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Review

Cold Plasma Processing on Fruits and Fruit Juices: A Review on the Effects of Plasma on Nutritional Quality

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Abstract: This review aims to present the effects of cold plasma technology on the nutritional quality of fruits and fruit juices. This review focuses on the chemical changes induced by plasma on several bioactive compounds, such as sugars, starch, lipids, vitamins, phenolic compounds, carotenoids, and anthocyanins. The main plasma-reacting species that reacts with fruit compounds are presented and discussed. The review presents the mechanisms that lead to the improvement and degradation of the main compounds, showing both the advantages and disadvantages of cold plasma technology.

Keywords: cold plasma; sugar; bioactive compounds; vitamins; carotenoids

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1. Introduction

Cold plasma technology has gained much interest from the food science and technology scientific community in the past 10 years. Since the first research on plasma application in food processing, this emerging technology has shown great potential in the food industry.

Early studies with cold plasma focused on the sanitization of several food products [1–4]. These studies proved that cold plasma was a suitable technology for sanitization, and apparently no significant change was noticed in food products' physical or sensory aspects. Until the mid-2010s, many scientists claimed that cold plasma did not induce any change in the food nutritional quality. By the end of the 2010s, cold plasma knowledge evolved. Many studies showed that cold plasma changed the nutritional quality of food products, but these changes were mostly to improve food quality [5–8]. Nowadays, cold plasma can be used to modulate several sensory and nutritional properties of food products [7,9,10].

Several fruits and fruit juices have a short shelf-life, and preservation and sanitization of these products were the scopes of several studies early on. Most studies focused on preserving whole fruits, such as tomatoes, blueberries, strawberries, and others [4,11–13]. Many articles and reviews have published a significant amount of information and knowledge regarding microbial inactivation and shelf-life preservation [14–18].

As more studies with this technology were carried out, changes in the nutritional quality of these plasma-processed fruits were noticed, and it became the focus of new research [5,6,8,19–22]. Chemical changes were noted in sugars and oligosaccharides [5,23,24], starch [25], lipids [22,26], vitamins [6,21,27], phenolics [6,9,21,26,28,29], anthocyanins [11,26,30], terpenoids, and other aroma compounds [7,31] during fruit and fruit juice processing. The chemical changes induced by plasma application were observed not only in fruit processing but also during the processing of other products, such as fish, meat, vegetables, and cereals [32–34].

Most articles correctly attribute the changes in the chemical compounds to the reaction with plasma-reacting species. However, simplistic explanations are given in most cases, attributing the changes to oxidation by reactive oxygen species (ROS) and reactive nitrogen species (RNS). This simple explanation holds true for many chemical changes observed

during plasma application. Still, plasma also induces many more reactions, including activating complex metabolic pathways, especially when fresh produce is treated, which is often the case when processing fruits.

This review addresses the chemical routes and mechanisms induced by cold plasma processing, which causes changes in several chemical compounds present in fruits: sugars, starch, lipids, carotenoids, vitamins, phenolic compounds, and anthocyanins.

2. Fundamentals, Cold Plasma Generation, and Technologies

Plasma is referred to as the fourth state of matter. It is an ionized gas comprising several excited atomic, molecular, ionic, and radical species, co-existing with electrons, positive and negative ions, free radicals, gas atoms, molecules in the ground or excited state, and quanta of electromagnetic radiation (UV photons and visible light).

Plasma is classified as thermal or nonthermal plasma. In thermal plasmas, the ionization and chemical processes are mainly governed by the temperature, which can reach more than 20,000 K. Thermal plasma systems are used for applications requiring enormous heat, such as coating technology, welding, cutting, and treatment of hazardous wastes [35].

In nonthermal plasmas, different temperatures can be achieved for different plasma species, mostly around room temperature. Nonthermal plasma uses energy more efficiently to gain better chemical selectivity. In nonthermal plasmas, the electron temperature governs ionization and chemical processes [35]. Plasma is in a metastable state with a roughly zero net electrical charge [36,37]. Only nonthermal plasma is applied to food products.

Cold plasma can be generated in gases like helium, argonium, oxygen, nitrogen, and a mixture of these. Most studies with fruits and fruits juices have applied air or modified atmospheres. Some early studies have used helium or argonium, but studies with these gases have diminished due to their high cost, which would impact the price of the treated product.

Nonthermal plasma can be formed by electrical, microwave, and radiofrequency power sources that generate a high electrical potential difference between two or more electrodes. Among all technologies used to form cold plasma, dielectric barrier discharge (DBD) plasma, jet plasma, and glow discharge plasma are the most studied technology concerning application with fruit and fruit juices. This review will not focus on the description of these plasma technologies since several good reviews presenting good insights on the different technologies used to generate cold plasma are available [14,38–40].

There is high interest in atmospheric pressure cold plasma technologies in food applications because they do not require vacuum systems and enable continuous material processing [41]. Glow discharge plasma (also called vacuum plasma) has been used in food and materials processing; however, since its application requires a vacuum, this technology cannot be directly applied to volatile materials. Jet plasma has been widely studied, but this technology has been less used due to the small surface areas that can be treated [42].

Among all technologies available, atmospheric dielectric barrier discharge plasma seems adequate to treat fruits and fruit juices rather than vacuum plasma. Several sensory characteristics of fruits come from volatile chemical compounds that may readily volatize when a vacuum is applied, inducing changes in aroma and flavor. On the other hand, vacuum plasma showed increased vitamin C and phenolic content in fruits, improving the nutritional quality of the product. Thus, the best technology depends on the goals established for the product and the main chemical components that characterize the fruit.

3. Plasma Reactive Species

As previously stated, plasma consists of an ionized gas comprising several excited atomic, molecular, ionic, and radical species, co-existing with electrons, positive and negative ions, free radicals, gas atoms, molecules in the ground or excited state, and quanta of electromagnetic radiation (UV photons and visible light). The reactive species generated during plasma application depend on the gas and operating conditions applied.

Helium and argonium plasma generate plasma species that do not react with many bioactive compounds and are mainly used for sanitization [38,43–45]. The first studies with cold plasma on food consisted primarily of applying jet plasma using helium and argonium, which produced reactive species that are mainly inert to the chemical compounds present in food. These first studies with inert gases, and therefore, inert plasma, induced minimal chemical changes on the main food constituents. The results attained by these studies helped to propagate the idea that cold plasma did not alter the food quality while being very efficient in sanitizing it.

As the application of cold plasma migrated to the use of nitrogen, air, and modified atmosphere, several studies began to report slight to significant changes in the chemical composition of fruit and fruit products. Air plasma is a potent source of reactive oxygen species (ROS) and reactive nitrogen species (RNS). These reactive species tend to react when in contact with living cells or with organic compounds. Normal metabolism may be disrupted when plasma reactive species contact living cells or organisms, and several chemical reactions may occur when these species encounter organic molecules.

Plasma reactive species can be in the form of atoms, molecules, or ions with unpaired electrons. Free radicals are very unstable and usually react very fast with other molecules. Knowing how to use these free radicals is extremely important in plasma treatment. Working efficiently with plasma technology allows us to explore several chemical reactions with these non-toxic, free radicals, opening opportunities to improve product quality.

Air plasma free radicals include hydroxyl (HO•), superoxide (O_2•$^-$), alkoxyl (RO•), peroxyl (ROO•), and nitric oxide (NO•). Air plasma nonradical species include ozone (O_3), hydrogen peroxide (H_2O_2), and singlet oxygen ($^1O_2$). The concentration of these species in plasma depends on the plasma technology being used, the operating conditions used to generate plasma, and environmental conditions (such as temperature and relative humidity).

Among the reactive oxygen species (ROS), hydroxyl radical is the most potent oxidant, followed by ozone and alkoxyl radicals [46]. These species react very rapidly with nearby molecules when they are formed. Thus, the generation of high amounts of hydroxyl radical, ozone, and alkoxyl radical may significantly change the fruit and fruit juices. Plasma species such as superoxide, nitric oxide, and lipid hydroperoxides are less reactive, allowing a higher control of the chemical reactions induced by plasma application.

Singlet oxygen ($^1O_2$) is formed when the two unpaired electrons of the molecular oxygen pair up into two different orbitals. This species is very reactive and a powerful oxidant. The species has two electrons that react quickly with unsaturated molecules, such as unsaturated lipids, phenolics, amino acids, or anthocyanins [47].

Superoxide radical (O_2•$^-$) is formed when a single electron reduces oxygen. This species is highly reactive and the precursor of several other plasma species due to its tendency to abstract hydrogen atoms. It reacts mainly with compounds on the product's surface because this species does not permeate through membranes due to its charge [48]. The superoxide radical, therefore, is an important species in sanitizing processes.

