Effects of non-thermal plasma on food nutrients and cereal-based raw materials

Efeito do plasma não térmico nos nutrientes de alimentos e matérias-primas à base de cereais

Efecto del plasma no térmico sobre los nutrientes de alimentos y materias primas a base de cereales

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Jefferson Henrique Tiago Barros
ORCID: https://orcid.org/0000-0002-9475-4289
Universidade Estadual de Campinas, Brazil
Instituto Federal de Ciência e Tecnologia do Acre, Brazil
E-mail: jefferson.barros@ifac.edu.br
Ulliana Marques Sampaio
ORCID: https://orcid.org/0000-0003-4664-7988
Universidade Estadual de Campinas, Brazil
E-mail: ulliana.unicamp@gmail.com
Flávio Martins Montenegro
ORCID: https://orcid.org/0000-0003-0476-4764
Instituto de Tecnologia de Alimentos, Brazil
E-mail: flavio.martins.montenegro@gmail.com
Caroline Joy Steel
ORCID: https://orcid.org/0000-0002-0911-5889
Universidade Estadual de Campinas, Brazil
E-mail: steel@unicamp.br
Jayr de Amorim Filho
ORCID: https://orcid.org/0000-0002-9250-4681
Instituto Tecnológico de Aeronáutica, Brazil
E-mail: jayr.de.amorim@gmail.com
Maria Teresa Pedrosa Silva Clerici
ORCID: https://orcid.org/0000-0002-8445-336X
Universidade Estadual de Campinas, Brazil
E-mail: mcleri@unicamp.br

Abstract
Non-thermal plasma (NTP) is an emerging technology that has been used for surface sterilization and food decontamination processes. However, studies have also shown promising results in modifying the characteristics of food raw materials with improvement in their physicochemical and technological properties. This review presents the different types of NTP and their effects on macronutrients, including carbohydrates, proteins, and lipids, and raw materials, such as wheat and rice flours, to modify their physicochemical characteristics. The studies have shown that NTP induced carbohydrates, lipids, and proteins oxidation, hydrolysis reactions, and pH reduction. These modifications occurred without using chemical agents, thus showing NTP as a promising alternative for use in the food industry. However, the modifications are dependent on the type of NTP, type of gas used, and complexity of the raw material composition. The challenges facing the industrial application of NTP include the mechanism of action of plasma, the regulatory aspects, scale-up, and consumer acceptance of the processed product.
Keywords: Cold plasma; Chemical modifications; Reactive species; Eco-friendly technology; Physical modification.

Resumo
O plasma não térmico (PNT) é uma tecnologia emergente que não gera resíduos ao meio ambiente. Inicialmente, o PNT foi aplicado em processos de esterilização de superfícies e descontaminação de alimentos, mas estudos mostraram resultados promissores na modificação das características de matérias-primas alimentícias com melhoria nas suas propriedades físico-químicas e tecnológicas. Nesta revisão, foram apresentados os diferentes tipos de PNT, bem como sua aplicação em macronutrientes, como carboidratos, proteínas e lipídios, e em matérias-primas, como farinhas de trigo e de arroz para modificação de suas características físico-químicas. O PNT foi capaz de promover modificações como a oxidação de carboidratos, lipídios e proteínas, hidrólise de ligações químicas, redução de pH. Essas modificações aconteceram sem o uso de agentes químicos, sendo promissor para uso na indústria de alimentos. No entanto, essas modificações são dependentes do tipo de PNT, tipo de gás utilizado e da complexidade de composição da matéria-prima. O mecanismo de ação do plasma, aspectos regulatórios, *scale up* e a aceitação pelos consumidores são os desafios encontrados para aplicação industrial do PNT.
Palavras-chave: Plasma frio; Modificações químicas; Espécies reativas; Tecnologia ecologicamente correta; Modificação física.

Resumen
El plasma no térmico (PNT) es una tecnología emergente que no genera residuos en el medio ambiente. Inicialmente, el PNT fue aplicada en procesos de esterilización de superficies y descontaminación de alimentos, no obstante, los estudios han mostrados resultados prometedores en la modificación de las características de las materias primas alimentarias mejorando sus propiedades fisicoquímicas y tecnológicas. En esta revisión se presentaron los diferentes tipos de PNT, así como su aplicación en macronutrientes, como carbohidratos, proteínas y lípidos, y en materias primas, como harina de trigo y arroz, para modificar sus características fisicoquímicas. El PNT promueve modificaciones como la oxidación de carbohidratos, lípidos y proteínas, hidrólisis de enlaces químicos, y la reducción del pH. Estas modificaciones sucedieron sin el uso de agentes químicos, siendo prometedoras para su uso en la industria alimentaria. Sin embargo, las modificaciones dependen del tipo de PNT, el tipo de gas utilizado y la complejidad de la composición de la materia prima. Por tanto, el mecanismo de acción del plasma, los aspectos regulatorios, el escalado y la aceptación del consumidor son los desafíos que se tienen para la aplicación industrial del PNT.
Palabras clave: Plasma frío; Modificaciones químicas; Especies reactivas; Tecnología ecológica; Modificación física.

1. Introduction

Raw materials or food ingredients are inputs used in manufacturing processed foods. They can be of the plant (fruits, wheat flour, rice, beans, and vegetable fats), animal (meat, milk, seafood), and mineral (water and salt) origin. Each raw material is composed of different chemical compounds, including carbohydrates, proteins, lipids, vitamins, and minerals, which nourish, interact with each other, and provide specificity to the product (Sikorski, 2007). For example, wheat flour is used to produce bread and pasta due to the formation of a protein network when mixed with water (Cauvain, 2015).

Chemical and physical characteristics of food components contribute to the texture, color, flavor, and final shape of the processed food product. Various technological processes have been used to modify ingredients, including physical (milling, dehydration, cooking, pasteurization, sterilization), chemical (modified starches, hydrolyzed proteins, interesterified fats, invert sugars), enzymatic (amylases, proteases, cellulases, lipases, etc.) treatments, as well as the addition of additives, such as colorants, emulsifiers, preservatives, among others (Brennan, 2006). The increasing consumer demand for clean label products, with the appeals of healthiness and environmental sustainability, has encouraged scientists and industry to look for emerging technologies that can meet these challenges, emphasizing non-thermal plasma (NTP).

NTP, also known as cold plasma, is a green technology, which emerged intending to replace technologies that use low processing temperatures (> 60 ˚C) and thus obtain foods with greater freshness, sensory properties, and better nutritional composition and physicochemical properties (Sonawane et al., 2020). The advantages of NTP include low temperatures (< 60 °C), no waste generation, no need to use water or solvent, low energy consumption, and rapid action on the material surface due to the high diffusivity of the plasma species. Moreover, it presents high efficiency in the inactivation of microorganisms and the possibility of use in solid and liquid systems (Misra et al., 2016a).

The main ingredient modifications by NTP were oxidation of lipids (Bahrami et al., 2016), starches (Yan et al., 2020), proteins, and amino acids (Meinlschmidt et al., 2016; Takai et al., 2014), in addition to starch depolymerization (Chaiwat et al., 2016; Thirumdas et al., 2017), and formation of protein cross-links (Bahrami et al., 2016; Misra et al., 2015). All these modifications will be discussed in this review.

This critical review aims to present the physical fundamentals of equipment and the advances in the use of NTP to modify the technological and physicochemical properties of macronutrients, such as carbohydrates, proteins, and lipids, as well as the future perspectives in this research field.

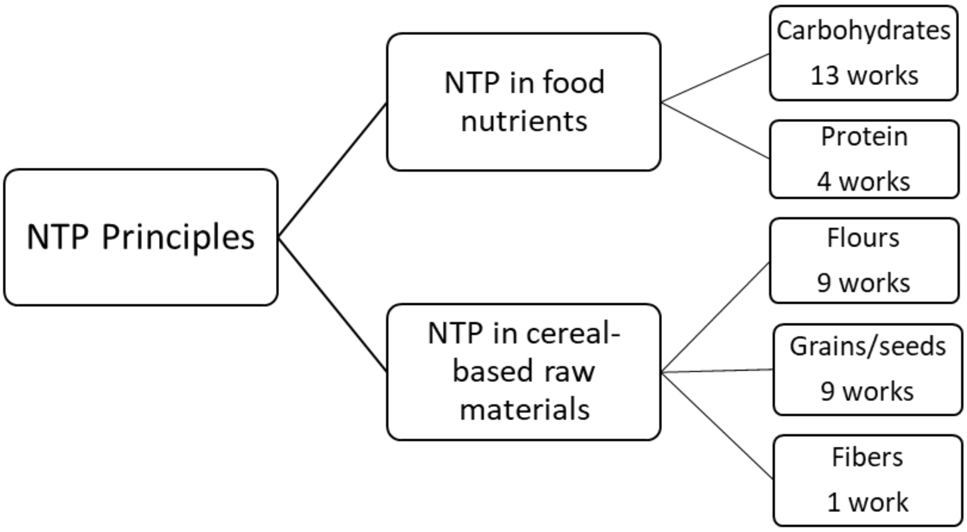
2. Methodology

This review established non-thermal plasma terminology to refer to plasma applied to food, although occurrences of the term's cold plasma or non-equilibrium plasma can be found. A few pioneering studies were used, emphasizing studies published in the last five years that used NTP in macromolecules or food raw materials, with a primary focus on macronutrients and cereals, such as wheat and rice. Other ingredients, such as lignocellulosic and whey, were also addressed, with significant results for discussing the action of NTP.

A narrative literature review Lakatos and Marconi (2010) was carried out due to the diversity of research found on different processes and effects of non-thermal plasma in various cereal-based raw materials. A survey of scientific articles and patents published between the period 2014 to 2021 was carried out to help the reader to understand the state of the art of each nutrient subjected to non-thermal plasma in different food matrices.

