the two major factors that affect the structural changes in protein molecules. Gharbi & Labbafi (2019) [122] studied the effects of atmospheric plasma treatment on the structure and functionality of egg white protein, finding that the plasma treatment caused changes in protein conformation, leading to reduced surface hydrophobicity and increased surface charge, which resulted in improved emulsification properties. Another study by Wu et al. (2020) [123] investigated the effects of atmospheric plasma treatment on the allergenicity of egg white protein. They found that plasma treatment led to a decrease in IgE binding, and suggested that the mechanism may involve changes in protein structure and the generation of reactive oxygen species.

In a study by Zhang et al. (2021) [124], cold plasma treatment was applied to soybean protein isolate, and its effects on functional properties and allergenicity were investigated. They found that plasma treatment improved protein solubility and emulsifying properties, and also resulted in a significant decrease in IgE binding. Furthermore, Taha et al. (2022) [125] studied the effect of plasma treatment on the structure and properties of bovine serum albumin (BSA). They found that plasma treatment caused changes in protein structure and resulted in improved solubility and emulsification properties, but also led to an increase in protein aggregation. Overall, these recent studies suggest that plasma treatment can be a promising approach for improving the functional properties of proteins and reducing their allergenicity, but the specific effects may depend on the type of protein, plasma treatment conditions, and other factors.

6.2. *Protein derivatives*

Physical treatments have emerged as a promising method for improving the properties of biocomposite films. Plasma treatment is one such method that has been investigated for its effectiveness in modifying the surface properties of biopolymer films. Plasma treatment involves subjecting the surface of a film to low-pressure gas plasma discharge, resulting in chemical and physical changes in the surface properties of the film. Chen et al. (2019) [3] investigated the effect of plasma treatment on zein films composited with chitosan. They found that shortduration plasma treatment (60 s) led to an improvement in tensile strength (TS) and elongation at break (EAB) of the film. However, longer treatment times resulted in decreased TS and EAB. In addition, the plasma treatment led to reduced water vapor permeability (WVP), indicating an improvement in shelf life. Similarly, Wu et al. (2020) [123] obtained comparable results for cold plasma treatment of casein-based edible films. The plasma treatment led to higher EAB and TS values, and a lower WVP, which could be attributed to improved toughness and compactness of the film, as confirmed by SEM imaging and ATR-FTIR results.

To enhance the properties of zein films, Cui et al. (2020) [126] developed zein films incorporated with pomegranate peel extract (PPE) encapsulated in chitosan nanoparticles (CNP) and modified the surface of the nanocomposite film using cold nitrogen plasma. They found that the plasma-treated film showed a rough surface due to the etching effect, and X-ray photoelectron spectroscopy (XPS) spectra analysis revealed additional C and N molecules after plasma treatment. Plasma treatment improved intermolecular interaction between zein biopolymers and CNPs, reducing the rate of release of PPE and slowing down the growth of L. *monocytogenes*, resulting in a lower release rate of PPE from the plasma-treated film than from the untreated one. Moosavi et al. (2020) [127] studied the modification of whey and gluten films using cold plasma treatment with air and argon gases. AFM results showed a significant increase in whey film roughness, while the opposite was observed for gluten film, which was attributed to the shadowing effect. ATR-FTIR analysis showed an increase in β-sheet structure and cross-linking formation on the surface of both biopolymers due to conformational changes during plasma treatment. Cold plasma treatment of both protein films led to an increase in wettability and reduced contact angle, which could be attributed to the formation of polar groups on the surface of both biopolymers.