Hydrogen peroxide (H_2O_2) has a long half-life and easily diffuses between cells. Thus, this species will continue to induce chemical changes to the product for long periods after plasma treatment. In fruits and minimally processed fruits, hydrogen peroxide is controlled by peroxidase enzymes and may have a lower effect. The decay of hydrogen peroxide produces water or hydroxyl radical, which is one of the most reactive plasma species.

Hydroperoxyl radical (HO_2•) will be formed at a low pH by the protonation of superoxide radicals. This species is more reactive than its precursor. Because of its lack of charge, hydroperoxyl radicals can permeate the membrane phospholipid bilayer and react with bioactive compounds present in this cell region.

Hydroxyl radical (HO•) is one of the most reactive plasma species. The decay of ozone generates it in the presence of water or water vapor, the cleavage of water, and the decay of hydrogen peroxide (H_2O_2). Plasma technologies that generate higher quantities of UV light may produce more hydroxyl radicals due to their capacity to cleave H_2O_2,

yielding two hydroxyl radicals. Fruits have defense systems that control hydroxyl radical concentration, but fruit juices have lower or no ability to control hydroxyl radicals. This species reacts with several molecules, including amino acids and terpenoids [49].

Ozone (O_3) is generated by the bombardment of oxygen molecules by electrons in the plasma discharge [50]. The concentration of ozone in plasma depends on several factors. Still, it is very dependent on the concentration of oxygen in the environment and on the excitation frequency of the plasma system. Higher excitation frequencies (>800 Hz) tend to form higher concentrations of ozone. Ozone has high oxidizing power and reacts with several compounds, including vitamins, phenolics, proteins, lipids, and nucleic acids. Ozone molecules react with water at the product surface or microorganism surface, thus having high sanitization power [51]. The half-live of ozone can be as high as 6 h after plasma application [51].

The reaction of oxygen plasma species with carbon-based compounds, such as lipids, proteins, amino acids, and nucleic acid, generates alkoxy radical (RO•) and peroxyl radical (ROO•) [52]. Both radicals are good oxidizing agents. The peroxyl radical (ROO•) has a long half-life and can diffuse considerably in fruit cells. As such, this radical has a good penetration capability and can induce changes in the interior of large products.

Nitric oxide (NO•) is the main reacting nitrogen species formed in plasma treatment. This species is a natural cellular messenger involved in several physiological processes. Thus, the formation of this species can trigger several physiological responses in fruit and fruit juices. An excess of nitric oxide is cytotoxic and can induce the formation of other nitrogen plasma species, such as peroxynitrite anion ($ONOO^-$) and nitrite anion (NO_2^-). Nitric oxide and its anions react with oils, proteins, sulfhydryls, lipids, and nucleic acids [53].

Nitric oxide also scavenges peroxyl and alkoxyl radicals, reducing the adverse effects of plasma in fruit and fruit juices with high contents of oils and lipids [54,55].

Fruits are constantly exposed to reactive oxygen species produced as by-products of metabolism, respiration, stress, and oxidation. Their antioxidant defenses are responsible for regulating the concentration of reactive oxygen species at acceptable levels to the cells. Oxidative stress occurs when an imbalance between the reactive oxygen species and antioxidant defenses is observed. Plasma application forms high concentrations of reactive oxygen species quickly, resulting in oxidative stress that the fruit antioxidant defense system cannot control. Thus, chemical changes are likely to occur.

In processed fruit-based products, where cells are not alive anymore, the reactive oxygen species will probably cause higher chemical modifications since no defense system will be available to regulate the reacting oxygen species' concentration. However, enzymes may still be viable and activated by some reactive oxygen species in some processed fruit juices, participating in some chemical modifications. This means that the chemical changes induced by plasma treatment in a blueberry may not have the same outcome as the changes induced in blueberry juice, dried blueberry, or blueberry jam. Thus, the analysis of the changes caused by plasma should consider its defense system.

Few studies have been conducted on the characterization and quantification of reactive oxygen species and reactive nitrogen species generated by plasma technologies used in food treatment. The detection of plasma-reacting species is usually done with optical emission spectroscopy. Most labs working with plasma still do not have an optical emission spectrometer, and complete characterization of reactive plasma species is rare and usually not reported in most studies. Thus, the characterization and quantification of reactive plasma species is a knowledge gap that needs to be filled.

Studies show that dielectric barrier discharge plasma applied in air generates reactive nitrogen species and reactive oxygen species, with nitrogen species dominating plasma composition. The presence of oxygen singlet and hydroxyl radical are detected in lesser quantities [56]. The production of ozone in dielectric barrier discharge plasma depends on the excitation frequency of the power source. Low frequencies (<200 Hz) produce low quantities of ozone, whereas higher frequencies (>800 Hz) produce higher amounts of

ozone. In systems where water or moisture are present, DBD plasma tends to increase the generation of OH radicals and restrict ozone production [57].

The plasma environment generated in glow discharge plasma is very different from that of dielectric barrier discharge plasma. Although nitrogen species are still prominent in glow discharge plasma, OH radicals and nitric oxide concentration are much higher than DBD plasma. Due to the high concentration of nitric oxide, air glow discharge plasma efficiently fixates nitrogen in organic materials [58].

The gliding arc plasma environment is characterized by OH radicals, NO, and other nitrogen species, as it is the technology that produces the greatest amount of OH radicals. Despite the generation of OH radicals from the dissociation of water in the air, only small quantities of other hydrogen species are generated. The formation of OH radical is dependent on the electrical current applied. Increasing the current increases the formation of OH radicals and decreases the formation of nitrogen species [59].

Different plasma technologies and operating conditions generate different compositions of reactive plasma species. Thus, the chemical modification induced by plasma will depend on the technology and operating conditions that are applied. In a general analysis, dielectric barrier discharge plasma will tend to cause more oxidation and hydrogenation reactions. Glow discharge plasma can induce hydrolysis, nitrogen fixation, and the formation of alcohols. Gliding arc plasma will induce more reactions with OH radicals. In comparison, jet plasma will be less reactive with most organic compounds because it operates mainly with inert gases.

4. Chemical Transformations

The plasma species formed during cold plasma application will likely induce chemical changes in most fruit and fruit juices. The extent of the changes will depend on several factors, such as the plasma technology used, treatment time, treatment conditions, and environmental conditions.

Cold plasma-reacting species can react with nearly all fruit constituents, such as sugars, starches, amino acids, lipids, vitamins, carotenoids, terpenes, phenolics, anthocyanins, and other bioactive compounds. These changes will affect several characteristics of fruit juices, such as pH, taste, aroma, color, and texture. In this review, we will focus on the chemical changes induced in several classes of compounds.

4.1. Sugars and Oligosaccharides

Plasma treatment can decrease or increase the concentration of sugars in fruit juices. The outcome depends on the kind of plasma technology being used, the sugar profile of the product, and whether the product is fresh, minimally processed, or processed.

Sugars are an energy source in most primary and secondary metabolisms. As such, sugars in fresh and minimally processed fruit products can decrease because they will be consumed to provide energy for the defense mechanism. In this case, sugars are converted to phosphoenolpyruvate and erythrose-4-P by glycolysis and the pentose phosphate pathway, respectively. This was the case of minimally processed apples and apple juice subjected to DBD plasma, where sugar content decreased by up to 46% [24]. A significant reduction was achieved when operating at high excitation frequencies (>400 Hz).

In fresh and minimally processed fruits, plasma application may induce a stress response, with sugars being consumed for the biosynthesis of phenolics. Phosphoenolpyruvate and erythrose-4-P will participate in the shikimate pathway converted into L-phenylalanine, which participates in the phenylpropanoid pathway to generate several phenolic compounds [60]. Thus, the decrease in sugar concentration can increase the concentration of phenolics when this mechanism is activated. The application of DBD plasma to minimally processed apples and apple juice presented this pattern. Total phenolic content increased ($\leq$62%) following a decrease in sucrose, fructose, and glucose ($\leq$57%, $\leq$34%, and $\leq$58%, respectively) [24,61].

An increase in sugar content is not expected after plasma application, but some studies have reported its increase. This increase is usually related to the depolymerization of starch and other oligosaccharides. An increase in fructose and glucose may be associated with the breaking down of sucrose. A slight increase in sugar content can also be caused by the extraction of sugars from the suspended pulp.

Cold plasma treatment induces a decrease in the average chain length of oligosaccharides. Depolymerization of oligosaccharides can occur to various extents, depending on the type of oligosaccharide and operating conditions. Usually, larger molecules are more susceptible to cleavage by plasma than smaller molecules. The decrease in the concentration of oligosaccharides with a higher degree of polymerization can increase the concentration of oligosaccharides with a lower degree of polymerization [5]. Prebiotic oligosaccharide depolymerization usually occurs at α-1,6-glycosidic bonds due to oxidation by ozone formed during plasma application [5,62]. An example of the effect of this mechanism was observed for orange juice enriched with prebiotic oligosaccharides subjected to DBD plasma, where 22% of the prebiotic oligosaccharides were depolymerized at different levels.