We performed the search from October 2018 to October 2021 in Scopus, Web of Science, Elsevier, Google Academic, Capes Periodicals, and Science Direct search engines, and for the search for patents, the INPI (National Institute of Industrial Property) search engine was used.) available at (https://busca.inpi.gov.br/pePI/jsp/patentes/PatenteSearchBasico.jsp) using the following filters: cold plasma, non-equilibrium plasma, cold plasma in food, cold plasma in carbohydrates, cold plasma in proteins, cold plasma in lipids, cold plasma in cereals, cold plasma in flours. After the identification of the works, they were grouped into (i) Principles of non-thermal plasma, which contributes to the basic knowledge of plasma and introduces essential concepts for the understanding of NTP in foods; (ii) NTP in food nutrients, which was divided into carbohydrates and proteins, and a section for lipids, fibers, and enzymes were inserted in the body of the work due to the few works found; (iii) NTP in cereal-based raw materials, such as wheat flour, rice flour, pea flour, insect flour, grains, and seeds, which aims to relate how the effects of NTP on nutrients affect the techno-functional properties of cereal-based raw materials. A flowchart of the selection of scientific publications is presented in Figure 1. With this, we selected thirty-six publications presented in Tables 1, 2, 3, and 4 and provide relevant information about the non-thermal plasma production process and its effects on nutrients in food and on the techno-functional properties of cereal-based raw materials.

Figure 1. Flowchart for the selection of scientific publications.



NTP: Non-Thermal Plasma. Source: Own authorship.

3. Understanding Non-Thermal Plasma

Most studies in food science and technology have focused on the use of NPT for the physical and chemical modification of nutrients. However, the equipment description and the process variables may not be fully described. Thus, some fundamental concepts have been addressed in this section.

Plasma is an ionized gas consisting of free electrons, positive and negative ions, atoms and/or reactive molecules, neutral molecules, and photons, with optical, electrical, and magnetic properties different from a neutral gas. The plasma most used in equipment is gas plasma, although plasmas in liquid and solid states are also found (Misra et al., 2016a). The plasma state can be achieved by providing sufficient energy (heat or electrical energy) to a gas or a mixture of gases, occurring at high pressure or low pressure (Loureiro & Amorim, 2016).

Plasma is electrically conductive and strongly interactive with electromagnetic fields due to the free electric charges (Tendero et al., 2006). Plasmas can be found in nature, such as in stars or the aurora borealis, lightning, or be produced in the laboratory. Those created in the laboratory can have reactive chemical species, which makes them effective for many emerging applications, such as sterilizing surgical materials in medicine, water treatment, and food preservation (Bermudez-Aguirre, 2020).

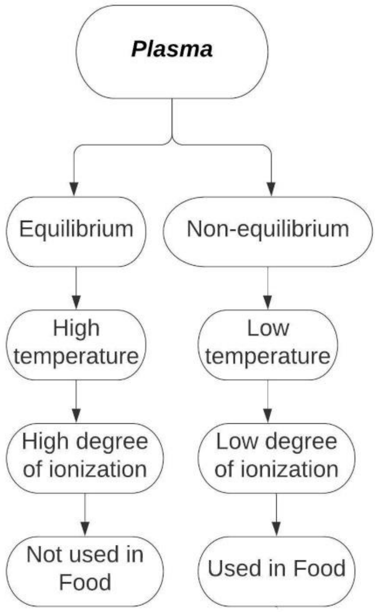
Plasmas can be in a state of equilibrium (thermal) and non-equilibrium (low temperature). When it is in a state of local thermal equilibrium, the electrons, ions, and neutrals have approximately the same temperature and energy, i.e., $T_{ion} \approx T_{electron} \approx T_{neutron}$. These plasmas commonly have an overall temperature around 10,000 K (Kelvin) and a degree of ionization (n_e / n_{total}) close to 100%; thus, they are not suitable for use in food and biological products (Loureiro and Amorim, 2016). On the other hand, in the non-equilibrium state, plasmas have electrons and ions at different temperatures. Whereas the electron temperature is around 10,000 K, the ions and neutral atoms will be close to room temperature (Demirci et al., 2020), which favors their use for compound modifications, including food ingredients. Furthermore, these NTPs are associated with a relatively low degree of ionization, $n_e/n_{total} \geq 10^{-6}$ (Loureiro and Amorim, 2016). The classification of plasma concerning the state of equilibrium is presented in Figure 2, where NPT can be considered a non-equilibrium plasma.

Plasma systems using high or low pressures are characterized as discontinuous processes that rely on closed devices. In this way, the continuous method opens up new possibilities for the safe and precise usage of plasma on food and biological surfaces, including medical applications (Turner, 2016). The wide variety of plasma generating equipment (Figure 3) has allowed the most different modifications of macromolecules, which will be discussed below.

3.1 Plasma chemistry and its sources

The plasma chemistry depends on the feed gas composition, humidity, power, and applied voltage. For a discharge initiated using a gaseous mixture containing oxygen, nitrogen, and carbon dioxide, the ions involved in the reaction include O^-, O_2^-, O^+, N^+, O_2^+, N_2^+, NO^+, and CO_2^+. Hydroxyl, nitrogen oxides, and atomic oxygen radicals are very reactive species formed by subsequent reactions. In addition to the particles generated in the plasma, their interactions with the biopolymer surface result in other free radicals and secondary electrons, which return to the gas phase, leading to the formation of cross-linking or grafting of oxygen-containing groups and depolymerization, as observed for starch granules (Misra et al., 2016a).

Figure 2. Classification of plasma.



Source: Authors.

The gas or gas mixture plays a significant role in forming NTP ionization products. The most commonly used gases are air, oxygen, nitrogen, and noble gases like helium and argon. However, plasma discharges with higher oxygen concentrations are desirable because they are associated with higher active oxygen species, including atomic oxygen and ozone (Misra and Jo, 2017; Pankaj et al., 2014). Atmospheric air has the greatest advantage over other gases due to its availability, cost, and oxygen in the composition (Sharma and Singh, 2020).

3.2 High-pressure plasma

The most common energy source used for the generation of high-pressure plasma is the application of electric fields, using electrodes inside or outside the discharge vessel. Direct current (DC) discharges require vacuum equipment to create a reduced pressure environment and are a closed system used for batch processes. On the other hand, a low-pressure environment requires low power levels to change the gas phase, producing the plasma (Loureiro and Amorim, 2016).

Recent progress in plasma device engineering has led to a new class of discharges operating at atmospheric pressure, which has no vacuum system and is easily operated, allowing continuous processes. Due to its non-thermal character and possibility of operation at atmospheric pressure, NTP at atmospheric pressure may offer a suitable approach for treating heat-sensitive foods such as fruits and vegetables. In addition, the high diffusivity of the plasma species allows for fast action with easy access to the entire food surface. In this case, atmospheric pressure NTPs offer high microbial inactivation efficiency at low temperatures (< 50 ˚C), allowing to extend the shelf life and contributing positively towards supply chain performance (Misra et al., 2016a).

3.3 Low-pressure plasmas

Currently used low-pressure plasmas are generated from radio frequency (RF) and microwave (MW). Electromagnetic waves drive these NTPs at frequencies of 13.56 MHz or 27 MHz for RF or 2.45 GHz for MW (Bárdos and Baránková, 2010). RF discharges can be created by electrodes immersed in the plasma or by using electrodes or coils outside the reactor. Microwave discharges are generated by a magnetron that delivers the microwaves coupled to the process chamber

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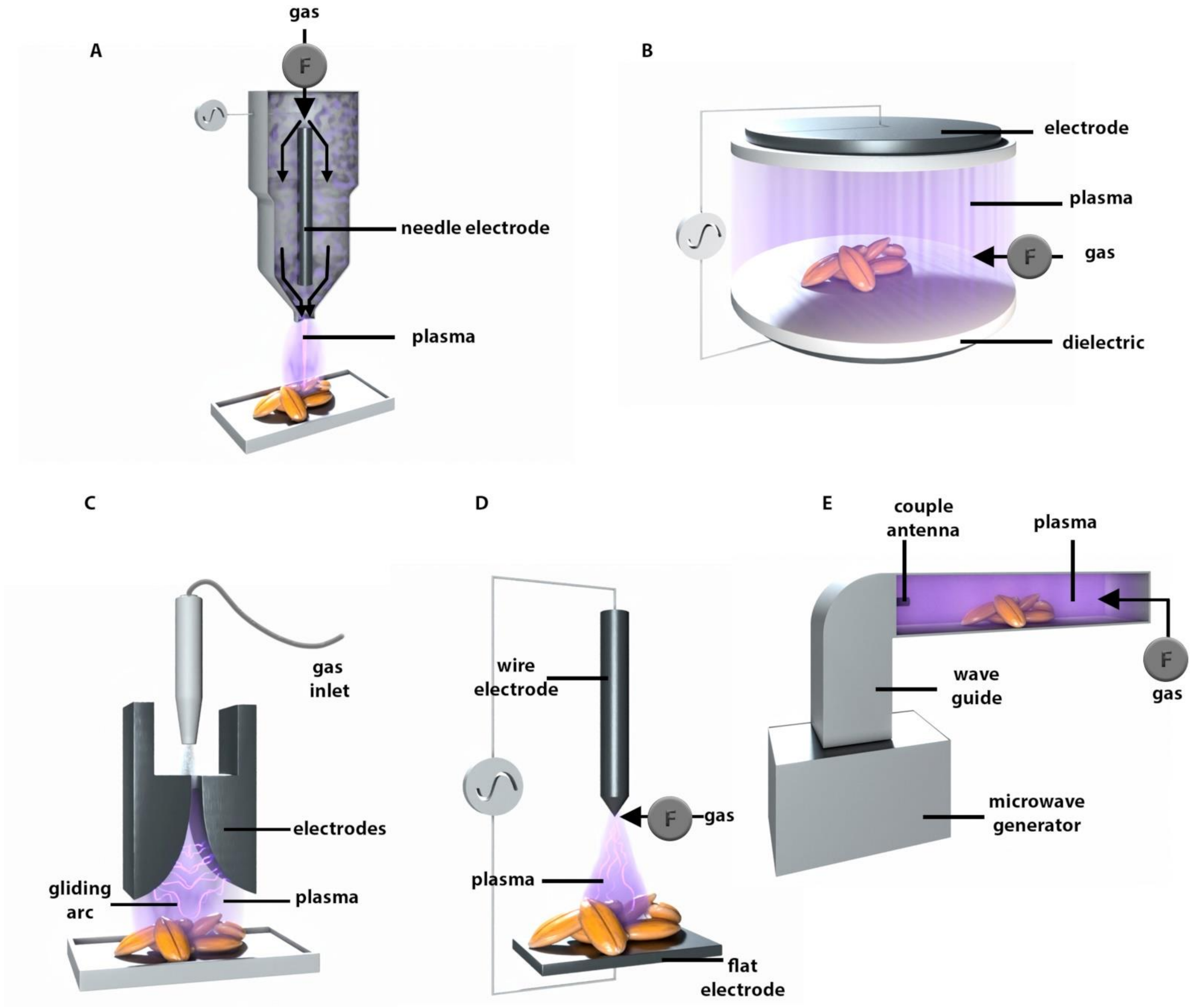
via a waveguide or coaxial cable. The electric field is absorbed mainly by electrons colliding with the gas after acceleration, causing heating, excitation, and ionization by inelastic collisions (Loureiro & Amorim, 2016).