In summary, physical modification methods, such as cold plasma treatment, have been shown to enhance the properties of biopolymer films by improving mechanical properties, reducing WVP, increasing wettability, and improving intermolecular interaction between the biopolymers and active ingredients. These improvements can lead to increased shelf life and slower release rates of active ingredients, making them suitable for food packaging applications. Furthermore, the application of plasma treatment to various protein-based biopolymers can result in unique surface properties, enabling the development of new functionalized materials with a wide range of applications. Table 2 summarizes investigations on the effects of plasma treatment on the physicochemical properties of proteins, categorized by their respective resources [3,7,85,116,127–135]. For each investigation, changes in different parameters are presented as vertical arrows, with upward arrows indicating an increase and downward arrows indicating a decrease.

7. Application of plasma-treated proteins and their derivatives

7.1. Health-beneficial effect

Applying the plasma treatment to numerous proteins in order to obtain modified and desired properties is not a new method. However, over time scientists tend to exploit this technique for different points of view or novel protein derivatives. As will be discussed in the following section, food scientists have used plasma treatment to lower or eliminate the allergenicity of special proteins in food formulations which is a new perspective on the plasma treatment of proteins. For example, Venkataratnam et al. (2019) [120] reported the significant effect of the plasma treatment on the reduction of allergenicity of dry, defatted peanut flour protein (Ara h 1). The effectiveness of reduction was 43 % which was attributed to the alteration in the secondary structure of this protein. In another study, the effect of plasma treatment on the immunoreactivity of soy protein isolate was investigated. In this study, cold plasma treatment was compared with gamma and UV radiation. The findings showed that the highest reduction (91–100 %) in immunoreactivity was observed in the case of plasma treatment which revealed the potential of this method in comparison with the other approaches [136]. Many investigations revealed that plasma treatment is an effective method in the reduction of the activity of trypsin inhibitors and this raises the nutritional quality of proteins and protein-containing food products [137–139]. It is said that trypsin inhibitor is the prominent anti-nutritive factor in soybeans which is said to be inactivated using plasma treatment due to the significant alteration in secondary and tertiary structures. For example, Xu et al. (2022) [140] found out that dielectric-barrier discharge cold plasma resulted in the elimination of soybean trypsin inhibitors. They also reported that this non-thermal processing caused a substantial alleviation for other destructive effects of soybean trypsin inhibitors such as causing an interruption in the function of the liver and kidney. A similar finding was reported by Li et al. (2017) [141] when they studied the effect of plasma treatment on soybean trypsin inhibitors. It was reported that applying the plasma treatment for 21 min at 51.4 W resulted in the highest (86.1 %) reduction in trypsin inhibitors.

7.2. Technological benefits

There are numerous technological applications of proteins including using them as emulsifiers, texturizing, and foaming agents in the food industry. Nowadays, improving these properties through protein modification has gained great attention and one of the successful approaches is found to be plasma treatment. For example, enhancing the physical properties of the protein-based composite film was done by Song et al. (2019) [142]. They applied the plasma treatment to a composite film prepared using whey protein concentrate and starch. It was concluded that due to the new cross-linked network formed after the plasma treatment the thermal stability of the composite film substantially increased. A proper treatment time (60 s) and power (400 W) endowed the highest tensile strength. In addition, the plasma treatment enhanced the barrier properties of composite films which means the water vapor permeability (WVP) and oxygen permeability reduced significantly after plasma treatment. Findings also revealed that the foam properties of soybean protein isolate can be improved using air cold plasma treatment. In this case, lower frequencies resulted in higher foam-forming properties while higher frequencies endowed higher foam stability [116]. Another techno-functional application of protein is using them as emulsifiers. Ji et al. (2018) [85] stated that the emulsification properties of peanut proteins gain the highest point when the plasma treatment was conducted at 35 V for 2 min. They claimed that this phenomenon is due to an increase in β-sheets and a reduction in the number of α-helices and β-turns. In addition, gelation is another interesting property of proteins such as pea protein, gelatin, and myofibrillar proteins in the food industry. Zhang et al. (2021) [124] reported the mechanism of the modification of gelling properties of pea protein using a transmission electron microscope and FTIR spectrometer. Applying the plasma treatment at 3500 Hz for 10 min resulted in the highest gelling properties. They stated that higher hydrogen bonds and higher surface hydrophobicity resulted in higher disulfide bonds and led to stronger gels in comparison with native proteins.