Plasma application was shown to be useful in decreasing total sugar content in some juices and allowing DBD plasma technology to produce low-sugar-content fruit juices. Glow plasma technology could also be used to produce low-sugar-content fruit juices, but at the expense of longer exposure times [63]. Table 1 presents the changes induced by cold plasma on the sugars of fruits and fruit juices.

Table 1. Changes in sugars induced by cold plasma application.

Sugar	Technology	Treatment Details and Its Effects on Sugar Content	Reference
Fructose	DBD plasma	Increase of 5% in orange juice treated for 1 min at 50 Hz, 70 kV	[8]
		No significant change in clarified tomato juice treated for 15 min at 50 Hz, 60 kV	[63]
		Decrease of 20% in pitaya treated for 5 min at 50 Hz, 60 kV	[33]
		Decrease of 34% in apple juice treated for 15 min at 900 Hz, 20 kV	[61]
		Decrease of 20% in apple cubes treated for 15 min at 900 Hz, 20 kV	[61]
Fructose	Glow plasma	Increase of 32% in apple juice treated for 10 min at 10 mL/min air flow	[61]
Glucose	DBD plasma	Increase of 14% in orange juice treated for 1 min at 50 Hz, 70 kV	[8]
		No significant change in clarified tomato juice treated for 15 min at 50 Hz, 60 kV	[63]
		Decrease of 21% in pitaya treated for 5 min at 50 Hz, 60 kV	[33]
		Decrease of 58% in apple juice treated for 15 min at 900 Hz, 20 kV	[61]
		Decrease of 25% in apple cubes treated for 15 min at 900 Hz, 20 kV	[61]
Glucose	Glow plasma	Increase of 92% in apple juice treated for 30 min at 10 mL/min air flow	[61]
Sucrose	DBD plasma	Increase of 5% in orange juice treated for 1 min at 50 Hz, 70 kV	[8]
		Decrease of 57% in apple juice treated for 15 min at 900 Hz, 20 kV	[61]
		Decrease of 52% in apple cubes treated for 15 min at 200 Hz, 20 kV	[61]
Sucrose	Glow plasma	Decrease of 51% in apple juice treated for 30 min at 30 mL/min air flow	[61]
Prebiotic Oligosaccharides	DBD plasma	Decrease of 22% in prebiotic orange juice treated for 1 min at 50 Hz, 70 kV. Decrease in DP6, DP7, and DP8, and increase in DP3	[5]

DP—degree of polymerization.

4.2. Starch

Starch consists of amylose and amylopectin molecules, which are assembled in granular form. Cold plasma processing changes in starch's chemical, physical, and mechanical properties depend on the type of starch, the plasma technology, and the operating conditions. Operating conditions such as voltage, pressure, and gas type induce more significant changes in starch properties.

Cold plasma treatment reduces the amylose content and increases the amylopectin content due to the formation of branches in the linear starch structure. An example of the branched structure formation was observed for aria starch, where the application of DBD plasma increased the content of amylopectin from 62 to 77%. The mass fraction of amylopectin increased with an increasing electrical potential difference between the plasma equipment electrodes [25].

Reactive oxygen species can oxidize starch, forming carbonyl groups. The increase in carbonyl group content reduces starch pH due to increased acidic functional groups [64].

Cold plasma reduces the moisture content of starch due to the interactions of plasma-reactive species and the water surrounding starch molecules. The interactions break the bond between water and starch, facilitating water removal during processing [65].

Plasma treatment may induce several changes in the molecular structure of starch. These changes include depolymerization, cross-linking, the formation of functional groups, and grafting [65].

Depolymerization of starch can occur to various extents, depending on the type of starch and operating conditions. Similar to the effects of plasma on oligosaccharides, larger starch molecules are more susceptible to plasma than smaller molecules. Large starch molecules have more water trapped in the starch structure, facilitating chemical reactions and depolymerizing starch [66].

Cross-linking of starch molecules can occur, increasing the molecular weight, which can be correlated to the increase in the contents of C-O-C bonding and the decrease in OH groups. Cross-linking has been reported after applying glow discharge plasma using argon and helium [67]. Extended exposure to plasma treatment depolymerizes newly formed cross-links and leads to a net reduction in molecular weight. Thus, processing time should be optimized when cross-linking starch is desired.

Grafting reactions may occur between starch and plasma species. Ethylene can be grafted in some starches using low-pressure glow discharge plasma [68].

Plasma treatment has little to no effect on the polymorph type of starch [65,67–70]. However, plasma may reduce the crystallinity of some starches due to molecular scission and granular corrosion [70]. The lamella structure of starch can be affected by plasma application, with the semi-crystalline lamellae showing an increase in repeating distance [67,71]. Crystalline lamellae are more prone to changes than amorphous lamellae. The higher water content trapped inside the crystalline region leads to more chemical changes with the plasma-reacting species. These changes depend on the type of starch and the operating conditions applied during plasma treatment. Helium plasma induces more changes in starch crystallinity than oxygen plasma.

Plasma induces the formation of fissures, cavities, corrosions, and deposits on the surface of starch granules [65]. Corona discharge plasma causes surface roughness or even extended surface damage due to electrical current passing through the granules. Extensive damage may occur by increasing the plasma intensity [72].

4.3. Ascorbic Acid (Vitamin C)

Ascorbic acid does not degrade significantly during plasma application, and only minimal losses have been reported. On the other hand, some studies have reported a significant increase in vitamin C in fresh fruits and fruit juices [9,28].

The decay mechanism of ascorbic acid occurs by deprotonation of the molecule forming ascorbate—ascorbate radical—ending in the formation of dehydroascorbate (Figure 1). The deprotonation of ascorbic acid can occur by reaction with hydrogen, superoxide, hydrogen peroxide, or tocopheroxyl radical.

In living cells, dehydroascorbate is regenerated by the dehydroascorbate reductase enzyme, forming ascorbic acid. Plasma technology that induces the hydrogenation of molecules can chemically transform dehydroascorbate into ascorbic acid, reversing the decay mechanism and increasing vitamin C content even in products not containing living cells.

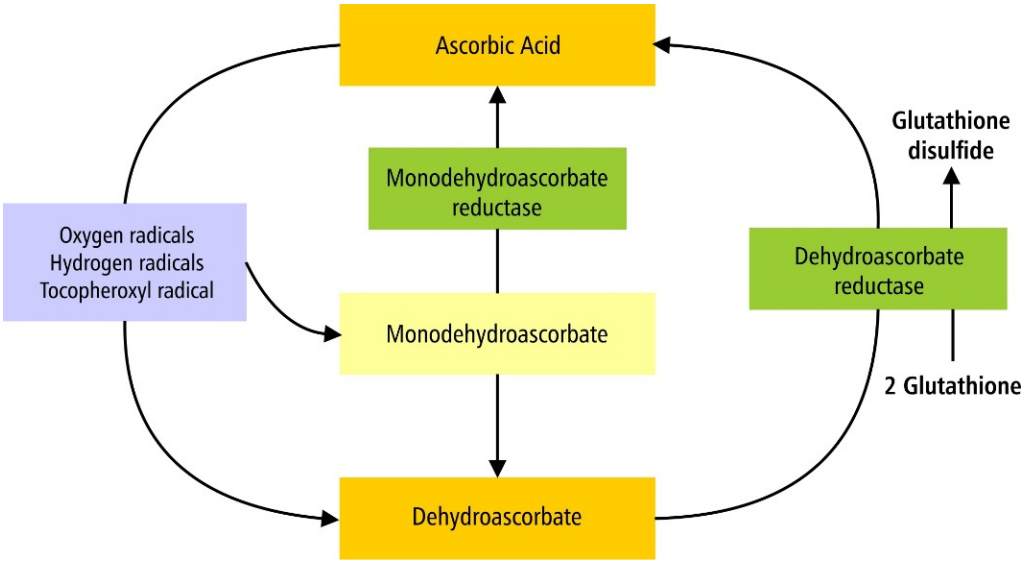


Figure 1. Ascorbic acid decay and regeneration cycle.

Ascorbic acid content can decrease due to reaction with ozone, either by direct reaction or by the Criegee mechanism [73]. Thus, operating conditions that produce large ozone quantities are not recommended for vitamin C-rich fruit products. A reduction in ascorbic acid can occur during storage due to the food matrix's accumulation of ozone and other long half-life reactive plasma species.

Table 2 presents the changes induced by cold plasma treatment on fruits and fruit juices. A significant increase in vitamin C was observed after DBD and glow plasma application on camu-camu juice and blueberries [9,28,31]. The positive effect of plasma application on vitamin C is mainly dependent on voltage and time of exposure to plasma. Larger voltages (80 kV) may increase vitamin C content but only when applied for a short period (<5 min). Reducing the applied voltage (20 to 25 kV) seems to lead to a higher vitamin C increase even after long exposure periods (15 to 30 min).