Figure 3. Different schematic set-ups of plasma sources.



(a) atmospheric pressure plasma jet; (b) dielectric barrier discharge (c) gliding arc discharges; (d) streamer corona discharge (point-to-plane); (e) microwave discharge. Based of Surowsky et al. (2015). Source: Authors.

MW plasmas are considered advantageous compared to RF discharges once they provide higher power density to the system and can be easily ignited in the air due to the absence of electrodes. A lower gas requirement to generate large reactive species is another attractive characteristic of low-pressure discharges (Bárdos and Baránková, 2010). Although MW plasmas are spatially limited, applications requiring large surfaces need a series of discharges comparable to plasma jet arrangements.

3.4 Atmospheric pressure plasma

Common plasma sources for NTP generation at atmospheric pressure include plasma jets (PJ), dielectric barrier discharges (DBD), corona discharges (CD), and microwave discharges (MW) (Loureiro & Amorim, 2016).

3.4.1 Plasma jet (PJ)

Plasma jet discharges (Figure 3a) can have different configurations, allowing for outdoor operation. The species generated in plasma expand into the air and can treat materials. These small plumes produced by a pulsed radiofrequency plasma jet produce NTP with temperatures a few degrees above room temperature, favoring biological applications (Amorim et al., 2013). Common noble gases can be used in the operation of these jets, with flow rates of 10 L / min, which can be a limitation for food treatment due to the high cost, which can be minimized by using atmospheric air (Misra et al., 2016a).

3.4.2 Dielectric barrier discharges (DBD)

DBD (Figure 3b) is generated by an alternating voltage applied between two electrodes, which are held apart using dielectric materials. The main advantage of DBD reactors is the generation of low-temperature plasma under atmospheric pressure conditions, with chemical reactions like in the low-pressure glow discharge. DBD reactors are usually implemented by depositing an insulator over one or both metal electrodes, and the most commonly used dielectric materials include Pyrex, quartz, polymers, and ceramics. These dielectrics have the function of preventing the formation of sparks (Spyrou and Amorim, 2019).

DBD operates at frequencies from 10 Hz to 100 kHz, with different gases, many electrode geometries, and low gas flow rates. They require ignition voltages around 10 kV and a current of ~100 mA, with pulses on the order of 10 ns (nanoseconds) (Fridman et al., 2005). DBD plasmas exhibit a homogeneous/diffuse or filamentary regime depending on various experimental parameters such as gas used, electrode geometry, high voltage (HV) waveform voltage, amplitude and frequency, and dielectric material (Spyrou and Amorim, 2019).

DBDs can exhibit active species formed during the discharge due to the relatively low discharge current density, mainly through direct electronic collisions with neutral species (Demirci et al., 2020). When in the metastable state of molecular nitrogen N_2 ($A^3\Sigma^+{}_g$) and due to several factors including long lifetime, relatively high density, and high potential energy, these active species participate in energy transfer and formation of NOx (nitrogen oxides) molecules, influencing the discharge chemistry and contributing to the surface treatment of biological samples (Loureiro and Amorim, 2016).

3.4.3 Gliding arc discharges (GAD)

GADs (Figure 3c) are generated in a reactor with two or more metal drift electrodes, which are fed with a potential difference of around 9 kV and 100 mA of free air current. The inlet gas is pumped into the discharge duct, leading to an electric arc between the electrodes, and then expelled at the inlet duct in the divergent region. These reactors can be used for surface and liquid treatments and the degradation of chemical contaminants such as organics, solvents, industrial waste, and bacterial decontamination (Chizoba Ekezie et al., 2017).

3.4.4 Corona discharge (CD)

CDs (Figure 3d) are partial discharges, characterized by a weakly luminous discharge that appears at atmospheric pressure near sharp electrode geometries such as tips and wires. In this region, the electric field is vast and accelerates the electrons present in the medium to produce ionization reactions of the surrounding gas. Corona discharge reactors usually work at high voltage and occur predominantly at one electrode. The sharp electrode can be either positive or negative, i.e., positive corona or negative corona. The corona reactor is simple and easy to operate equipment (Bárdos and Baránková, 2010), and its main limitation is the minor treatment area with non-uniform production of reactive species (Chizoba Ekezie et al., 2017).

3.4.5 Microwave discharge (MW)

MWs (Figure 3e) are generated by electromagnetic fields originating from a magnetron. These discharges do not require electrodes and typically operate at a frequency of 2.45 GHz (Bárdos and Baránková, 2010). The electric field is conducted into the reactor chamber by a waveguide or applied to a launcher via a coaxial cable. Electrons absorb the energy of the microwave field, thus accelerating and leading to ionization reactions of atoms and molecules that make up the surrounding gas. The significant advance of these discharges is the absence of electrodes and the ease of operation. They can be ignited in various gases, including ambient air. Although spatial limitation is a disadvantage, a series of discharges can be implemented to cover large treatment areas (Loureiro and Amorim, 2016).

3.5 Plasma Device Settings

New plasma devices have emerged with several critical applications in food. There are two approaches, classified as indirect and direct, based on plasma at atmospheric pressure. The indirect treatment uses the post-luminescence elements of plasma generated in a remotely located plasma discharge. On the other hand, direct treatment uses direct contact of the material with the plasma luminescence, which brings charged energetic particles to the plasma/material interface (Fridman et al., 2007).

Various equipment used for NTP production has been described in the literature, leading to different changes in the same ingredient; thus, a detailed description of NTP and its variables is necessary for process reproducibility. Shortly, studies will lead to further discussion on the effects of NTP on foods concerning the type of plasma and its parameters, including treatment time, amount and type of reactive species, type of gas, type of food/nutrient, and its physical and chemical characteristics before and after the treatment (Bahrami et al., 2016; Bie et al., 2016; Chaple et al., 2020; Chizoba Ekezie et al., 2019; Gao et al., 2021; Hernandez-Perez et al., 2021; Misra et al., 2015; Okyere et al., 2019; Thirumdas et al., 2017).

Regarding the industrial application in the food area, the significant challenges include scalability, adaptability to different food processes, and the peculiarities of the processes. NTP is a novel technology and requires extensive knowledge of various ingredients and foods from various segments. Further studies are needed to elucidate the plasma chemistry and its mechanism of action in a complex matrix once few studies have been addressed on these interactions. Misra et al. (2015) evaluated the composition of ambient air DBD plasma by optical emission spectroscopy and identified reactive oxygen and nitrogen species and their effects on wheat flour.

