Cold plasma surface treatments to prevent biofilm formation in food industries and medical sectors

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Abstract

Environmental conditions in food and medical fields enable the bacteria to attach and grow on surfaces leading to resistant bacterial biofilm formation. Indeed, the first step in biofilm formation is the bacterial irreversible adhesion. Controlling and inhibiting this adhesion is a passive approach to fight against biofilm development. This strategy is an interesting path in the inhibition of biofilm formation since it targets the first step of biofilm development. Those pathogenic structures are responsible for several foodborne diseases and nosocomial infections. Therefore, to face this public health threat, researchers employed cold plasma technologies in coating development. In this review, the different factors influencing the bacterial adhesion to a substrate are outlined. The goal is to present the passive coating strategies aiming to prevent biofilm formation via cold plasma treatments, highlighting antiadhesive elaborated surfaces. General aspects of surface treatment, including physico-chemical modification and application of cold plasma technologies, were also presented.

Key points

- Factors surrounding pathogenic bacteria influence biofilm development.
- Controlling bacterial adhesion prevents biofilm formation.
- Materials can be coated via cold plasma to inhibit bacterial adhesion.

Keywords Biofilm · Cold plasma · Antiadhesive · Surface treatment

Introduction

The presence and growth of bacterial species on many natural and synthetic surfaces leading to the formation of biofilms are a major problem affecting different fields, especially food and medical sectors (Abdallah et al. 2014; Ciofu et al. 2015; Galié et al. 2018). As a matter of fact, the biofilm formation is a complex process characterized by a succession of steps already described by different works (Stoodley et al. 2002; Donlan and Costerton 2002; Abdallah et al. 2014). In the biofilm formation system, the bacteria switch from a free floating (Planktonic) state where they function as individuals, to a sessile state where they function as communities. Adsorption, or reversible adhesion of bacteria, is the first and essential step in biofilm formation on abiotic surfaces. The adherent bacteria in this step are not all initiated to be in the differentiation mechanism leading to biofilm formation, and many can actually escape from the surface and return to the planktonic lifestyle (Stoodley et al. 2002; Khelissa et al. 2019). This phase is reversible and promoted by numerous non-covalent interactions. Indeed, when microorganisms reach a certain distance from the surface (between 2 and 50 nm), bacterial adhesion is induced, by non-covalent forces, such as Van der Waals, acid–base, and electrostatic interactions. The resulting force of these interactions allows bacterial adsorption on the support. Moreover, environmental conditions surrounding the bacteria, like temperature, pH, and organic matter may influence the bacterial and surface properties leading to a modification in the bacterial adhesion behavior. At this stage, exopolymeric substances (EPS) are secreted by bacteria that become irreversibly attached to the surface. Adhered
bacteria multiply and form microcolonies while secreting an extracellular matrix containing a mixture of polysaccharides, nucleic acids, proteins, and lipids. Then, the biofilm maturation process leads to development of a mature and complex biofilm. The final step of a biofilm lifecycle is the detachment or dispersion of bacterial cells from the biofilm and colonizing new surfaces. This step has a key role in the dissemination of bacteria and the spread of infections (Fig. 1).

These persistent pathogenic structures are responsible for a variety of nosocomial infections and foodborne illnesses (Abdallah et al. 2014; Veerachamy et al. 2014). Moreover, the plans of disinfections carried out by hospitals and industrials do not remove completely the biofilms formed on the equipment, especially the resistant ones (Kostakioti et al. 2013). These plans aiming to restrain the biofilm formation have a negative environmental footprint and an important economic impact on these fields (Pace et al. 2006). Indeed, it is of significant relevance to find solutions to get rid of bacterial contamination and biofilm formation. The development of surfaces that limit the formation of biofilms is an aim that researchers and industrials have been trying to reach. Several investigations have been carried out to elaborate effective, harmless, and stable antiadhesive and antimicrobial coatings in order to prevent biofilm structuration. In the passive approach aiming to prevent biofilm formation, surfaces have an antiadhesive property towards pathogenic microorganisms. In this approach, the surface’s chemical and physical aspect modification is investigated. The resistance to bacterial adhesion is owed, on those films, to the interactions between bacteria and modified surface. The surface properties are adjusted to inhibit bacterial adhesion mechanisms. Indeed, physical properties like surface wettability, roughness, and surface charge are adapted according to the desired characteristics (Rodrigues 2011; Guo et al. 2016). One of the possible ways to develop these modified surfaces is cold plasma treatments (Saulou et al. 2012).