Table 2. Changes in ascorbic acid induced by cold plasma application.

Product	Technology	Treatment Details and its Effects on the Ascorbic Acid Content	Reference
Fruits	DBD plasma	Minimal loss in strawberry, blueberry, cherry tomato Increase of 5% in kiwis treated for 40 min at 15 kV. Slight reduction (7%) after 4 days of storage	[74] [75]
Fruits	In-package DBD plasma	Increase of 57% in blueberries treated for 1 min at 50 Hz, 80 kV; followed by 16% reduction when treated for 5 min	[28]
Juices	DBD plasma	Increase of 40% in camu-camu juice treated for 15 min at 960 Hz, 24 kV Decrease of 5% in clarified tomato juice treated for 15 min at 50 Hz, 60 kV	[31] [64]
Juices	Gliding arc plasma	Decrease of 4% in tomato juice treated for 5 min at 50 Hz, 3.8 kV, 40 W	[4]
Juices	Glow discharge plasma	Increase of 18% in camu-camu juice treated for 20 min at 50 kHz, 80 W, 30 mL/min air plasma Decrease of 10% in seriguela juice treated for 10 min at 50 kHz, 80 W, 30 mL/min nitrogen plasma	[9] [76]
Whey beverage	Glow discharge plasma	Increase of 120% in guava flavored whey beverage treated for 10 min at 50 kHz, 80 W, 10 mL/min air plasma	[77]

4.4. Other Vitamins

Reports on cold plasma effects on vitamins A, B, D, and K are scarce, and the mechanisms are still unknown. Glow discharge plasma application increased the contents of pro-vitamin A at low fluences (time x plasma flow). However, decay of this pro-vitamin

was observed at higher fluences. An increase in vitamin B3 and B6 has been reported, but no decay was observed at higher fluences [76]. Table 3 presents the main findings concerning vitamins A and B.

Table 3. Changes in vitamins A, B3, and B6 induced by cold plasma application.

Vitamin	Product	Technology	Treatment Details and Its Effects on Vitamin Content	Reference
A	Juices	Glow plasma	Increase of 49% in seriguela juice treated for 10 min at 50 kHz, 80 W, 10 mL/min of nitrogen plasma. Decrease of 9% when treated for 10 min at 80 kV, 30 mL/min of nitrogen plasma.	[76]
B3	Juices	Glow plasma	Increase of 19% in seriguela juice treated for 10 min at 50 kHz, 80 W, 10 mL/min of nitrogen plasma.	[76]
B6	Juices	Glow plasma	Increase of 56% in seriguela juice treated for 10 min at 50 kHz, 80 W, 20 mL/min of nitrogen plasma.	[76]

4.5. Phenolics

Cold plasma can induce both an increase and a decrease in phenolics. Four phenomena can change the phenolic content in food products. Phenolics can increase due to the extraction of phenolics from the cell membrane or by leaking cytoplasm due to cell breakdown. Phenolics can also increase due to the depolymerization of tannins. In plants, phenolics can increase due to the activation of cell defense mechanisms when exposed to plasma-reactive species. On the other hand, phenolics can decrease due to oxidation.

The extraction of phenolics from fruits occurs due to changes in the cell membrane, either by chemical modification or physical modification. Reaction with ROS and RNS may damage the cell membrane with the consequent extraction or leakage of phenolic compounds to the outer cell environment. Furthermore, ROS, RNS, and UV radiation induce chemical changes that release phenolic compounds bonded to the cell membrane.

UV radiation and ROS can act as abiotic elicitors participating in the regulation of stress responses in plants. UV radiation stimulates hormesis and induces beneficial reactions in fruit tissue, such as accumulating secondary metabolites, including phenolic compounds [34,78,79]. Reacting oxygen species are signaling molecules that trigger exogenous ROS regulators that mediate phenolic content in stress responses, contributing to an increase in phenolic content [59]. The biosynthesis of phenolics resulting from stress response occurs through the phenylpropanoid pathway (Figure 2).

The biosynthesis of phenolics in this pathway is done mainly by the enzymes PAL, C4H, and 4CL. Cold plasma can increase the expression of these enzymes, increasing the production of phenolic compounds. In the phenylpropanoid pathway, PAL biosynthesizes cinnamic acid from phenylalanine, C4H biosynthesizes *p*-coumaric acid from cinnamic acid, and 4CL biosynthesizes cinnamoyl-CoA also from cinnamic acid [80]. Phenolics such as caffeic acid, protocatechuic acid, *p*-hydroxybenzoic acid, and gallic acid are biosynthesized from *p*-coumaroyl CoA in the phenylpropanoid pathway (Figure 3) [81]. Plasma processing of cashew apple juice by DBD plasma activated the phenylpropanoid pathway, reducing glucose, fructose, and phenylalanine, and increasing the total phenolics [20,21].

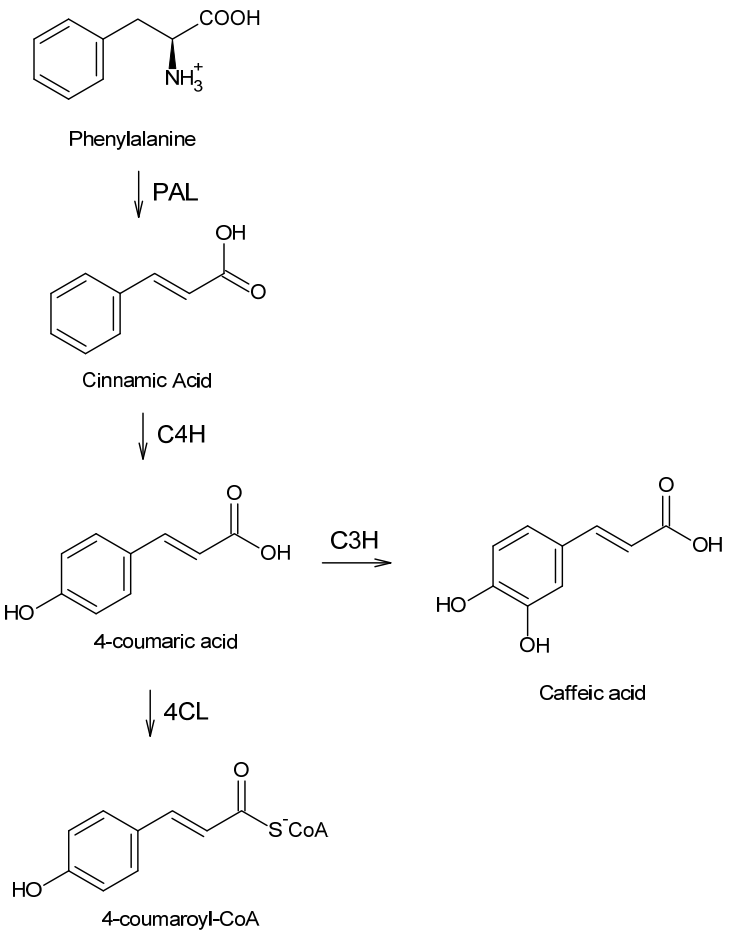


Figure 2. Phenylpropanoid pathway [58].

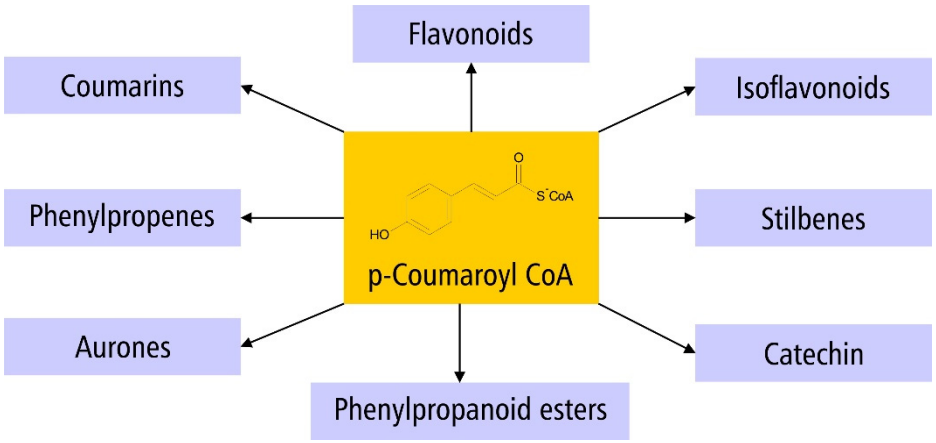


Figure 3. Compounds generated from *p*-Coumaroyl CoA in the phenylpropanoid pathway [81].

The application of high voltages (>60 kV) may generate higher concentrations of reactive oxygen species and ozone, which reacts with the phenolic aromatic rings, decreasing the content of phenolic compounds by oxidation.