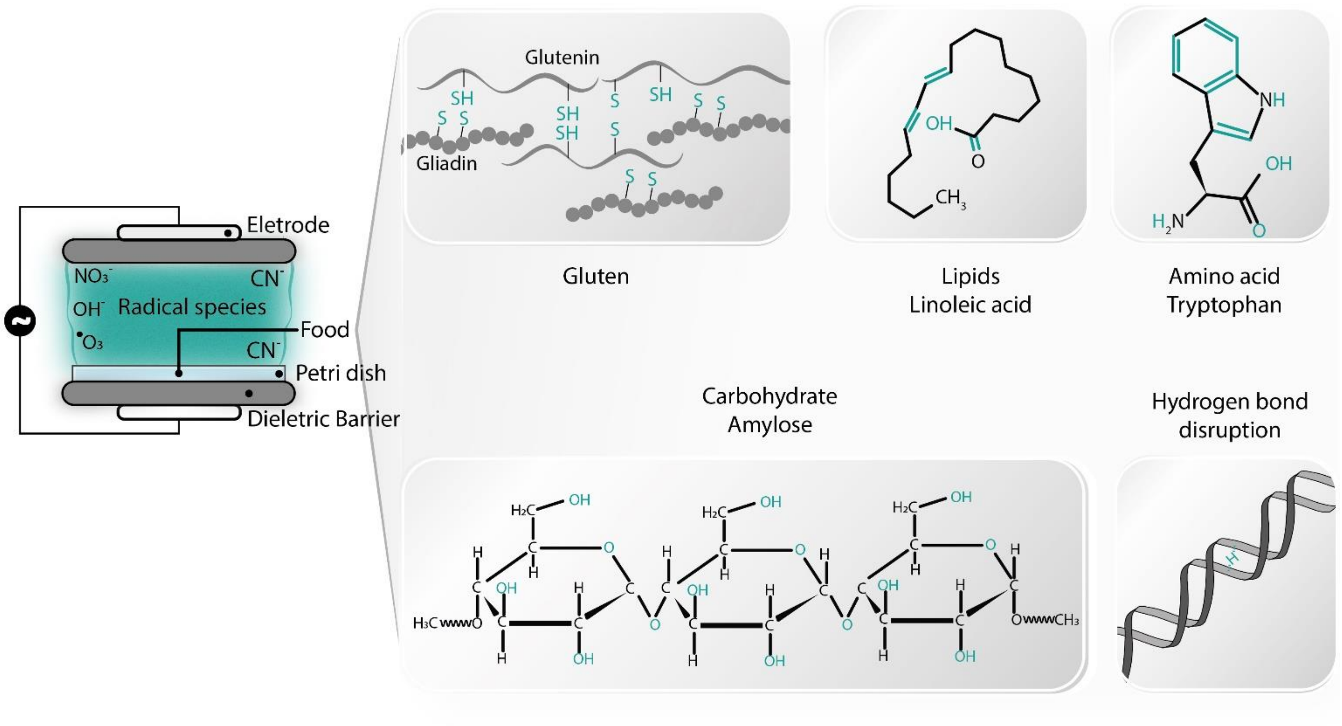
4. Effects on Food Ingredients and Raw Materials

To elucidate the use of NTP in food ingredients, this study presented the researches on nutrients, and studies and patents using raw materials (different flours, grains, and lignocellulose material), to investigate the possible interactions between the different nutrients.

4.1 Nutrients

Studies using isolated nutrients (model systems) allow understanding the main modifications that occurred during the process and contribute to understanding a more complex matrix containing several nutrients. The main macromolecules (carbohydrates, lipids, and proteins) will be addressed in this study due to the scarcity of studies involving NTP in vitamins and minerals. It is worth mentioning that each nutrient has different physicochemical properties; thus, only indicative changes of the isolated compounds will be addressed. Figure 4 shows the main effects of non-thermal plasma on food components.

Figure 4. Effect of non-thermal plasma on food components.



Proteins: denaturation, disruption of bonds, oxidation; Lipids: oxidation at double bonds, formation of free radicals, hydrolysis of triacylglycerols into free fatty acids; Amylose: oxidation (carbonyls), hydrolysis with formation of reducing sugars, disruption of bonds between C_2 and C_3 of glucose. Source: Bahrami et al. (2016) and Misra et al. (2016).

4.1.1 Carbohydrates

Within the class of carbohydrates, starch and dietary fiber are the most used as body and texture agents in foods. The most desired modifications of starch granules for use as an encapsulating agent, film-forming agent, and fat replacer include increased water absorption, emulsification capacity, reduced retrogradation upon cooling, enzyme susceptibility, and changes in composition, structure, and thermal properties (Banura et al., 2017; Bie et al., 2016; Thirumdas et al., 2017; Zhu, 2017).

Concerning fibers, the modifications are focused on increasing water absorption, water holding capacity, and reducing cellulose/hemicellulose chain size to produce soluble fibers (Fuller et al., 2016).

4.1.1.1 Starch

Starch is a reserve homopolysaccharide of plants formed by glucose units. It is widely used in industry for its abundance, low cost, thickening profile, and energy value (Huber and BeMiller, 2009). The main starches produced on an industrial scale are corn, cassava, and potato. These starches have different amylose/amylopectin ratios, providing different pasting properties. They can be modified by various chemical, physical, enzymatic, and genetic methods, emphasizing physical methods for not generating effluents (Maniglia et al., 2021). Alternative starch sources and physical modification processes have been studied to extend the applicability of this carbohydrate, including the use of NTP in the unconventional buckwheat starch (Gao et al., 2021) and banana (Yan et al., 2020).

The main starch modifications after the NTP application are presented in Table 1. Maniglia et al. (2021) and Zhu (2017) reported the interaction between reactive plasma species and amylopectin side chains. The formation and/or increase of particle size, cross-linking, depolymerization, and the inclusion of functional groups have been reported in several studies (Clerici et al., 2009; Li et al., 2021; Muhammad et al., 2018; Thirumdas et al., 2017; Zhang et al., 2014; Zhu, 2017), and the degree of modification depends on the type of plasma, type of gas, process parameters, and amylose to amylopectin ratio.

The starch modifications presented below are the most common and reported in the literature, showing a certain consensus for different types of starches.

4.1.1.1.1 Moisture

The reduction in moisture can occur by the interaction between reactive plasma species and water molecules from starch, leading to the rupture of hydrogen bonds and, consequently, causing the water release from the system. Chaiwat et al. (2016) reported a reduction from 13.5% to 6.2% moisture content in cassava starch subjected to NTP with processing time from 30 to 180 min. In a granted patent, Clerici et al. (2009) reported moisture contents close to 1% in starch at the end of the process, with no visible changes in starch color even after vacuum treatment and initial moisture of 8%.

4.1.1.1.2 pH

The pH reduction in starch is related to the introduction of new functional groups into the molecular structure or formation of acidic groups (carboxyl groups and carbonyls) due to oxidation of hydroxyl groups (Banura et al., 2017), which can lead to hydrolysis of the C_2-C_3 bonds of the glucose molecule. Thirumdas et al. (2017) studied rice starch treated with NTP with atmospheric air and reported a reduction in pH from 7.42 to 6.96.

4.1.1.1.3 Molecular Structure

The structural modifications of the amylose and amylopectin molecules are based on the chemical reaction prevailing during the plasma treatment. Cross-linked bonds can be formed, favoring, for example, a more robust gel structure and reduced paste luminosity (Clerici et al., 2009; Wongsagonsup et al., 2014), or starch depolymerization, leading to a reduction of the amylose content by disruption of the polymer structure due to the action of free radicals and electrons present in plasma (Clerici et al., 2009; Thirumdas et al., 2017) generating low molecular mass molecules and simple sugars. On the other hand, Banura et al. (2017) observed an increase in the number of molecules with linear chains in cassava starch (17.21 to 20.06 g / 100 g), due to depolymerization of amylopectin side chains. Therefore, determining the amylose content in plasma-treated starches may give a false result of increased amylose in the starch granule.

Starch depolymerization can increase the binding capacity with polyphenols by approximately 10-fold, as observed by Gao et al. (2021), or the water solubility, as reported by Yan et al. (2020) in banana starch.

Sifuentes-Nieves et al. (2021) evaluated corn starches with different amylose contents in the presence of HMDSO (Hexamethyl di-siloxane) subjected to NTP with different gases (atmospheric air, O_2, N, Ar) and reactors (RF and DBD). The authors reported that the results favored the ordering and stability of the amylose molecule; thus, it can be used in the food industry and/or other fields such as semiconductors.

Table 1. Modifications in starch properties after application of non-thermal plasma.

Plasma device	Process conditions	Sample	Modificação em comparação com o controle	References
DBD[1] Low pressure	Gas: Air Time: 1, 5 and 10 min Parameters: 50 V, 1,5 A and 75 W The sample was conditioned in a reactor with 40 % RH	Corn starch	↑ Oxidation; fissures ↓ Molecules with a low molecular weight; viscosity	(Bie et al., 2016)
PJ[2] Atmospheric pressure	Gas: Argon (1 L/min) Time: 5 min Power: 50 and 100 W Frequency: 600 MHz Constant stirring of samples during treatment	Cassava starch slurry 20 % (w/w)	↑ Thermal stability of the gel structure at 50 W ↓ Paste clarity	(Wongsagon sup et al., 2014)
		Cassava cooked starch 2 % (w/w)	↑ Paste clarity ↓ Strong gel structure	
RF[3] Low pressure	Gas: Air Time: 5 and 10 min Power: 40 and 60 W Pressure: 0.15 mbar Frequency: 13.56 MHz	Rice Starch	↑ Water absorption index; Fissures on the granule ↑ Final viscosity (RVA) ↑ Amylose content and turbidity	(Thirumdas et al., 2017)
Semi-continuous Low pressure	Gas: Argon Time: 30 to 180 min Power: 60 W Pressure: 12 mbar Samples passed through the system in 1, 3 and 6 cycles	Cassava starch	↑ Breakdown (RVA); paste clarity; depolymerization ↓ Moisture	(Chaiwat et al., 2016)
RF Low pressure	Gas: Air Time: 10 and 20 min Power: 40 and 60 W Frequency: 13.56 MHz	Corn starch	↑ Water binding capacity ↓ pH; peak temperature and enthalpy gelatinization	(Banura et al., 2017)
		Cassava starch	↑ Amylose content; ↑ Water binding capacity; enthalpy gelatinization ↓ pH	
DBD Low pressure	Gas: Oxygen Time: 30, 45 and 60 min Voltage: 245 V 12 % moisture (samples)	Corn starch and Potato starch	↓ Molecular weight	(Zhang et al., 2014)