8. Comparison of plasma treatment effects on non-starch hydrocolloids and their derivatives

8.1. Factors affecting plasma treatment effects

To understand the different effects of cold plasma treatment of hydrocolloids and proteins, it is essential to appreciate the major factors affecting the modification of such macromolecules. Based on the above-mentioned information about plasma chemistry and the changes that occurred in hydrocolloids and proteins, the type of reactive groups presents in the macromolecule, and the distance of these groups from reactive species can be regarded as intrinsic factors and the amount and

Table 2
Effects of plasma treatment on the physicochemical properties of proteins (categorized by resource). Changes in different parameters are presented as vertical arrows, with upward arrows indicating an increase and downward arrows indicating a decrease.

Protein source/ derivative	Results					Reference
	Mechanical properties/ particle size	Hydrophilicity or solubility	Interfacial properties	Chemical changes		
				Functional groups/ amino acids	Structure	
Wheat flour	↑ Dough strength	–	–	–	↑ α-Helix ↑ Stability of the second structure ↑ β-Turn ↓ β-Sheet ↑ Disulfide bond	[7]
Peanut		↑	↑		↑ β-Sheet ↑ Random coils ↓ α-Helix	[85]
Glycosylated Peanut protein		↑	↑	↓ Lysin ↓ Phenylalanine	↓ α-Helix ↓ β-Sheet ↑ β-Turn ↓ Compactness	[128]
Glycosylated Peanut protein		↑ For short-time treatment ↓ For long-time treatment	↑			[129]
Chocolate milk drinks	↑ Particle size ↑ Viscosity	↓	↑	↓ Carbonyl groups ↓ Free sulfhydryl groups	↑ Disulfide bond	[130]
Zein bio-film	↑ TS & EAB (For short-time treatment) ↓ TS & EAB (For long-time treatment) ↓ WVP	↑	–	–	–	[3]
Casein-based edible film	↑ TS & EAB ↓ WVP	↑	–	–	–	[131]
Whey and gluten bio-films	↑ TS (After 5 and 10 min) ↓ TS (>10 min) -WVP was unchanged ↓ Oxygen permeability	↑	–	Dissociation of C—H ↑ Oxygen-containing groups ↓ Free sulfhydryl groups	↑ β-Sheet	[127]
Zein film	↓ WVP	↑	–	↓ Free sulfhydryl groups	↑ Hydrogen bonds ↑ α-Helix and ↑ Disordered conformation ↑ S—S bonds	[132]
Myofibrillar fish-based film	↑ TS ↑ EAB ↑ WVP (After 2 min) ↓ TS ↓ EAB ↓ WVP (After 5 min)	↑	–	–	–	[133]
Casein	–	–	–	↓ Content of amino acids (For long-time treatment)	↑ β-Sheet ↑ β-Turn ↑ α-Helix -Random coils remained unchanged	[134]
Whey proteins	–	–	–	–	↓ α-Helix ↑ β-Sheet (Spark discharge) ↓ β-Sheet (Glow discharge) ↓ β-Turn (Glow discharge) ↓ β-Turn (Spark discharge) -Random coils were unchanged	[134]
Soybean protein isolate solution		↑ Hydrophilicity (For mild oxidation) ↓ Hydrophilicity (For harsh oxidation)	↑ Emulsification capacity ↑ Foam forming capacity	↓ Free sulfhydryl groups ↓ Carbonyl groups	↓ α-Helix ↓ Random coil	[116]
Grass pea protein isolate	↓ Particle size	↓ Hydrophilicity ↓ Solubility	↑ Emulsification capacity	↓ Carbonyl groups ↓ Cross-linked amino acids ↓ Free sulfhydryl groups ↓ Hydrogen bonds	↑ Compactness in the third structure	[135]