Flavan-3-ols (catechin, epicatechin, procyanidin) tend to oxidize when subjected to DBD plasma. The oxidation process increases with processing time. Hydroxycinnamic acids are not much affected when subjected to DBD or jet plasma. They are less effective reducing agents than other phenolic compounds and react slowly with plasma-generated radicals [82].

Dihydrochalcones (phloretin, phloridzin) and flavonols tend to increase when subjected to short-time DBD processing (up to 30 min) and then decrease after extended exposure to plasma (60 min).

A decrease in gallic acid may occur due to cleavage of its aromatic ring due to the formation of phenoxyl radicals during electron/proton transfer, leading to the formation of small phenolic acids [83,84].

Ellagitannins can be hydrolyzed, releasing gallic acid (Figure 4) [85]. Reacting plasma species have sufficient electrical energy to break covalent bonds and improve the depolymerization of ellagitannins [86].

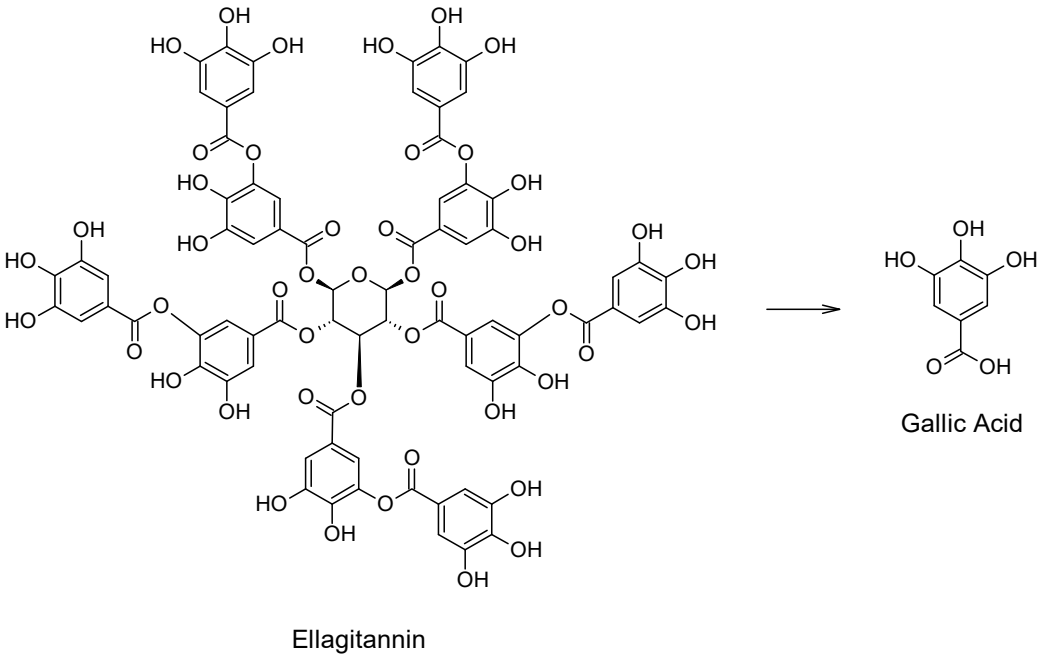


Figure 4. Depolymerization of ellagitannin [86].

Reacting plasma species have sufficient electrical energy to break covalent bonds, inducing several chemical reactions [86]. These chemical reactions can include hydrogenation, dehydrogenation, hydrolysis, or dehydration. The main reaction route depends on the plasma technology and the operating conditions that are applied. DBD plasma tends to hydrogenate and dehydrate molecules in many applications, whereas glow plasma tends to hydrolyze molecules. For example, an increase in flavonols can be related to incorporating hydroxyl radicals into the aromatic rings of phenolic compounds.

Table 4 presents a list of phenolic compounds and the changes observed after plasma treatment. The application of jet plasma and glow discharge plasma presented promising results in increasing the contents of most phenolic compounds. The application of DBD plasma to fruits and fruit juices showed mixed results, some positive and some negative. DBD plasma operating at high voltage (60 kV) and low frequency (50 Hz) tended to increase hydroxybenzoic acids.

Table 4. Changes in phenolics induced by cold plasma application.

Phenolic	Technology	Treatment Details and Its Effects on Phenolic Content	Reference
4-coumaroylquinic acid	DBD plasma	Decrease of 15% in apples treated for 30 min at 13 kHz	[75]
4-hydroxy benzaldehyde	DBD plasma	Increase of 5% in strawberry treated for 15 min at 50 Hz, 60 kV	[83]
Caffeic acid	DBD plasma	No significant change in apples treated for 30 min at 13 kHz Decrease of 17% in pitaya treated for 5 min at 50 Hz, 60 kV	[75] [33]
Caffeic acid	Jet plasma	Increase of 48% in sour cherry juice treated for 3 min at 25 kHz, 4 W, 1.25 L/min argon flow rate No significant change in chokeberry juice treated for 5 min at 25,000 Hz, 4 W, 0.75 L/min argon flow rate Decrease of 43% in pomegranate juice treated for 3 min in argon plasma at 25 kHz, 1.0 L/min	[30] [87] [86]
Caffeoylquinic acid	DBD plasma	No significant change in apples treated for 30 min at 13 kHz	[75]
Catechin	DBD plasma	Decrease of 20% in apples treated for 30 min at 13 kHz	[75]
Catechin	Jet plasma	Increase of 88% in pomegranate juice treated for 5 min in argon plasma at 25 kHz, 1.2 L/min	[86]
Chlorogenic acid	DBD plasma	Increase of 46% in strawberry treated for 15 min at 50 Hz, 60 kV Increase of 31% in clarified tomato juice treated for 15 min at 50 Hz, 60 kV	[83] [64]
Chlorogenic acid	Jet plasma	Increase of 138% in pomegranate juice treated for 3 min in argon plasma at 25 kHz, 1.2 L/min No significant change in chokeberry juice treated for 5 min at 25,000 Hz, 4 W, 0.75 L/min argon flow rate	[86] [87]
Coumaroylquinic acid	DBD plasma	Increase of 4% in apples treated for 30 min at 13 kHz	[75]
Ellagic acid	Jet plasma	Increase of 245% in pomegranate juice treated for 7 min in argon plasma at 25 kHz, 1.2 L/min	[86]
Epicatechin	DBD plasma	Decrease of 17% in apples treated for 30 min at 13 kHz	[75]
Ferulic acid	Jet plasma	Increase of 35% in pomegranate juice treated for 3 min in argon plasma at 25 kHz, 1.2 L/min	[86]
Gallic acid	DBD plasma	No significant change in strawberry treated for 15 min at 50 Hz, 60 kV Increase of 51% in clarified tomato juice treated for 15 min at 50 Hz, 60 kV Increase of 107% in pitaya treated for 5 min at 50 Hz, 60 kV	[83] [64] [33]
Gallic acid	Jet plasma	Decrease of 25% in pomegranate juice treated for 3 min in argon plasma at 25 kHz, 0.7 L/min	[86]
Hyprin	DBD plasma	Increase of 14% in strawberry treated for 30 min at 50 Hz, 60 kV	[83]
Myricetin rhamnoside	DBD plasma	No significant change in apples treated for 30 min at 13 kHz	[75]
Neochlorogenic acid	Jet plasma	Increase of 4% in chokeberry juice treated for 5 min at 25,000 Hz, 4 W, 0.75 L/min argon flow rate Increase of 50% in sour cherry juice treated for 3 min at 25 kHz, 4 W, 0.75 L/min argon flow rate	[87] [30]