MW[4]	Gas: $N_2$and N_2-O_2 (80:20) Gasflow: N_2 (1 L/min) e N_2-O_2 (2.5 L/min) Time: 20 min Power: 900 W	Corn starch citrate	↑ Swelling power ↑ Peak viscosity using N_2, but decreased using N_2-O_2 ↑ Setback using N_2, but decreased using N_2-O_2 ↓ Resistant starch ↓ Solubility and gelatinization temperature ↓ Formation of ester bonds between starch and anhydrous citric acid	(Kim & Min, 2017)
RF Low pressure	Gas: Argon, Oxygen, Helium and combinations between them Time: 5 to 30 min Power: 30 to 70 W Samples up to20 % moisture	Corn starch	↑ Porosity; facility of starch to be digested by amylolytic enzymes; digestibility ↓ Moisture; viscosity	(Lambert, et al., 2018)
DBD	Gas: Air Time: 30 s Voltage: 20 kV	Buckwheat starch	↑ Formation of quercetin complex ↓ Consistency coefficient; Crystallinity; resistant starch ↓ Flow behavior index;digestion velocity	(Gao et al., 2021)
RF Low pressure	Gas: Carbon dioxide-argon and argon Time: 60 min Power: 120 W Frequency: 13.56 MHz Flow rate: 25 standard cubic centimeters per minute (sccm) and 15 sccm	Waxy rice, maize and potato starch	↑ Enthalpy of gelatinization ↓ Setback and final viscosities (RVA) ↓ Crystallinity of waxy potato starch	(Okyere et al., 2019)
RF Low pressure	Gas: Hexamethyldisiloxane (HMDSO) Pressure: 0.4 mbar Time: 10 and 30 min Power: 90 W Frequency: 13.56 MHz Flow rate: 0.35 sccm	Normal and high amylose corn	↓ Paste Viscosity; film flexibility	(Hernandez-perez et al., 2021)
DBD and RF Low pressure	Gas: Hexamethyldisiloxane (HMDSO) Power (RF): 90 W Frequency (RF): 13.56 MHz Voltage (DBD): 12 kV Frequency (DBD): 25 kHz Flow rate: 0.35 cm^3/min	Corn starches with different amylose/amylopectin	↑ RF: Amylose and thermal stability ↑ DBD: Stability of starch molecules	(Sifuentes-Nieves et al., 2021)

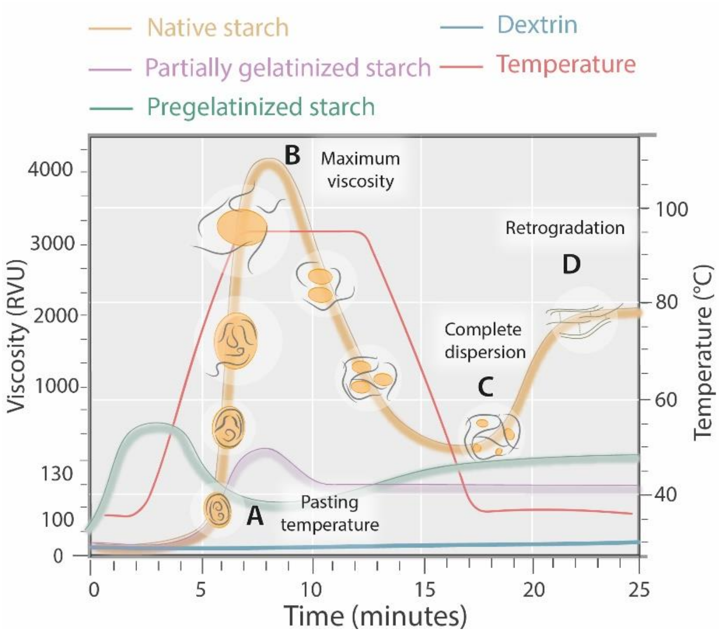
	Time: 10 min			
DBD	Voltage: 30 – 50 V, Time: 3 min	Banana starch	↑ Solubility; Depolymerization, decomposition and plasma etching ↓ Paste Viscosity	(Yan et al., 2020)

[1]DBD: Dielectric Barrier Discharge; [2]PJ: Plasma Jet; [3]RF: Radio Frequency; [4]MW: Microwave;↑: increase; ↓: decrease. Source: Authors.

4.1.1.1.4 Starch paste viscosity

Studying the behavior of starch granules during heating in water is essential to determine several parameters, including the swelling power and viscosity, initial paste temperature, stability to prolonged heating under stirring, and retrogradation during cooling. These parameters are used to evaluate the production of soups, porridges, pregelatinized starches, and the energy expenditure required to make the starch accessible to the digestive process. These analyses can be done on devices that determine empirical rheology, such as RVA (Rapid Visco Analyser) and the Brabender viscometer. Figure 5 shows viscosity curves, with examples of each type of starch (native, partially gelatinized, pregelatinized, and dextrin).

Figure 5. Viscosity curves of different types of starches and dextrin.



Source: Authors.

A decrease in paste viscosity upon heating is observed due to starch depolymerization and retrogradation during cooling, showing possible starch dextrinization, as reported by several authors (Bie et al., 2016; Clerici et al., 2009). This behavior may allow starches in products in which starch retrogradation is not desired, such as gluten-free bread and cakes. Table 1 shows changes in paste luminosity and weaker gel formation, which matches the increased fragility of the starch granule, which breaks more easily during gelatinization. These events lead to a decrease in gelatinization temperature, which may represent savings in the process since starch gelatinization occurs under continuous stirring, with a significant increase in viscosity up to 95 ºC, as shown in Figure 5.

4.1.1.1.5 Porosity and other properties

The formation of cracks or cavities on the surface of starch granules, mainly in potato starch (Zhang et al., 2014), can occur at higher powers and treatment times. Potato starch is classified as a B-type crystalline structure, consisting of starch granules with more inter-helical water molecules; therefore, it is more susceptible to plasma active species, such as oxygen-free radicals, which produce hydrogen-free radicals upon contact with water present in the molecule. The radical grater formation leads to increased starch degradation, reducing its molecular weight (Zhang et al., 2014).

The higher surface porosity of NTP-treated starches can also increase the digestibility and accessibility of amylolytic enzymes, turning the raw intact starch into a damaged starch (Clerici et al., 2009). Gao et al. (2019) reported this behavior who studied sorghum starch and found a 48.29% increase in digestibility in the rapidly digestible starch (RDS) content compared to the untreated sample. According to the authors, these characteristic favors hydrolysis processes, such as bioethanol production, fermentation, maltodextrin production, and several other processes involving chemical modifications, such as acid hydrolysis and oxidation, due to the greater accessibility of reagents to the interior of the granule.

4.1.1.2 Fibers

Dietary fibers have nutritional benefits, such as regulation of the digestive system (Dhingra et al., 2012; Fuller et al., 2016), as well as acting in the food industry as a thickener, emulsifier, sugar, and fat substitute, and promoting sensory changes, as reported by Attia et al. (2010).

Few studies with NTP-modified dietary fibers have been found, and the main changes were observed for the water absorption capacity of the matrix. Therefore, this field may be promising for modifying the properties of dietary fibers.

Misra et al. (2018) evaluated the properties of xanthan gum after treatment in DBD-generated plasma at atmospheric pressure using ambient air. The authors reported an increase in brightness and viscosity of the gum solution (1% w/w), probably due to an increased ability of the material to form hydrogen bonds, thus generating a more ordered network. In addition, greater emulsion stability was observed with increasing viscosity, reducing the mobility and collision between the dispersed phase droplets. Thus, the modifications in xanthan gum may be effective for food applications such as salad and instant dry soup formulations.

On the other hand, studies showed that non-food fibers - lignocellulosic materials - presented several technological benefits after the NTP treatment (Barra et al., 2015; Macedo et al., 2020). Lignocellulosic materials are mainly composed of cellulose, hemicellulose, and lignin and are used to produce high value-added products such as xylitol, oligosaccharides, and biofuels, among others. A review study by Pereira et al. (2021) reported that the pretreatment with NTP led to the breakdown of bonds responsible for the structure of lignocellulosic materials such as wheat straw, natural coconut fibers, sugarcane bagasse, etc.

Amorim et al. (2013) performed the breaking of the molecular structure of sugarcane bagasse lignin using argon plasma jet at atmospheric pressure for more efficient bioethanol production. In the pretreatment with NTP, the authors observed that, in the plasma discharge. Reactive electrons broke the aliphatic chain and the aromatic ring of the lignin structure of the sample, which was not observed when the treatment was performed in the post-discharge (sample 1 cm distant from the jet). According to the authors, the generated species probably did not present the same reactivity when reaching the sample surface. In contrast, the patent of Xu et al. (2016) showed efficiency in applying DBD-NTP to micronized cereal lignocellulose to provide an increase in enzyme efficiency, facilitating saccharification.

Fazeli et al. (2019) also demonstrated that cellulose fiber subjected to RF-NTP increased surface roughness and porosity due to the partial attack and ablation of amorphous regions, improving adhesion to thermoplastic starch. This event improved the mechanical properties of biocomposite films compared to untreated fiber. Furthermore, an increase in fiber hydrophilicity was observed due to the chemical reactions favored during the treatment that induced an increase in functional groups such as COO^-, OH, and C=O.

4.1.2 Lipids

Lipids are organic molecules formed from fatty acids and glycerol (polyol). Oils are more affected by NTP than fats due to their unsaturated fatty acid composition, facilitating oxidation (Larsson et al., 2006). Thus, oxidative and/or hydrolytic rancidity generated by NTP treatment in lipid matrices can compromise food quality, as Gavahian et al. (2018) and Sarangapani et al (2017) reported.

Cui et al. (2017) and Sarangapani et al. (2017) reported that NTP from oxygen and atmospheric air are primarily responsible for inducing lipid oxidation. The authors stated that reactive oxygen species (ROS) are responsible for initiating the oxidation process by interacting with the double bonds from lipid chains. According to Bahrami et al. (2016), lipids with a high degree of unsaturation, such as those with linoleic (18: 2) and linolenic (18: 3) fatty acids, are the most sensitive to these changes.