type of reactive species generated in the plasma environment are called extrinsic factors. These factors are affected by voltage, frequency, time, and the type of inlet carrier gas. The carrier gas, as mentioned earlier, can be nitrogen, argon, or ambient air which can react with water or water vapor and generate nitrate ions or ozone as durable reactive species or hydroxyl and oxygen radicals as short-term reactive species. On the one hand, the presence of a diverse range of functional groups such as hydroxyl, carboxylic acid, and sulfhydryl causes versatile outcomes. On the other hand, non-starch hydrocolloids possess hydrogen and hydroxyl groups as the predominant reactive groups available to interact with reactive species. However, some of these macromolecules are accompanied by other functional groups such as the amino/acetamido group in chitosan, amine and carboxyl group in gum Arabic, and ester sulfate groups in carrageenan which lead to more and more types of modified hydrocolloids. In the following section, the differences between treated and untreated non-starch hydrocolloids and their derivatives are discussed in detail and the distinctive differences will be debated.

8.2. Similarities and differences in plasma treatment effects

Understanding the parameters which ought to be measured after the plasma treatment of hydrocolloids is crucial for scientists who investigate the plasma treatment of such macromolecules. These parameters differ in different investigations and are based on the purpose of the research. When the hydrophobic modification of hydrocolloids is aimed, researchers tend to measure the chemical composition, molecular and structural differences, solubility, and interfacial properties after the plasma treatment. Crystallography evaluation is another prominent parameter that should take into account. The importance of this measurement arises from the pattern by which water molecules can penetrate the structure and hydrate the powder or the composite film [143]. In addition, sometimes hydrocolloids are blended to form a film-forming formulation. When these biopolymers are miscible and compatible with each other, the crystallinity of the resulting film will be more than the corresponding individual hydrocolloid used [144]. It is also known that the crystalline content of non-starch hydrocolloids is affected by chemical alteration during plasma treatment [145]. Color properties can be important when the modified hydrocolloid is regarded as an ingredient in food formulation. Therefore, any undesirable changes in the color of hydrocolloids resulting from plasma treatment make this method a challenging approach to modifying the ingredient. Besides, color properties indicate important changes such as oxidation and microbial growth in samples. For instance, a decrease in the lightness of plasma-treated hydrocolloids is attributed to the oxidation reaction [95]. Rheological properties are also another factor, which play an important role in food formulation. Since the predominant application of hydrocolloids stems from the viscosifying effect of these biopolymers, measurement of this parameter is vital in research [146]. In addition, thermal properties are also an indication of chemical changes, changes in crystalline regions, and the stability of samples during heat processing. Thus, measurement of this parameter can be beneficial to validate the XRD, FTIR, and NMR results.

Comparatively, scientists consider some similar properties of both hydrocolloids and proteins when they are investigating the plasma modification of such macromolecules. For example, gellan and gelatin have been extensively used as viscosifier or gel-forming agents in food formulation and researchers tend to measure the modified viscosity of both polysaccharide-based and protein-based hydrocolloids after plasma treatment. Furthermore, the film-forming ability is another common criterion considered by scientists after plasma treatment of both hydrocolloids and proteins. This is because many investigations have revealed the high efficiency of the plasma treatment of such biopolymers to form film-forming formulations with suitable mechanical properties and desired barrier characteristics [86,147]. Measurement of interfacial properties is also frequent among these macromolecules which has gained great attention among scientists because they believe that it is vital to develop natural and green surfactants and emulsifiers and avoid exploiting synthetic ones [148].