Table 4. *Cont.*

Phenolic	Technology	Treatment Details and Its Effects on Phenolic Content	Reference
p-coumaric acid	DBD plasma	Increase of 109% in pitaya treated for 5 min at 50 Hz, 60 kV	[33]
p-coumaric acid	Jet plasma	Increase of 91% in sour cherry juice treated for 3 min at 25 kHz, 4 W, 0.75 L/min argon flow rate Decrease of 25% in pomegranate juice treated for 3 min in argon plasma at 25 kHz, 0.7 L/min	[30] [86]
p-hydroxybenzoic acid	DBD plasma	Decrease of 7% in pitaya treated for 5 min at 50 Hz, 60 kV	[33]
Phloretin	DBD plasma	Increase of 25% in strawberry treated for 15 min at 50 Hz, 60 kV	[83]
Phloretin-2-O-(2″-O-xyosyl) glucoside	DBD plasma	Increase of 36% in apples treated for 30 min at 13 kHz	[75]
Phloridzin	DBD plasma	Increase of 127% in apples treated for 30 min at 13 kHz	[75]
Procyanidin dimer	DBD plasma	Decrease of 18% in apples treated for 30 min at 13 kHz	[75]
Procyanidin trimer	DBD plasma	Decrease of 25% in apples treated for 30 min at 13 kHz	[75]
Protocatechuic acid	DBD plasma	Increase of 132% in pitaya treated for 5 min at 50 Hz, 60 kV	[33]
Protocatechuic acid	Jet plasma	Decrease of 54% in pomegranate juice treated for 7 min in argon plasma at 25 kHz, 1.0 L/min	[86]
Punicalagin 1	Jet plasma	Increase of 36% in pomegranate juice treated for 5 min in argon plasma at 25 kHz, 1.2 L/min	[86]
Punicalagin 2	Jet plasma	Decrease of 77% in pomegranate juice treated for 7 min in argon plasma at 25 kHz, 1.2 L/min	[86]
Quercetin	DBD plasma	Increase of 28% in apples treated for 30 min at 13 kHz	[75]
Quercetin-3-galactoside	Jet plasma	Decrease of 15% in chokeberry juice treated for 5 min at 25,000 Hz, 4 W, 0.75 L/min argon flow rate	[87]
Quercetin-3-glucoside	Jet plasma	No significant change in chokeberry juice treated for 5 min at 25,000 Hz, 4 W, 0.75 L/min argon flow rate	[87]
Quercetin-3-vicianoside	Jet plasma	No significant change in chokeberry juice treated for 5 min at 25,000 Hz, 4 W, 0.75 L/min argon flow rate	[87]
Quercetin-3-rutinoside	Jet plasma	No significant change in chokeberry juice treated for 5 min at 25,000 Hz, 4 W, 0.75 L/min argon flow rate	[87]
Quercetin-O-glucoside	DBD plasma	No significant change in apples treated for 30 min at 13 kHz	[75]
Quercetin-O-rhamnoside	DBD plasma	Increase of 23% in apples treated for 30 min at 13 kHz	[75]
Rutin	DBD plasma	Increase of 2% in strawberry treated for 15 min at 50 Hz, 60 kV Decrease of 61% in apples treated for 30 min at 13 kHz	[83] [75]
Sinapic acid	DBD plasma	Increase of 48% in clarified tomato juice treated for 10 min at 50 Hz, 60 kV	[64]
Vanillin	DBD plasma	Increase of 4% in strawberry treated for 15 min at 50 Hz, 60 kV	[83]

Table 4. *Cont.*

Phenolic	Technology	Treatment Details and Its Effects on Phenolic Content	Reference
Total phenolics	DBD plasma	Increase of 5% in kiwi treated for 40 min at 15 kV	[75]
		Increase of 4% in clarified tomato juice treated for 10 min at 50 Hz, 60 kV; followed by a decrease by 4% when treated for 15 min	[63]
		No significant change in orange juice treated for 1 min at 50 Hz, 70 kV	[5]
		Decrease of 3% in apples treated for 30 min at 13 kHz	[75]
		Decrease of 38% in white grape juice treated for 5 min at 80 kV	[84]
Total phenolics	In-package DBD plasma	Increase of 10% in blueberries treated for 1 min at 50 Hz, 80 kV; followed by a decrease by 35% when treated for 5 min	[28]
Total phenolics	Glow discharge plasma	Increase of 75% in camu-camu juice treated for 20 min at 50 kHz, 80 W, 10 mL/min air plasma	[9]
		Increase of 58% in seriguela juice treated for 15 min at 50 kHz, 80 W, 20 mL/min nitrogen plasma	[76]
Total phenolics	Jet plasma	Increase of 49% in pomegranate juice treated for 5 min in argon plasma at 25 kHz, 1.0 L/min	[86]
		Increase of 74% in sour cherry juice treated for 3 min at 25 kHz, 4 W, 1.25 L/min argon flow rate	[30]
Total flavonoids	DBD plasma	Decrease of 20% in white grape juice treated for 5 min at 80 kV	[84]
Total flavonoids	In-package DBD plasma	Increase of 10% in blueberries treated for 1 min at 50 Hz, 60 kV; decrease of 27% when treated for 5 min at 50 Hz, 80 kV	[28]
Total flavonols	DBD plasma	Increase of 65% in white grape juice treated for 5 min at 80 kV	[84]

Jet, DBD, and glow discharge plasmas presented a net increase in total phenolics, with some variations on individual phenolic compound effects. All these plasma technologies can improve the total phenolic content in fruits and fruit juices, with a consequent impact on the antioxidant activity and health benefits. It is still unclear which operating conditions are most recommended, and more studies in this respect are still needed.

4.6. Anthocyanins

The concentration of anthocyanins can either increase or decrease. An increase in content is related to the extraction of anthocyanins from the vacuoles to the extracellular space. A decrease in content is related to oxidative degradation.

Anthocyanins are located in the cell vacuoles in most plants. Plasma-reacting species can break down cell and vacuole membranes, releasing these compounds to the extracellular space, where analytical instruments will more easily detect them. The increased anthocyanin content reported in the literature is mainly caused by better detection of anthocyanins rather than an actual net increase in concentration. This is probably what occurred with the application of jet plasma on sour cherry and pomegranate juices [30,88].

Plasma treatment with argon and oxygen mixture produces a mixture of Ar^+, •OH, $•O(^3P)$, and $•O_2$, which increases the breakdown of the fruit epidermis, releasing bioactive compounds from the vacuole and cytoplasm to the extracellular space [89].

Anthocyanins can undergo oxidative degradation during plasma treatment, reducing their content. Anthocyanins can also interact with other plasma-generated species, resulting in further degradation.

Although anthocyanins are susceptible to plasma-reacting species, the glycolyzed anthocyanins are less susceptible to degradation than the aglycon molecule (anthocyanidin) [90].

Many anthocyanins have intense color due to their chemical structure, which contains chromophores, which are chemical groups that absorbs light and give rise to color. Ozone

and hydroxyl radicals can cause oxidative cleavage of chromophores, resulting in the decay of anthocyanins and loss of color of the product [28,91]. Figure 5 presents the degradation of cyanidin-3-O-glucoside. Other anthocyanins show a similar degradation pattern.

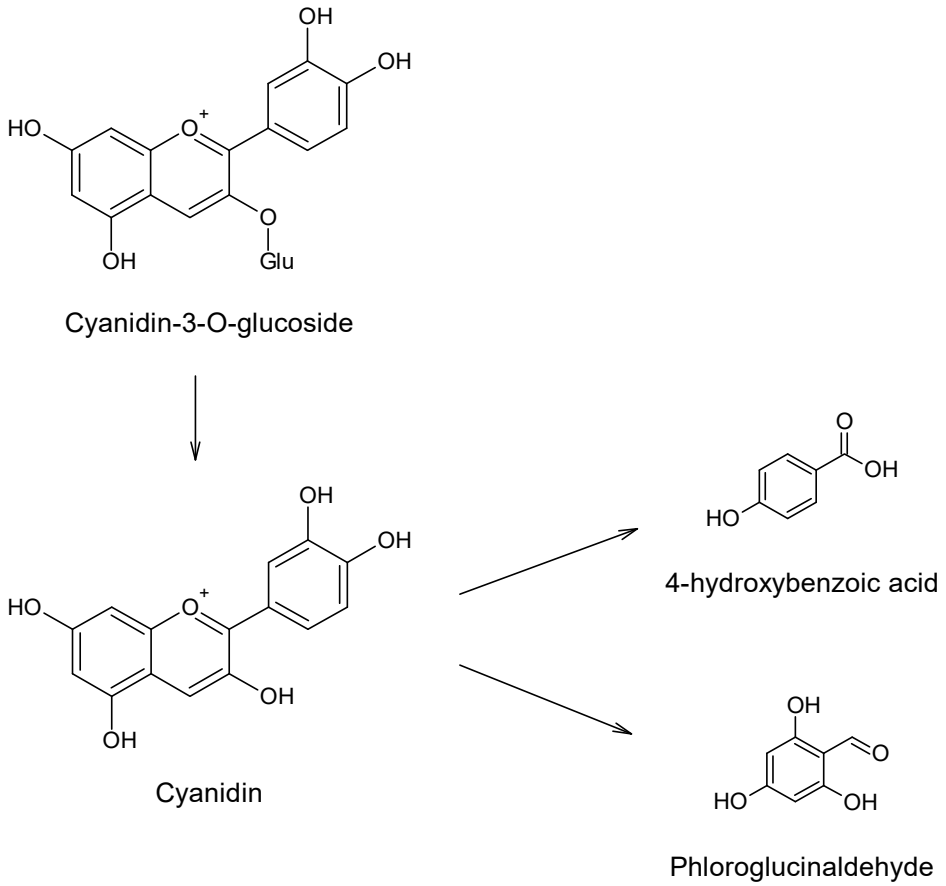


Figure 5. Degradation mechanism of cyanidin-3-O-glucoside subjected to plasma treatment [92].

Plasma systems that generate visible and UV-radiation may induce photodegradation of anthocyanins with their conversion into chalcones, which further decompose into benzaldehyde and benzoic acid [92].