In cereals, Bahrami et al. (2016) applied NTP at atmospheric pressure to wheat flour using power densities of 0.19 W / cm^2 and 0.43 W / cm^2 and observed an increase in lipid oxidation with increasing the exposure time. At high voltage (20 V) and treatment times of 60 and 120 s, the oxidation reduced phospholipids and free fatty acids while linolenic acid was completely oxidized.

The application of plasma on lipids has some advantages. Vandamme et al. (2015) accelerated the oxidation of fish oil via NTP and concluded that the plasma technology has advantages over the traditional tests to assess the oil stability and may be an effective alternative for this purpose.

NTP use in the transesterification process for biodiesel production (Cubas et al., 2006) and hydrogenation of oils (Puprasit et al., 2020) showed promising results. Puprasit et al. (2020) used NTP to produce margarine from palm oil and soybean oil without using a catalyst. The authors reported a reduction of the iodine value from 60.89 to 48.39 and trans-fat by 1.44 % after 4 h of treatment. This result represents a trans-fat generation rate of 6.12 times lower than a conventional method based on high temperature, high pressure, and the use of catalyst.

Fat-containing foods are one of the technological challenges for applying NTP; thus, the control of process parameters can contribute to minimizing the lipid changes. Techniques to prevent oxidation using antioxidant ingredients such as BHA (Cui et al., 2017), essential oils (Tyagi et al., 2012), and the use of vacuum packaging associated with plasma (Bauer et al., 2017) can assist in reducing these changes. However, studies on this subject are scarce. Concerning processes with plasma generation by electrical discharge, it is recommended to control the power, time, and treatment temperature at the lowest possible values and to use inert gases in the process when required (Bahrami et al., 2016; Gavahian et al., 2018).

Therefore, preventing the lipid oxidation of plasma-treated foods and the possible interactions with other nutrients may challenge the successful use of plasma.

4.1.3 Protein

The properties of food proteins depend on several factors, including the amino acids composition, the arrangement of bonds that stabilize the protein structure, molecular mass, amino acid sequence, structure (secondary, tertiary, and quaternary), net charge, presence of other proteins, and ability to react with other components (Phillips and Williams, 2011). Thus, studies on protein modifications are much more complex due to their greater diversity of functional groups when compared to carbohydrates and lipids, which makes the protein much more reactive. The biological functions of proteins may be structural, antibodies, biological catalysts, and hormonal; in foods, they may have emulsifying and foaming properties, besides providing a three-dimensional network, among others (Lorient et al., 1989).

Proteins can be of animal or plant origin, and plant-based proteins have been widely studied in NTP due to the greater interest as a complementary ingredient for nutritional intake in juices, meat derivatives, and dairy beverages. In addition to good nutritional quality and functional properties, these plant sources stand out for their technological properties, including foaming capacity, emulsification, stabilization, water, and fat binding, gel, and dough forming capacity, among others (Lorient et al., 1989), with specific effects during food processing and storage. The NTP has been applied in various protein sources, including protein meal (Bußler et al., 2015, 2016; Dong et al., 2017), protein concentrate (34 and 80% protein) (Segat et al., 2015), and protein isolate (protein content greater than 90%) (Chizoba Ekezie et al., 2019; Yang et al., 2018).

Sharma and Singh (2020) and Saremnezhad et al. (2021) reported that the mechanisms of NTP modification in proteins are related to the cleavage of peptide bonds, oxidation of amino acid side chains, and cross-link formation between polypeptide chains. Table 2 presents the main modifications in proteins by NTP, such as increased solubility (Bußler et al., 2016), thermal stability (Yang et al., 2018), and viscosity (Bahrami et al., 2016) (Table 2). Other modifications caused by the application of NTP are described below.

4.1.3.1 pH

As shown in Table 2, several authors have reported a reduction in pH due to the exposure time between plasma-generated active species [ROS and reactive nitrogen species (RNS)] and the sample during the treatment (Dong et al., 2017; Chizoba Ekezie et al., 2019; Segat et al., 2015). Dong et al. (2017) studied the effects of DBD on the physicochemical and structural properties of zein (corn protein) powder and reported an increase in the concentration of free sulfhydryl groups within 7 min of treatment at 75 V by indirect exposure, leading to a reduction in the pH of the solutions.

Table 2. Modifications in protein properties after application of non-thermal plasma.

Plasma device	Process conditions	Sample	Modification	References
DBD[1]	Gas: Atmospheric air Treatment time: 120 s Process parameters: 60 V, 1.0 A and 12 kHz The sample was conditioned in a reactor at 50 % RH and 20 ˚C	Ferritin (red beans)	↓ The molecular weight; denaturation temperature; hydrophobicity	(Yang et al., 2018)
PJ[2]	Gas: Atmospheric air (10-15 L/min) Treatment time: 0, 2, 4, 6, 8 and 10 min under stirring (250 rpm) Process parameters: 220 V, 10 A, 50 kHz, an output voltage of 7 kV	20 mg/mL solution of myofibrillar protein from raw shrimp	↑ Protein-protein interaction; mean particle diameter; ↓ pH and solubility	(Ekezie et al., 2019)
DBD	Gas: Atmospheric air Treatment time: 2 min Process parameters: 50, 75, 100 and 125 V	Zein powder (corn protein)	↑ The concentration of free SH groups ↑ Solubility at 75 V ↑ Depolymerization ↓ Average particle diameter	(Dong et al., 2017)
NS[3]	Gas: Atmospheric air Treatment time: 1 to 60 min Process parameters: 70 kV Quantity: 30 mL in 5 mm diameter Petri dish	Whey protein isolate	↑ Carbonyl groups (15 min) ↓ pH ↓ Free SH groups (15 min) ↓ Foaming and emulsifying capacity (30-60 min) ↑ Oxidation of proteins	(Segat et al., 2015)

[1]DBD: Dielectric Barrier Discharge; [2]PJ: Plasma Jet; [3]NS: Not specified; ↑: increase; ↓: decrease. Source: Authors.

4.1.3.2 Oxidation

Oxidation of amino acids occurs by exposure to ROS, including O3, and secondary by-products of oxidative stress. Oxidation induces protein unfolding, increasing their surface hydrophobicity, causing protein aggregation and polymerization due to intermolecular bonds (Zhang et al., 2014).

Chizoba Ekezie et al. (2019) evaluated the conformation and physicochemical properties of myofibrillar proteins extracted from king prawn treated by JP under atmospheric pressure and found that the secondary structure of proteins was affected within 10 min of treatment due to the action of reactive species on acidic residues of their amino acids, leading to molecule unfolding and increase in mean particle diameter from 654 to 2297 nm, with the promotion of protein-protein interactions, exposure of hydrophobic groups, and consequent reduction in water solubility. Segat et al. (2015) studied whey protein isolate and observed an oxidizing effect of NTP on the free SH groups of cysteine, leading to increased protein aggregation.

Considering these possible modifications, it is important to evaluate whether the traditional methodologies used for protein analyses are necessary after applying plasma technology. Osborne and Voorhees (1894) classified proteins according to their solubility in water, salt solution, alcoholic solutions, and diluted acid and base. The modifications caused by NTP, such as oxidation and new intermolecular bonds, can lead to the proteins losing their native form. Thus, studies are required to confirm whether these methodologies for protein characterization can be applicable as in native protein.

In addition, NTP can affect the protein extraction and purification processes, and oxidation of a single amino acid can affect protein functionality. Aromatic amino acids and sulfur-containing amino acids (-SH) such as cysteine and methionine, for example, are sensitive to oxidation (Mollakhalili-Meybodi et al., 2021; Surowsky et al., 2013). Furthermore, a review study by Esteghlal et al. (2019) showed that oxidation reduced the nutritional value of the food due to changes in the amino acid profile that affected bioavailability, as well as the sensory aspects, once free amino acids can alter the flavor of food, which has not been investigated.

4.1.3.3 Molecule structure

Yang et al. (2018) evaluated the effect of pH changes on ferritin (an iron-containing protein) from kidney bean extract subjected to NTP to improve its microencapsulation properties and reported changes in the secondary structure of proteins. The results showed that ferritin can be cleaved at pH 4.0 and then restructured as intact ferritin when increasing pH to 7.0, maintaining the native ferritin-like structure. This property allowed encapsulating curcumin molecules at the concentration of 12.7% (w/w). Regarding the protein structure, the treatment reduced α-helix content by 1.4% and β-sheet by 2.3%, with a 6.3% increase in random coil form. These changes affected hydrogen bonds and reduced the molecule's hydrophobicity, decreasing the thermal stability.

Dong et al. (2017) studied the effect of NTP on the physical and chemical structures of zein and confirmed changes in the secondary structure. The authors concluded that the reactive species interacted with the zein surface, reorganizing its conformation and promoting increased hydrophilicity. Segat et al. (2015) observed a certain degree of the unfolding of whey proteins, which resulted in improved foaming and emulsifying capacity when subjected to NTP for up to 15 min.

4.1.3.4 Enzymes

Enzymes are a class of proteins with biological functions, except for ribozymes (RNA), which interfere in food processing and preservation. They have the function of facilitating the reactions, decreasing the energy required for biochemical processes, and may have undesirable effects on food (Damodaran et al., 2007). The undesirable effects are directly related to the action of endogenous enzymes, which can cause browning reactions (peroxidase and polyphenol oxidase), pectin modifications (polygalacturonases, pectin methylesterase), and lipid oxidation (lipases). Research has shown that these undesirable effects can be minimized using NTP (Surowsky et al., 2013; Tolouie et al., 2021).

Han et al. (2019) and Misra et al. (2016b) reported in a review study that reactive plasma species can alter the conformational structures of enzymes, especially the secondary structure. The enzyme activity depends on the intact conformational structure of the enzyme; thus, a single oxidized amino acid in the protein can lead to a change in enzyme function (Takai et al., 2014; Zhang et al., 2015).