To compare non-starch hydrocolloids and their derivatives, there are some differences after plasma treatment which are rationalized according to the difference in nature of the hydrocolloid or the treatment parameters. For example, occurring oxidation during plasma treatment is validated for hydrocolloids and proteins which indicates the addition of oxygen-containing molecules to these biopolymers. Some studies reported an increase in the crystallinity of hydrocolloids and it was attributed to the new cross-linked structure formed after plasma treatment. However, some other investigations claimed a decrease in crystallinity. These controversial results may arise from the differences in hydrocolloids used in experiments and differences in the condition of treatment. Similarly, scientists face opposite results in comparison with other researchers' reports on color properties measurement. The above-mentioned reason is also involved in this circumstance. In contrast, many studies have reported that rheological characteristics of non-starch hydrocolloids improved after plasma treatment which was assigned to newly formed bonds and a more structured network. This stronger structure also results in more thermal stability for these biopolymers.

9. Cold plasma in the food industry

9.1. Dairy industry: enhancing sensory properties

The sensory properties of dairy products, including taste, texture, and aroma, are essential factors that influence consumer acceptance and preference. These properties are influenced by various factors, including processing, storage, and composition. Understanding the sensory properties and their importance can help dairy manufacturers develop products that meet consumer expectations, leading to increased sales and customer satisfaction. This section covers the discussion of how plasma treatment improves different aspects of the sensory properties of dairy products.

Volatile compounds in dairy products can have both positive and negative effects on sensory properties. Some volatile compounds contribute to desirable flavors and aromas, while others can cause off-flavors and off-odors. Regarding desirable volatile compounds, diacetyl, 2,3-pentanedione, aldehydes, ketones, and lactones contribute to buttery, creamy, fruity, floral, and nutty notes, enhancing the overall sensory experience and consumer acceptance of dairy products [130,131].

Coutinho et al. [132] assessed the impact of cold plasma process parameters, specifically processing time (5, 10, and 15 min) and nitrogen gas flow rate (10, 20, and 30 mL/min), on the volatile compounds of a chocolate milk drink. The findings of their study showed that the mild and severe cold plasma processing conditions led to the loss of some crucial volatile compounds. On the other hand, intermediate processing conditions resulted in higher levels of ketones, esters, and lactones, which are crucial flavor and aroma compounds in chocolate milk. This indicates that these important compounds were better preserved in the intermediate cold plasma conditions compared to the control sample (pasteurized product), thereby maintaining the desired flavor and aroma of the chocolate milk.

In another study, Coutinho et al. [133] assessed the impact of cold plasma processing parameters, including time and flow rate, on the physical properties of chocolate milk drinks. These properties include rheological parameters, which describe the flow behavior of the chocolate milk, and particle size, which is the size of the individual particles in the drink. Similar to the previous study, intermediate processing conditions displayed characteristics that were more comparable to the pasteurized product. The combination of parameters used in this formulation may produce some effects similar to those achieved by heat during pasteurization. This caused some disruption of the ingredients in the milk drink, resulting in slightly higher protein oxidation and aggregation. The slightly higher consistency of the sample treated in intermediate processing could be attributed to its particle size, as the particles in this formulation were slightly larger than those in the pasteurized product. In the treatment of a dairy beverage based on whey proteins, the rheological analysis showed that the plasma-treated beverages exposed to intermediate and high processing times had moderate viscosity and consistency, whereas the beverage exposed to low processing times exhibited high viscosity and consistency. Additionally, it was found that the plasma treatment improved consumer acceptability [134].

Enzymatic browning is a chemical reaction that occurs when enzymes in dairy products, such as phenol oxidase and peroxidase, react with oxygen in the presence of water to produce brown pigments called melanoidins. While enzymatic browning can enhance the flavor and color of some dairy products, such as cheese and butter, it can also have negative effects. The formation of melanoidins can alter the appearance, texture, and flavor of dairy products, resulting in a bitter taste, off-odors, and a darker color. Additionally, enzymatic browning can lead to a decrease in the nutritional content of dairy products, particularly vitamins and amino acids, due to the breakdown of these compounds during the reaction. The use of cold plasma was observed to inactivate enzymes that cause undesirable color changes and off-flavors in dairy products [135]. While several studies have investigated the impact of cold plasma treatment on the physicochemical and microbiological properties of dairy products, further research is needed to evaluate its effect on sensory properties. Thus, understanding the effect of cold plasma treatment on sensory properties is essential in determining its potential as a food processing technology for dairy products. Further research can help to identify optimal processing parameters that preserve the sensory quality of dairy products while ensuring food safety and extending shelf life.