Cold plasma treatment is a valuable coating technology since it imparts a homogenous and stable surface modification. Several studies have proven the stability of plasma-deposited coatings such as organosilicon, oxides, or fluorocarbon films. Cold plasma also permits researchers to tailor the functionalization of the surface according to the properties needed. Regarding the prevention of biofilm formation, surfaces elaborated by plasma can act passively with antiadhesive character towards bacteria, and actively with antimicrobial properties depending on the molecules and parameters used in the coating elaboration (Chan 1993; Saulou et al. 2012). In this review, the plasma technology for coating elaboration is highlighted. A special attention is given to the developed surfaces with antiadhesive property for its importance as a first step in preventing biofilm formation. Indeed, this research presents the cold plasma coating strategies, for biofilm formation prevention, as a passive approach.

**Factors influencing the bacterial adhesion to a substrate**

The factors influencing the initial adhesion of bacteria to substrates involves multiple parameters. The adhesion factors are linked to the microorganism, the target surface, and
the surrounding environment (An and Friedman 1998). The relative impact of these characteristics depends on the microbial strain studied (Katsikogianni and Missirlis 2004). However, these factors must be carefully considered in order to develop effective strategies in the prevention of microbial colonization. The most significant factors influencing this adhesion were discussed hereafter and summarized in Fig. 2.

A solid surface is systematically covered with a layer of organic contamination because of the air pollution (Corn 1961). After being cleaned, surfaces such as glass, plastics, or metallic materials are prone to re-contaminate themselves in order to acquire a thermodynamically stable state. During manufacturing processes, material surfaces might be contaminated by micro-particles. Since materials are frequently in contact with other material types, drugs, or foods during production processes (Bohinc et al. 2016). This contamination may affect the bacterial adhesion to the substrate. Indeed, the presence and accumulation of organic soil on surfaces affect its surface roughness and the bacterial adhesion behavior (James et al. 2017). A material adsorption with macromolecules like organic and inorganic compounds is called the “conditioning film.” Moreover, the surface modification by grafting molecules via multiple coating techniques, using classical chemistry or plasma technology, permits the elaboration of various coating types. All those reactions result in a significant modification of the physical and chemical characteristics of the support like its roughness, hydrophobicity, and charge.

It influences positively or negatively microbial adhesion. In addition, when microorganisms are detached from a surface by mechanical stress, the constituents of their membrane might remain adhered to the surface and promote other microbial attachment (Donlan 2002; Lorite et al. 2011; Chouirfa et al. 2019).

The chemical type of surface encountered by the microorganisms strongly influence the development of the biofilm, more specifically the first stage of bacterial adhesion. For example, Verheyen et al. (1993) showed that Staphylococcus aureus adheres more preferentially to the metal 316L steel than to the polymeric surface poly(L-lactide) due to the differences in chemical composition and polarity of these surfaces. Moreover, a study by Alam and Balani (2017) investigated the adhesion force of Staphylococcus aureus on different biomaterial surfaces. The UHMWPE surface (ultra-high molecular weight poly ethylene) showed a weak adhesion force (~4 nN) whereas stainless steel showed strong adhesion force (~15 nN) owing to their surface roughness and surface energy.

Surface roughness is one of the most discussed parameters influencing the bacterial adhesion. Indeed, it seems that microbial adhesion can be impacted positively or negatively, depending on the bacterial size and on the surface topography that includes several parameters like the width/depth of the “micro-cracks” and the presence of stripes. Thus, the presence of cracks and “Micro-cracks” increases the contact area and can promote adhesion mechanisms by protecting

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Fig. 2 Factors influencing bacterial adhesion on a substratum
bacteria from hydrodynamic shear stress and chemical disinfection agents. In fact, a study by Dantas et al. (2016) analyzed the relationship between the bacterial adhesion and surface roughness of acrylic polymethyl methacrylate substrates. This investigation showed that the increase in surface roughness of the samples was directly related to an increase in bacterial adhesion of Streptococcus sanguinis. On the same wave, Hage et al. (2021) studied a plasma-modified stainless steel by organosilicon monomer 1,1,3,3-tetramethyldisiloxane mixed with oxygen, using a nitrogen flow microwave post-discharge plasma polymerization process. The influence of cold plasma parameters on coating characteristics, coated surface structure, and attachment of Salmonella Enteritidis cells was investigated. The results demonstrated that the surface structure affected the rate of bacterial adhesion. Indeed, rough coatings did not repel Salmonella Enteritidis as the numbers of adhered cells on these surfaces ranged from 30 ± 4 to 65 ± 4 bacteria per microscopic field. However, the smoother coatings exhibited an anti-adhesive nature as the number of adhered cells was almost nil on these surfaces. In addition, Whitehead et al. (2005) showed that titanium surfaces, with holes of similar or larger size than those of the bacteria S. aureus (diameter ~ 0.5–1 µm) and Pseudomonas aeruginosa (diameter ~ 1–3 µm), as well as Candida albicans yeast cells (diameter ~ 2 µm), offer a better adhesion. For other authors, roughness has no influence on biofilm initiation and it inevitably grows after a period of time (Vanhaecke et al. 1990; Rodriguez et al. 2008). In addition, Flint et al. (2000) observed no correlation between arithmetic roughness of AISI 304L stainless steel surfaces (Ra between 0.5 µm and 3.3 µm) and the attachment of heat-resistant staphylococci. However, they have shown an interesting adhesion for a value of Ra equal to 0.9 µm, suggesting a trapping of microorganisms linked to their size. Moreover, according to other studies, the increase of surface roughness reduces the contact surface between the substrate and the microorganism when its size is higher than the surface roughness, promoting cell detachment (Boulangé-Petermann et al. 1997). The surface roughness parameter effect on bacterial adhesion is frequently associated with the surface wettability which is a very important parameter.