Although studies have shown the effect of reducing the enzyme activity by NTP (Tolouie et al., 2021), it is worth noting that this research is limited only to liquids or thin samples, which does not reflect a complex system, with enzymes interacting with other components, or systems containing other desirable enzymes that may be affected by the treatment (Han et al., 2019). For example, wheat flour contains amylases, which are essential for bread making, while polyphenol oxidases and peroxidases also present in wheat flour are responsible for the undesirable color formation in fresh pasta.

As NTP is a surface treatment, the inactivation of endogenous enzymes in the whole fruit is challenging. In addition, some authors have also reported the degradation of vitamin C (Xu et al., 2017) and oxidation of some amino acids (Tolouie et al., 2018). Therefore, enzymes and lipids are placed as limiting factors in processing by NTP.

4.1.3.5 Allergenicity

It is expected that plasma can contribute to treating proteins with recognized allergenicity, making them hypoallergenic. However, according to the review study of Chizoba Ekezie et al. (2017), allergenicity can reduce due to the formation of insoluble aggregates and protein depolymerization, changing the structure of linear and conformational epitopes, thus reducing their reactivity.

Recent studies have shown that NTP reduced the immunoreactivity of soy proteins (β-conglycinin and glycine) from 87% to 100% (Meinlschmidt et al., 2016). In shrimp tropomyosin, DBD plasma showed a 76% reduction in allergenicity within 5 min of treatment at 30 kV (Shriver and Yang, 2011). Similarly, a 37% reduction of allergenicity was observed for wheat gliadins after DBD plasma application (Lee and Koo, 2019). In contrast, milk allergens (casein, β-Lg, and α-La) were unaffected (Tammineedi et al., 2013).

In general, proteins can suffer denaturation, carbohydrates suffer disruption of hydrogen bonds, while lipids are subject to oxidation through unsaturated bonds (Figure 4).

4.2 Ingredients

Unlike the effect of plasma treatment on single components, with intrinsic interactions, the use of flours, which contain several constituents, becomes more complex. The interactions can be inter and intra component, and the results must be carefully analyzed. The main applications of NTP in starchy flours (Pal et al., 2015; Sarangapani et al., 2016) and protein flours (Bußler et al., 2016) are shown in Table 3.

4.2.1 Cereal Flours

Data in Table 3 show predominance in the use of DBD over the other types of plasma generators, generally using ambient air for the most effective ozone generation (O_3). The interest in O_3 is due to its oxidative properties, which have been presented as an alternative to chemical oxidants (such as ascorbic acid, azodicarbonamide, and potassium bromate) to promote the functionalization of wheat flours, aiming to strengthen the gluten network. More details about these studies are described below.

Table 3. Modifications in flour properties after application of non-thermal plasma.

Plasma device	Process conditions	Sample	Modification	References
DBD[1]	Gas: Air Time: 60 and 120 s Power: 40 and 90 W Voltage: 15 and 20 V	Refined Wheat flour	↑ Lipid oxidation, ↑ Gluten strength	(Bahrami et al., 2016)
RF[2] Low pressure	Gas: Air Time: 5, 10 and 15 min Power: 30, 40 and 50 W Pressure: 0.15 mbar Frequency: 13.56 MHz	Parboiled rice flour	↑ Water and oil absorption capacity; Antioxidant activity and total phenol content ↓ Swelling power due to amylopectin degradation and the presence of proteins and lipids ↓ Amylose content	(Sarangapani et al., 2016)
DBD	Gas: Air Time: 5 and 10 min Voltage: 60 and 70 kV 45 % relative humidity Samples packed with polyethylene terephthalate with film sealing	Long grain rice flour	↑ Lightness (L*) ↑ Syneresis by the effect of oxidation ↑ Peak viscosity (RVA)	(Pal et al., 2015)
		Short grain rice flour	↑ Transmittance about the control, but a reduction of this parameter along the storage period ↑ Swelling power ↑ Syneresis compared to long grain rice flour ↓ Peak intensity in X-ray diffraction	
DBD	Gas: Air Time: 5 and 10 min Voltage: 60 and 70 kV 45 % RH Samples packed with polyethylene terephthalate with film sealing	Refined Hard wheat flour	↑ Elasticity ↑ Oscillatory rheological parameters (elastic module G' and viscous module G")	(Misra et al., 2015)
		Refined Soft wheat flour	↑ Peak time (time of optimal dough development) ↑ Modifications were attributed to the ozone formed in the plasma	
NS[3]	Time: 30 and 45 min	Refined Wheat flour	↑ Color improvement ↑ Dough stability and dough strength ↑ Total and specific volume in breads ↓ Hydration capacity	(Menkovska et al., 2014)
DBD	Frequency: 50 Hz Time:5, 15 and 30 min	Wheat grain (*Triticum aestivum*)	Uniform growth of the seedlings	(Dobrin et al., 2015)

DBD	Voltage: 20 and 24 kV Frequency: 6 and 10 kHz Time: 5 to 35 min	Wheat germ	↑ Little effect on germination rate ↑ Inactivation with increased voltage and time ↓ Not valid after 25 minutes of treatment ↓ Some of its activities were recovered during storage	(Tolouie et al., 2018)
DBD	Gas: Air Time: 10 min under stirring (350 rpm) Parameters: 8.8 kV, 3.0 kHz	Pea flour	↑ Water solubility ↑ Viscosity cracking Structural (tryptophan) and compositional modifications of proteins	(Bußler et al., 2015)
DBD	Gas: Air Time: 1, 2.5, 5, 10 and 15 min Parameters: 8.8 kV, 3.0 kHz Quantity: 4.75 g under stirring (350 rpm)	Insect flour	↑ Absorption capacity of oil ↓ Higher protein solubility at pH4 than at pH 10 ↓ Water binding capacity of the protein fractions	(Bußler et al., 2016)
DBD	Gas: Air atmospheric Voltage: 80 kV Time: 10 – 30 min	Whole wheat grain and Refined wheat flour	↑ Paste Viscosity (RVA) ↑ Depolymerization of starch	(Chaple et al., 2020)

[1]DBD: Dielectric Barrier Discharge; [2]RF: Radio Frequency; [3]NS: Not specified; ↑: increase; ↓: decrease. Source: Authors.

4.2.2 Wheat flour

Wheat flour is one of the most used raw materials for producing bread and pasta, which require a strong gluten network to generate products with better technological quality. Therefore, most studies have been performed using refined flours, although some have used whole grain and whole wheat flour. The changes in refined wheat flour observed by Bahrami et al. (2016) and Misra et al. (2015) were mainly the lipid oxidation and improved gluten quality due to the formation of disulfide bonds between glutenin subunits, resulting in increased dough strength, higher viscoelasticity, and gas retention.

In contrast, Misra et al. (2015) studied wheat flour with different protein contents and sample packaging, contributing to greater O_3 retention. Bahrami et al. (2016) used no primary packaging and a shorter treatment time (60 and 120 s), aiming to minimize the side effects of the process, such as the formation of lipid oxidation products. The results proved that plasma technology could be efficient in making changes in the technological properties of wheat flour, and its parameters can be adjusted to reduce undesirable effects. However, it was not clear whether lipoproteins were formed in the samples' lipid oxidation and starch oxidation.

The gluten-forming proteins belong to the gliadin and glutenin classes (Cauvain, 2015) and contain many subunits of different sizes, structures, and conformations (Chiang et al., 2006). The α-helix, β-sheet, β-turn, and random coil structures, when together, constitute the secondary structure of wheat gluten (Bock and Damodaran, 2013).

Misra et al. (2015) reported a significant effect of the treatment time and the tension applied for the treatment of two types of flour (weak and strong) on the secondary structure of gluten. The researchers observed a decrease in β-sheets and an increase in α-helix and β-turns in the treated samples. The β-spiral structure, composed of consecutive β-sheets, is reported to be the structural element that contributes to the dough viscoelasticity (Wellner et al., 2005). An increase in the helical structural (α-helix) in wheat gluten can indicate a possible increase in the hydration capacity of gluten. Regarding the weak wheat flour (flour with low protein content, low gluten elasticity, and high gluten extensibility), an increase in α-helix along with β-sheet conformation was observed. These results indicate an ordering of the protein molecule and increased hydrogen bonding strength in weak wheat flour. Thus, the secondary structure of the protein became more stable in this plasma-modified flour (Misra et al., 2015).

On the other hand, Menkovska et al. (2014) reported a decrease in the wet gluten content due to the lower hydration capacity of gluten. In addition, the authors reported that the plasma treatment for 30 or 45 min allowed obtaining bread with higher specific volume and lighter crumbs compared to bread made with unprocessed flour.

Bahrami et al. (2016) reported no significant changes in total protein levels of wheat flour, while flour subjected to the most intense treatment (20 V for 120 s - DBD) showed significant changes in the distribution of higher molecular weight glutenin. These results confirmed the study of Misra et al. (2015), who reported that the changes in dough rheology were due to increased disulfide bonds between glutenin subunits. Therefore, molecules with large cysteine appear to be more sensitive to NTP treatment.

All these studies indicate that plasma technology can be used to modulate the functionality of wheat flour. It is well known that all rheological properties studied for wheat flours are quality indicators of the final product. However, many factors are involved, once the effects of plasma on starch and/or damaged starch, pentosans, minerals, lipids, and other proteins such as albumins and globulins are not known, which may have undergone reactions that can affect the quality of the final product.

4.2.3 Rice flour

Sarangapani et al. (2016) studied the application of NTP with atmospheric air at low pressure to treat parboiled rice flour and reported some modifications, including increased water and oil absorption capacity, and higher total phenol content, and antioxidant activity. According to the authors, the increased water absorption capacity may be due to the higher hydrophilicity from the action of reactive species on the sample surface. The increased oil retention capacity - related to the physical trapping of lipids within the starch structure - may have been favored by the formation of cross-linked bonds. There are many variables in the parboiling process, with significant changes in nutrients. Thus, the plasma-associated modification is studied only for comparison purposes.