9.2. *Meat industry: safety and shelf life*

Cold plasma is a promising technology that has been gaining attention in the meat industry as a potential solution to improve food safety and extend shelf life. In the meat industry, cold plasma can be applied to various stages of the production, from processing to packaging. One of the primary benefits of using cold plasma is its ability to inactivate microorganisms such as bacteria, viruses, and fungi. This property makes it a useful tool for reducing the risk of foodborne illnesses and improving product safety. By reducing the microbial load on the surface of the meat, cold plasma can help to slow down spoilage and preserve freshness. Given the continual advancements in plasma science, plasma technologies may eventually supplant traditional decontamination methods within the meat industry. In the foreseeable future, cold plasma could become a prominent tool for ensuring the safety and quality of meat products.

The reactive species are highly effective at breaking down the cell walls of microorganisms, such as bacteria. The charged particles in the plasma cloud also generate an electric field that disrupts the cell membranes of these microorganisms, leading to their destruction [149]. Additionally, the UV radiation emitted by the plasma cloud can cause damage to the DNA of microorganisms, impairing their ability to reproduce and survive. This combination of physical and chemical processes effectively eliminates the microorganisms present on the surface being treated [150]. Furthermore, cold plasma can penetrate small crevices and hard-to-reach areas, making it an effective tool for decontaminating complex surfaces. The non-thermal nature of cold plasma also means that it does not cause thermal damage to the treated surface and formation of carcinogenic compounds such as polycyclic aromatic hydrocarbons (PAHs), making it a safe and efficient decontamination method [151,152]. While studies have shown that cold plasma can improve the safety, quality, and shelf life of meat products, there is still much to learn about the optimal conditions for application and the potential long-term effects on the nutritional and sensory properties of meat products. Furthermore, the effectiveness of cold plasma technology can also be affected by the type and thickness of the meat product being treated. Different types of meat products may require different treatment times and conditions, and the thickness of the product can affect the penetration of the reactive species generated by cold plasma [153].

Among the various pathogens present in meat and meat products, *Escherichia coli* and *Listeria monocytogenes* are widely acknowledged to be the most resilient human pathogens [154]. According to Lee and colleagues [155], the use of an atmospheric plasma jet operating in both helium and a combination of helium, oxygen, and nitrogen, was found to be effective in treating *L. monocytogenes* on cooked chicken breast and ham, resulting in a longer shelf life for the treated products. The bactericidal effect of cold plasma treatment against *Escherichia coli* remains rather limited in comparison with other pathogens in meat across many documented studies that employed a comparable plasma source [151,156,157]. It has also been reported that applying plasma treatment at ambient pressure can reduce the *Listeria monocytogenes* count by 2.5 $\log_{10}$ in meat products [158,159].

9.3. *In-packaging cold plasma treatment*

In-packaging cold plasma treatment is a technology that has gained increasing attention in recent years, due to its potential for improving the safety and quality of packaged foods. The technology works by generating a non-thermal plasma within the food packaging and it is effective against a wide range of microorganisms, including bacteria, viruses, and fungi. This method offers the benefit of serving as a complementary sanitization process that eliminates cross-contamination [160]. Packaging material plays a key role in this process and by exposing the surface of the packaging material to reactive plasma species, the internal surface of the package is decontaminated. Cold plasma has proven to be effective in reducing the microbial load of various packaging materials, such as PET foils, polystyrene, LDPE, and other polymeric materials [161]. However, it is essential to evaluate all potential changes that the cold plasma may induce in the packaging material for safety and quality reasons. For example, the migration of molecules and alteration in barrier properties of packaging are included as crucial issues [162].