The decay of anthocyanins can happen during storage when the fruit or fruit-based product is treated under conditions that generate more significant amounts of ozone.

Table 5 presents a list of anthocyanins and the effects of plasma treatment during application. The improvement of anthocyanins by plasma technology showed to be largely dependent on processing time. Short treatments (<10 min) improved the number of anthocyanins available in fruits and fruit juices. However, prolonged exposure to plasma (>20 min) significantly decreased the number of anthocyanins, most probably due to the degradation of these compounds.

Table 5. Changes in anthocyanins induced by cold plasma application.

Anthocyanin	Technology	Treatment Details and Its Effects on Anthocyanin Content	Reference
Cyanidin-3-arabinoside	Jet plasma	Decrease of 25% in chokeberry juice treated for 5 min at 25,000 Hz, 4 W, 0.75 L/min argon flow rate	[87]
Cyanidin-3-galactoside	Jet plasma	Decrease of 23% in chokeberry juice treated for 5 min at 25,000 Hz, 4 W, 0.75 L/min argon flow rate	[87]
Cyanidin-3-glucoside	Jet plasma	Increase of 52% in pomegranate juice treated for 5 min at 25 kHz, 1.0 L/min argon flow rate	[88]
		Increase of 41% in sour cherry juice treated for 3 min at 25 kHz, 4 W, 1.0 L/min argon flow rate	[30]
		Decrease of 20% in chokeberry juice treated for 5 min at 25,000 Hz, 4 W, 0.75 L/min argon flow rate	[87]
Cyanidin-3-glucosylrutinoside	Jet plasma	Increase of 37% in sour cherry juice treated for 3 min at 25 kHz, 4 W, 1.0 L/min argon flow rate	[30]
Cyanidin-3-rutinoside	Jet plasma	Increase of 37% in sour cherry juice treated for 3 min at 25 kHz, 4 W, 1.0 L/min argon flow rate	[30]
Cyanidin-3-sophoroside	Jet plasma	Increase of 38% in sour cherry juice treated for 3 min at 25 kHz, 4 W, 1.0 L/min argon flow rate	[30]
Cyanidin-3-xyloside	Jet plasma	Decrease of 23% in chokeberry juice treated for 5 min at 25,000 Hz, 4 W, 0.75 L/min argon flow rate	[87]
Cyanidin-3,5-diglucoside	Jet plasma	Increase of 60% in pomegranate juice treated for 3 min at 25 kHz, 1.0 L/min argon flow rate	[88]
Delphinidin-3-glucoside	Jet plasma	Increase of 84% in pomegranate juice treated for 3 min at 25 kHz, 0.7 L/min argon flow rate	[88]
Delphinidin-3,5-diglucoside	Jet plasma	Increase of 61% in pomegranate juice treated for 3 min at 25 kHz, 1.0 L/min argon flow rate	[88]
Pelargonidin-3,5-diglucoside	Jet plasma	Increase of 171% in pomegranate juice treated for 3 min at 25 kHz, 0.7 L/min argon flow rate	[88]
Total anthocyanin	DBD plasma	Decrease of 37% in açai juice treated for 10 min at 50 Hz, 20 kV	[26]
Total anthocyanin	Glow discharge plasma	Increase of 20% in camu-camu juice treated for 10 min at 50 kHz, 80 W, 10 mL/min air plasma	[9]
Total anthocyanin	In-package DBD	Decrease of 40% in blueberries treated for 5 min at 50 Hz, 80 kV	[28]
Total anthocyanin	Jet plasma	Increase of 57% in pomegranate juice treated for 3 min at 25 kHz, 1.0 L/min argon flow rate	[88]
		Increase of 37% in sour cherry juice treated for 3 min at 25 kHz, 4 W, 1.0 L/min argon flow rate	[30]

4.7. Carotenoids

As occurs with most bioactive compounds subjected to plasma, carotenoids can increase or decrease with treatment. The mechanisms for carotenoid increase and decay are still unknown.

Glow discharge plasma was shown to increase the contents of carotenoids at low fluences (time × plasma flow). However, decay of carotenoids occurred at extended processing times.

Table 6 presents a list of carotenoids and the effects of plasma treatment during application.

Table 6. Changes in carotenoids content induced by cold plasma application.

Compound	Product	Treatment	Treatment Details and Its Effects on Carotenoids Content	Reference
β-carotene	Juices	Glow discharge plasma	Increase of 51% in seriguela juice treated for 10 min at 50 kHz, 80 W, 10 mL/min of nitrogen plasma	[76]
Lycopene	Juices	Gliding arc plasma	Increase of 5% in tomato juice treated for 2 min at 50 Hz, 3.8 kV, 40 W	[4]
Lycopene	Juices	Glow discharge plasma	Increase of 57% in seriguela juice treated for 10 min at 50 kHz, 80 W, 10 mL/min of nitrogen plasma	[76]
Total carotenoids	Fruits	DBD plasma	Decrease of 5% in kiwi treated for 40 min at 15 kV	[75]
Total carotenoids	Juices	Glow discharge plasma	Increase of 32% in seriguela juice treated for 10 min at 50 kHz, 80 W, 10 mL/min of nitrogen plasma	[76]
Total carotenoids	Juices	Gliding arc plasma	Increase of 12% in tomato juice treated for 5 min at 50 Hz, 3.8 kV, 40 W	[4]

4.8. Antioxidant Capacity

The antioxidant capacity is related to the sum of the individual antioxidant capacity of several bioactive compounds. Thus, an increase in total phenolic content or other bioactive chemical groups does not necessarily increase the antioxidant capacity. If the concentration of compounds with high antioxidant capacity decreases and the compounds with low antioxidant capacity increase, the net antioxidant capacity may eventually lead to a higher net concentration of bioactive compounds but a lower net concentration of antioxidant capacity.

Degradation of antioxidant compounds occurs due to oxidation, exposure to light, and enzymatic reactions. Most enzymatic degradation occurs due to ascorbate oxidase, polyphenol oxidase, cytochrome oxidase, and peroxidase.

Plasma application may increase the antioxidant capacity up to a certain processing time, and then overexposure to plasma species and their free radicals may decrease the antioxidant capacity of that product (Figure 6).

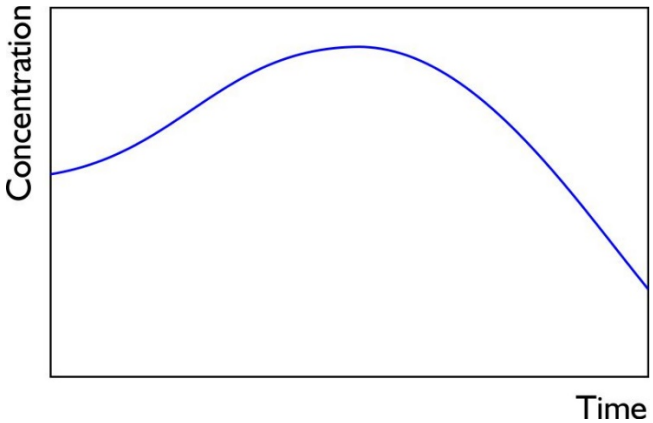


Figure 6. Typical theoretical profile of the concentration of bioactive compounds subjected to plasma processing showing the theoretical effect of overexposure to plasma.

Table 7 presents the changes in antioxidant capacity observed after plasma treatment. Glow plasma technology is probably the best plasma technology to improve the antioxidant content of fruits and fruit juices, mainly due to its ability to increase the amount of total phenolics in these products. The application of DBD plasma showed mixed positive and

negative results. The increase in antioxidant capacity was higher when applying glow discharge plasma than DBD plasma (when positive results were attained with this technology).

Table 7. Changes in antioxidant capacity induced by cold plasma application.

Method	Technology	Treatment Details and Its Effects on the Antioxidant Capacity	Reference
ABTS	DBD plasma	Increase of 4% in kiwi treated for 40 min at 15 kV. Decrease of 8% after 4 days of storage	[75]
		Decrease of 20% in apples treated for 30 min at 13 kHz	[75]
		Decrease of 48% in orange juice treated for 1 min at 50 Hz, 70 kV	[5]
ABTS	Glow discharge plasma	Increase of 120% in camu-camu juice treated for 20 min at 50 kHz, 80 W, 30 mL/min air plasma	[9]
		Increase of 23% in seriguela juice treated for 15 min at 50 kHz, 80 W, 20 mL/min nitrogen plasma	[76]
DPPH	DBD plasma	Increase of 14% in strawberry treated for 10 min at 50 Hz, 60 kV; decrease of 7% when treated for 30 min	[83]
		No significant change in orange juice treated for 1 min at 50 Hz, 70 kV	[5]
		Decrease of 2% in apples treated for 30 min at 13 kHz	[75]
		Decrease of 12% in white grape juice treated for 5 min at 80 kV	[84]
		Decrease of 2% in kiwi treated for 40 min at 15 kV	[75]
DPPH	Glow discharge plasma	Increase of 25% in camu-camu juice treated for 20 min at 50 kHz, 80 W, 30 mL/min air plasma	[9]
		Increase of 66% in seriguela juice treated for 15 min at 50 kHz, 80 W, 10 mL/min nitrogen plasma	[76]
FRAP	DBD plasma	Decrease of 4% in apples treated for 30 min at 13 kHz	[75]
		Decrease of 9% in kiwi treated for 40 min at 15 kV	[75]
FRAP	Glow discharge plasma	Increase of 30% in camu-camu juice treated for 20 min at 50 kHz, 80 W, 10 mL/min air plasma	[9]
TEAC	DBD plasma	Decrease of 11% in white grape juice treated for 5 min at 80 kV	[84]

4.9. Lipids

Cold plasma induces lipid oxidation, mainly when reactive oxygen species are formed in plasma. Lipid oxidation adversely affects the sensory and nutritional quality of foods. This is a significant problem in beef and fish subjected to plasma treatment, but fruits and fruit juices are less prone to significant changes in lipids.