Regarding the content of total phenols and antioxidant activity, Sarangapani et al. (2016) also found an increase in the parboiled rice flour treated for 5 minutes regardless of the power used. The release of phenolic compounds from glycosidic components and the formation of compounds with shorter chain lengths may be responsible for increasing total phenol content. However, increasing the treatment time (10 and 15 minutes) with increasing power (30 and 50 W) led to the degradation of these compounds due to free radicals from plasma, which led to the reduction of both phenolic content and antioxidant activity.

Pal et al. (2015) studied the treatment of long- and short-grain white rice flour with NTP and observed a reduction in swelling power due to starch depolymerization, which may also have been influenced by the presence of proteins and lipids once proteins compete for water while lipids form a complex with amylose. The formation of the amylose-lipid complex may also have contributed to the reduced transmittance of the starch paste of these rice flours during the 5-day storage time. For both samples, the degree of oxidation of the molecules affected the starch gel, forming a weak gel, and the high degree of depolymerization provided an increase in syneresis.

4.2.4 Flours from other sources

Due to their nutritional contribution, several types of flour have been subjected to NTP to improve their functionality, allowing its use as an ingredient in various foods. Bußler et al. (2015) used DBD plasma to modify the functional properties of different fractions of pea flour (*Pisumsativum* 'Salamanca') and reported that the treatment induced a pH reduction to 3.4. According to Sakiyama and Graves, (2009), the pH reduction is due to acidification by generating nitric acid and nitrous acid (via NO to NO_2). The authors reported that fluorescence emission intensity led to structural and/or compositional changes in the proteins with increasing treatment times.

Bußler et al. (2016) used semidirect plasma at atmospheric pressure on insect meal (*Tenebrio molitor*) and observed that in addition to the effective decontamination, the protein solubility at pH 4 decreased to a minimum of 54%, the water holding capacity decreased from 0.79 to 0.64 gwater / g, and the oil binding capacity increased from 0.59 to 0.66 goil / g.

It is worth noting that these changes in different flours reported in several studies are only interesting from a technological point of view, when applicable, once the reduction of water or oil absorption is not always required for the development of the products.

5. Innovations

Consumers have driven innovation in the food industry as they are increasingly seeking foods with fewer chemical additives and subject to processes with less environmental impact (Cullen et al., 2017; Sarangapani et al., 2018).

The search for patents using NTP technology in cereals has shown great potential for future studies. The possibilities of using NTP in the cereal processing chain, particularly the wheat processing chain, are shown in Table 4. The presence of pests is one of the problems encountered in the wheat production chain. The physical method, such as drying, is the most used, which can also be combined with chemical and biological methods. Current patents using NTP for grain treatment have focused on the improvement seed quality (Changyong et al., 2019; Xiuwu, 2019), increase in germination potential (Jiangang et al., 2015; Xiuwu, 2019), and the increase in seed yield at planting (Changyong et al., 2019; Jiangang et al., 2015; Ling et al., 2018; Xiuwu, 2019).

In the wheat milling industrial sector, Akira et al. (1996) used plasma generated at high frequency, low pressure [HF

(LP)] device, and argon and oxygen gases to replace flour conditioning additives that aim to accelerate ripening. The results showed that the argon-generated NTP was effective in reducing undesirable odors and oxidation of the wheat flour, as well as improving the specific volume of the pancakes compared to pancakes produced with the untreated flour, indicating that the pancakes produced with the untreated flour were effective that the treatment improved the wheat flour properties.

The scarcity of patents filed shows that exploiting NTP technology in the wheat production chain may be a promising approach, especially in the milling and processing stages.

6. Regulatory Aspects

Although there are no regulatory standards for the plasma treatment of foods worldwide, there is a guide elaborated by the Senate Commission on Food Safety of the German Research Foundation (Cullen et al., 2017). Within the jurisdictions of the United States and Europe, this new technology can be employed after approval of the Environmental Protection Agency (EPA), Food and Drug Administration (FDA), United States Department of Agriculture (USDA), and European Food Safety Authority (EFSA), showing that NTP poses no risk to the consumer and the environment (Keener et al., 2018).

7. Future Prospects

The current review presented only laboratory studies. It is worth mentioning that some process variables may limit the use of NTP on a large scale, including the small amount of sample treated, the form of treatment (layered material, within packages, etc.), type of sample (liquid, dry, granular material), and the process parameters (continuous or non-continuous). There are few studies on the process conditions, plasma source, and the technological benefits provided to the plasma-treated materials (Hanliang, 2017; Jiangang et al., 2015; Ling et al., 2018), thus impairing the use of this technology.

Table 4. Existing patents related to cereals, wheat flour and non-thermal plasma.

Plasma Device	Process conditions	Samples	Modifications	References
NS[1] Vacuum	Gas: Helium Time: 12 to 18 s Power: 60 to 100 W	Rice and wheat seeds	↑ Yield for different varieties of hybrid and brown rice ↑ Germination potential of rice varieties ↓ Cadmium in rice and wheat samples	(Ling et al., 2018)
NS Vacuum	Gas: Not specified Power: 10 to 25 W/8 to 18 s for seeds with thinner hull; 220 to 290 W/15 to 30s for seeds with thicker hull	Seeds with different hull densities	Improvement of seed characteristics (does not specify what improvement was achieved)	(Hanliang, 2017)
DBD[2]	Gas: a mixture of N_2/atmospheric air; Atmospheric N_2, atmospheric air or atmospheric O_2 Gas flow rate: 0.01 to 1.2 L/min Time: 1 to 60 s of treatment of the material Power: 300	Cereal lignocellulose	↑ Enzymatic efficiency, as the process facilitates the access and enzymatic saccharification of lignocellulose ↓ Energy consumption Simplicity and ease of operation Non-polluting	(X. Xu et al., 2016)
HF[3] Pressure 100 kPa	Gas: Argon and Oxygen Gas flow rate: 100 mL/min Time: 15, 30, 40 and 60 min Power: high-frequency microwaves and radio waves, 2.45 GHz, at 1.35 Kw	Wheat flour	↑ Specific volume of the pancakes that used the experimental flour ↓ Degree of oxidation of the flour	(Akira et al., 1996) (expired in 2019)
MW[4] Atmospheric pressure	Gas: Atmospheric air Power: 500 to 800 W Times: 3 to 10 s	Rice seeds	↑ Germination potential of rice seeds ↑ Flowering period ↑ Resistance to seed diseases	(Changyong et al., 2019)
NS	Power: 77 to 208 W Times: 70 to 250 s	Wheat seeds	↑ Germination ↑ Water absorption capacity ↑ Resistance to drought ↑ Weight of 1000 grains ↑ Selenium and riboflavin contents ↓ Plant diseases	(Xiuwu, 2019)
MW Vacuum	Gas: Helium Power: 300 W Times: 25 to 35 s	Rice seeds	↑ Vitality of the grains ↑ Germination rate ↑ Harvest Yield	(Yongxin et al., 2015)

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			↓ Seed growth stress ↓ Process cost	
DBD	Gas: Helium Power: 60 to 140 W Times: 15 to 20 s	Seeds of wheat, rice and oilseeds	↑ Yield ↑ Germinative power ↑ Seed vigor	(Jiangang et al., 2015)
NIR[5] Vacuum	Gas: Helium Power: up to 500 W Times: 15 to 20 s	Wheat seeds	↑ Germinative ability; ↑ Root growth ↑ Seed yield ↑ Plant size ↑ Seed production per plant	(Xin et al., 2016)

[1]NS: Not specified; [2]DBD: Dielectric Barrier Discharge; [3]HF: High Frequency; [4]MW: Microwave; [5]NIR: Non-ionizing radiation; ↑: increase; ↓: decrease. Source: Authors.

Standards and regulations are required for the industrial use of this technology, which requires guidance and approval from competent agencies (Cullen et al., 2017). In addition to the regulatory aspects, increasing the investment and development time of the technology is key to increasing the use of NTP. However, due to the process versatility, many innovative applications are expected in the future, with new studies integrating the scale trials in laboratories and full-scale production.

The use of larger area discharges without compromising the uniformity of the plasma and the treated food is still a challenge. Furthermore, the large-scale industrial application requires product and process efficacy, efficient control, validation of the application, and consumers acceptance of the product (Cullen et al., 2017). According to Cullen et al. (2017), Puač et al. (2018), and Sarangapani et al. (2017), some steps need to be elucidated, including the formation mechanisms and types of radicals generated, the interactions with nutrients, bioactive compounds, plasma, and packaging, the process optimization for each food matrix, and the methods of dissemination and the benefits to consumers.

8. Final Considerations

The present review presented various studies on NTP technology to modify isolated food constituents and/or more complex ingredients such as flours, seeds, and lignocellulosic material.

It was found that the dielectric barrier discharge and ambient air are the most common source and the gas used to produce NTP for food purposes. DBD is one of the safest processes and can operate over a wide range of gas pressures, providing a homogeneous discharge even at a low gas flow rate. Atmospheric air is a good alternative due to its low cost and easy acquisition, essential for full-scale production. Plasma chemistry is also prominent as the source for obtaining highly reactive species, such as ROS and RNS.

Therefore, research should be focused on new pilot studies about the repeatability and reproducibility of parameters to enable the technological, nutritional, and sensory evaluation and storage stability of processed products. Improvements in the production layout to produce greater amounts of treated material are also necessary. Furthermore, in vitro and in vivo studies are required to ensure the use and consumption of plasma-treated foods and ingredients and sensory studies to evaluate possible impacts on the sensory properties of the processed products.

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