Misra and colleagues [163] evaluated the effectiveness of in-packaging cold plasma treatment of strawberries. Dielectric barrier discharge with the air as feeding gas was applied to polypropylene packaging material containing fresh strawberries. The results showed that without significant effects on the respiration rate, the plasma treatment could reduce the total count (aerobic mesophilic bacteria, yeast, and mold) by 2 $\log_{10}$. In a study that assessed the decontamination of fish, Chiper and colleagues [164] found that the in-package treatment of smoked salmon by argon as feeding gas can reduce the *Photobacterium phosphoreum* count by approximately 3 $\log_{10}$.

9.4. *Modification of packaging materials*

As mentioned in the previous section, cold plasma treatment can modify the surface chemistry of food packaging polymers by introducing functional groups, such as carboxyl, hydroxyl, and amino groups, which can enhance surface reactivity and improve adhesion to coatings and other materials. This can improve the barrier properties of the packaging material, such as reducing gas permeability, which can help to maintain product freshness and extend shelf life. Cold plasma treatment can also modify the surface morphology of food packaging polymers by inducing surface roughness, which can enhance surface area and improve printability. Ongoing research and development in this area are likely to lead to further improvements and expanded applications of this technology in the food industry.

Polyethylene is one of the most commonly used packaging materials in the food industry due to its versatility, cost-effectiveness, and excellent barrier properties. Polyethylene has low surface energy because it is a non-polar material, which can make it challenging to adhere to or print on its surface [165]. Studies have shown that cold plasma treatment leads to higher roughness and lower contact angle of Polyethylene films [166]. Polypropylene is also a widely used packaging material in the food industry because it is versatile, chemically inert, and thermally stable. The low surface energy of polypropylene can be challenging in printing, coating, and lamination applications, which may necessitate additional surface treatment to increase its surface tension [167]. It has been reported that cold plasma treatment of polypropylene makes it a suitable candidate to be laminated with other biodegradable active polymers such as carboxymethyl cellulose [168] and chitosan [169].

Overall, the use of cold plasma treatment in the modification and treatment of biopolymers offers several advantages, including biocompatibility, elimination of toxic solvents, and suitability for heat-sensitive biodegradable polymers. Numerous investigations have shown that cold plasma treatment can improve the structural, mechanical, and thermal properties of film composites, as well as the surface characteristics of protein-based films. While it does not affect water vapor permeability, cold plasma-treated edible packaging films have demonstrated significant improvement in their antimicrobial activities [170]. For example, it has been reported that cold plasma treatment can reduce the water vapor permeability of zein films [171], zein-chitosan composite films [3], and starch-based films [172]. Hu and colleagues [173] found that cold plasma treatment of poly (lactic acid) films resulted in an improvement in antimicrobial activities of the film due to higher absorption of nisin after plasma treatment of poly (lactic acid) films. Plasma treatment of active chitosan films has been shown to improve the thymol diffusion into films and the higher loading capacity of this thymol led to higher antimicrobial activities of biodegradable active packaging [88].

10. Challenges and opportunities in using plasma treatment on non-starch hydrocolloids and their derivatives

10.1. Challenges

As mentioned earlier, the surface-effective characteristic of plasma is regarded as its primary challenge which can limit its potential in biopolymer modification. In addition, working with plasma as a tool to modify the chemical or physical properties of hydrocolloids can also be challenging. This is because controlling the condition of the reaction such as temperature is rather difficult. Furthermore, sometimes, applying the same parameters of reaction such as power and time for a particular biopolymer is found to result in unidentical outcomes. Thus, controlling the reactions in the plasma environment and attaining the expected result is another challenging factor for scientists in the modification of hydrocolloids and other biopolymers. Future research requires a focus on developing efficient and cost-effective plasma treatment methods to modify non-starch hydrocolloids. Moreover, efforts should be made to investigate the potential of plasma-treated biopolymers in various food and non-food applications, including encapsulation techniques, drug delivery systems, and novel biopolymer-based antimicrobial agents and antioxidants.