Plasma treatment induces the transformation of unsaturated lipids into saturated lipids, increasing palmitic and stearic acids and decreasing palmitoleic, oleic, and linoleic acids. Polyunsaturated lipids are more prone to peroxidation than monounsaturated lipids. The changes in lipids are caused by hydroperoxyl radicals, superoxide radicals, and singlet oxygen that reacts with unsaturated fatty acids. Contact with light and the absence of active defense mechanisms increase lipid peroxidation.

The hydroperoxyl radical ($HO_2\bullet$) can permeate into the membrane phospholipid bilayer of cells. This radical can abstract hydrogen from the bis-allylic position of phospholipid polyunsaturated acyl radical inside the membrane, initiating lipid peroxidation. Hydroperoxyl radical reacts with lipid hydroperoxides, generating peroxyl radicals and hydrogen peroxide [93]. Hydrogen peroxide does not react significantly with lipids but attacks other bioactive compounds. Ozone reacts with unsaturated lipids, forming aliphatic radicals and carbonyl compounds. Ozone can form several free radicals that can induce lipid peroxidation [48].

Propagation of lipid peroxidation occurs through the peroxyl and alkoxy radicals that are produced in the process. Polyunsaturated lipids have more than one bis-allylic position and can form more than one lipid free radical, increasing the concentration of this radical

considerably. Termination occurs when the lipid radicals (L•, LO•, LOO•) react, forming a nonradical product. Several compounds can be created by termination of lipid radicals due to the structural diversity of lipid molecules.

Most fruits are poor in lipids, so lipid oxidation is not of great concern. There are very few studies with lipid-rich fruits, such as avocados; thus, there is not much information on how plasma affects these kinds of fruits.

5. Differences between Fruit and Fruit Juice Processing

Significant differences in chemical changes are observed when plasma treatment is applied to fruits, minimally processed fruits, and fruit juices. Overall, the changes induced in minimally processed fruits and whole fruits are less significant than those in fruit juices because of whole fruits' more complex tissue structure.

Whole fruits are protected by the epicarp or exocarp (peel), making it more difficult for several plasma-reacting species to penetrate the fruit. In whole fruit, plasma-ionic species tend only to act on the surface, not penetrating the epicarp or mesocarp. Plasma non-ionic species, on the other hand, can penetrate the epicarp, reaching some millimeters deep into the mesocarp. As such, most of the whole fruit will not directly react with plasma species during plasma application. However, this does not mean that only slight changes will occur. The plasma species acting in the epicarp and a small fraction of the mesocarp can trigger the fruit's defense system, activating a series of metabolic responses.

The significant decrease in sucrose, glucose, and fructose observed in pitayas [33]; the increase in vitamin C in blueberries [28]; and the increase in phenolic compounds in strawberries, blueberries, and pitayas [83] are linked to these metabolic responses rather than a direct reaction with plasma-reacting species.

Minimally processed fruits usually have an exposed mesocarp, which facilitates the penetration of reactive plasma species into the fruit tissues. More chemical reactions are expected to occur in comparison with whole fruits. Minimally processed fruits can be considered fruit in stress conditions, with highly active defense mechanisms in progress. It is not uncommon for minimally processed fruits to present high concentrations of superoxide and hydrogen peroxide, which will increase the effects of other reactive plasma species.

Apple cubes subjected to plasma presented a significant reduction in sucrose, fructose, and glucose, which is usually linked to the activation of the defense systems [61]. The increase in vitamin C observed for minimally processed kiwi may be related to the direct reaction with plasma-reacting species, which was facilitated by the exposure of the mesocarp to plasma processing. In some cases, mesocarp exposition results in deleterious effects, such as the degradation of catechin, epicatechin, and procyanidin in minimally processed apples [75], which would not be likely to occur when treating the whole fruit.

Fruit juices do not have the protection of complex tissue structures, and therefore are more prone to chemical modification induced by plasma treatment. Most changes in composition were reported for plasma-treated fruit juices, as expected. Studies with fruit juices usually apply plasma to the surface of a static volume of liquid. Even though plasma is applied to the surface of the fluid, both ionic and non-ionic plasma-reacting species can permeate more deeply into the juice due to simple mass transport driven by the concentration gradient between the surface and the core of the fluid.

The application of plasma inside fruit juices is still a gap that needs to be addressed by researchers. In the past few years, plasma-activated water has been applied to the sanitization of foods, and the same technology could be used to inject plasma into fruit juices. The application of plasma as microbubbles inside fruit juices could increase the effects of plasma, increasing reactions rates and chemical transformations.

6. Plasma Treatment Benefits and Future Challenges

Cold plasma treatment of fruit and fruit juices could be viewed as a new unit operation aiming to improve products, and not only as a unit operation for sanitization. The

chemical reactions induced by plasma treatment can provide benefits such as improving the nutritional and sensory quality of fruit and fruit juices.

Plasma technology can increase the phenolic contents of fruit juices, improving the antioxidant capacity of fruit juices, with direct health benefits. Such improvements have been attained for most juices treated by glow discharge plasma and dielectric barrier discharge plasma (under some operating conditions).

Other more complex changes can be induced using cold plasma treatment. Plasma technology and operating conditions can be studied and optimized to cause designed changes to fruit products. Changes induced in sugar molecules have been designed to modulate the sweetness perception of apple juice [24]. The ability of plasma treatment to change the sucrose:fructose:glucose ratio in juices can be used to change the sweetening power of the juice, with a consequent impact on sweetness perception. Furthermore, the sugar:acid ratio can also be modulated to improve the perceived sourness intensity of fruit juices.

Pasteurized fruit juices tend to produce off-flavors derived from the thermal treatment. Oxidation induced by dielectric barrier discharge plasma can be used to reduce off-flavors caused by terpinol alcohols, as observed in orange juice [94], converting off-flavors back into characteristic flavor molecules.

The aroma can also be modulated using plasma treatment, increasing or decreasing several primary and secondary odor characteristics and conferring better aroma characteristics to fruit and fruit juices [7,95]. Such application requires a good knowledge of the aroma compounds, the chemical reactions and mechanisms induced by plasma treatment, and the odor thresholds of each aroma compound.

The improvement of several characteristics of fruits and fruit juices can be achieved by plasma treatment. However, the modulation of these characteristics requires deep knowledge of the chemical and biochemical reactions that may be induced by plasma treatment. Although several chemical mechanisms that occur during plasma application have been elucidated, there is still a lack of information on the effects of plasma on several chemical compounds, such as aldehydes, short-chain esters, ketones, furans, pyrans, pyrroles, and several amino acids. Further challenges consist of better understanding how different plasma environments and the concentration of the main reactive plasma species increase or decrease these chemical reactions.

7. Conclusions

Plasma technology produces many reactive species that react with fruit and fruit juices compounds. The reactions potentially affect all bioactive compounds, inducing several chemical transformations. Until now, most studies with cold plasma have presented a generic analysis of total phenolics, anthocyanins, and carotenoids, and antioxidant capacity. More complex studies on bioactive compound transformations are still required to understand the full potential of plasma technology applied to food processing. Comprehensive knowledge of the chemical pathway induced by plasma treatment may allow scientists and the industry to produce healthier and tastier foods.

Besides its known advantage in the sanitization and preservation of fruit and fruit juices, cold plasma technology can improve several nutritional aspects of these products. The technology can be used to reduce sugar content and improve phenolics, vitamin C, carotenoids, and the antioxidant capacity of fruit and fruit juices. Further studies should still be carried out to overcome some disadvantages of the technology, such as the degradation of lipids, oligosaccharides, and anthocyanins.

Future challenges are mainly related to the scale-up of the technology. Most studies were conducted in lab-scale equipment, and few works with pilot or industrial-scale equipment have been carried out. The key parameters that will lead to successful scale-up should be focused on in future studies.

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