10.2. Opportunities

Cold plasma treatment has emerged as a promising technology for enhancing the properties of non-starch hydrocolloids. While the conventional application of hydrocolloids for viscosifying has been well established, novel applications such as emulsion stabilizing, microencapsulation, and preparation of packaging materials using modified hydrocolloids are gaining momentum. For example, the surface-effective properties of plasma treatment are beneficial in the preparation of food packaging materials. Scientists tend to treat the packaging polymers to obtain some desired functionalities such as printability, controlled barrier properties, and antimicrobial and antioxidant activities. Applying this technology to enhance the structure of composite film with two or more biopolymers is also another bonus. Therefore, researchers can conduct more studies on such properties of cold plasma for non-starch hydrocolloids. Moreover, there is a limited number of studies on increasing the bioavailability of nutraceuticals through a reaction with non-starch hydrocolloids. Therefore, this can be a technique not only to functionalize these biopolymers but also to raise their nutritional quality when they are used as emulsifiers, edible packaging material, or viscosifiers.

11. Conclusion and future prospects

Taken together, the use of cold plasma treatment has emerged as a novel and promising technology for modifying non-starch hydrocolloids. Reactive species such as excited atoms and molecules, ozone, radicals, and electrons play a critical role in altering the molecular structure, weak bonds, and surface functionality of these biopolymers. The oxidation, cross-linking, and hydrogen bonding effects of cold plasma treatment result in significant changes in crystallinity, chemical structure, and hydrophilicity. Although surface effectiveness and high energy requirements are limiting factors, plasma treatment is a green, non-toxic, and facile modification strategy for preparing diverse packaging materials and functional ingredients. The alteration in crystallinity of hydrocolloids through plasma treatment can lead to unpredictable changes due to the surface-effective properties of plasma and reactions of reactive species with the polymer components. Nonetheless, increasing the viscosity of solutions due to cross-linking effects and newly formed hydrogen bonds is a predictable outcome.

In the case of proteins, plasma treatment can enhance the stability of the secondary and tertiary structure, increase water-holding capacity, and improve emulsion stability. The effects of plasma treatment on protein-based edible films can vary, but it can enhance the conjugation of proteins and conjunction agents resulting in proper characteristics as a packaging material. Therefore, based on the findings of this review paper, it can be said that cold plasma treatment has the potential to revolutionize the field of food science and technology by providing an efficient and cost-effective modification method for non-starch hydrocolloids. Future research should focus on optimizing the plasma treatment conditions and exploring the potential of plasma-treated biopolymers in various food and non-food applications. Regarding its industrial potential, plasma treatment is a promising approach for enhancing the sensory properties of dairy products. It has been demonstrated to improve the volatile compounds, color, and rheological properties of dairy products, leading to an improved flavor, texture, and aroma. The application of plasma treatment in the dairy industry offers a safe and effective way of extending the shelf life of dairy products while maintaining their sensory quality and nutritional value. However, further research is still required to optimize the processing parameters and investigate the effects of plasma treatment on various food products. Despite this, the latest findings indicate that plasma treatment has a significant impact on the sensory properties of dairy products, making it a promising technology in the food industry.

Ethical approval

This study does not involve any human or animal testing.

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CRediT authorship contribution statement

Shahriyar Sahraeian: Conceptualization, Data curation, Methodology, Formal analysis, Software, Investigation, Writing – original draft. **Ali Rashidinejad:** Supervision, Investigation, Validation, Writing – review & editing. **Mehrdad Niakousari:** Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare no conflict of interest.

Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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