The hydrophobicity and surface free energy of a material are recognized to influence bacterial adhesion (Quirynen et al. 1994; Subramani et al. 2009). The non-specific physico-chemical interactions are constituted of Van der Waals forces, electrostatic and acid–base interactions, which characterize the surface free energy of a substratum (Grivet et al. 2000). Results from several investigations that relate surface wettability to bacterial adhesion are conflicting (Grivet et al. 2000; Oh et al. 2018). However, it is known that according to the bacterial adhesion thermodynamic model, hydrophobic bacteria preferentially colonize hydrophobic substrates and vice versa (Mabboux et al. 2004; Wassmann et al. 2017). A recent study investigated the effects of surface texture and roughness on the bacterial adhesion. Staphylococcus aureus adhesion behavior, towards a bio-ceramic joint implants with different roughness grades (Ra 205–1.1 nm) and surface texture (uniform and unidirectional textures), elaborated via polishing technologies, was studied. The results showed that when the surface roughness reduces from the sub-micron scale to the nano-scale level, the surface state gradually changes from hydrophobic to hydrophilic. In this case, it turns unsuitable for the adhesion of Staphylococcus aureus which is hydrophobic. Moreover, the anchoring points for bacterial adhesion gradually disappear, and then the bacterial-surface bonding strength weakens. It was concluded that the preparation of a smooth surface and the elimination of unidirectional surface textures can inhibit the initial adhesion of Staphylococcus aureus on those surfaces, reducing the occurrence of implant-related infections (Lu et al. 2020). Another study aimed to investigate bacterial adhesion on different ceramic and titanium surfaces, and analyzed the relationship between surface hydrophobicity and surface roughness defining the predominant factor for bacterial adhesion on each material. Results showed that the variations in surface roughness did not show any differences in the adhesion of Staphylococcus epidermidis. However, higher surface roughness showed an increase in Streptococcus sanguinis adhesion. In contrast, for Staphylococcus epidermidis, the bacterial adhesion rates detected were higher on the hydrophobic surfaces than on the hydrophilic surfaces but not for Streptococcus sanguinis. The adhesion potential of Streptococcus sanguinis was higher on the ceramic surfaces than on the titanium surfaces while no such preference was detected for Staphylococcus epidermidis. Indeed, both surface wettability and roughness can impact the adhesion behavior of bacteria on biomaterials. In this context, the predominant factor is dependent on the bacterial species (Wassmann et al. 2017).

Several studies have established that the surface charge of materials plays an important role during cell adhesion (Behrens and Grier 2001; Palmer et al. 2007; Choi et al. 2017). A recent study demonstrated the relationship between the surface charge and the bacterial adhesion. Indeed, Guo et al. (2018) elaborated layer-by-layer films via branched polyethylenimine and synthesized polyanions bearing either alkylcarboxylic or poly (ethylene glycol) side chains. This development was carried out with control over wettability and surface charge parameters. Adhesion test results showed that the Escherichia coli and Staphylococcus aureus adhesion was guided by surface charge and wettability.

Several factors linked to the microorganism properties influence the bacterial adhesion. Investigations showed that when the microbial concentration increases, the number of adhered cells gets higher, until the surface is completely covered (Piette and Idziak 1992). In addition,
the presence of primary microorganisms colonizing a surface can facilitate the occurrence of other microorganisms (Beloin et al. 2008). This phenomenon, known as “co-aggregation” has been highlighted in a study of the oral cavity presenting adhered bacteria to the teeth (Whittaker et al. 1996). Other studies have demonstrated the existence of this cooperation in food, agriculture, and biomedical sectors (El-Azizi and Khardori 1999). The biochemical composition and the architecture of the bacterial cell surface (presence of proteins, fimbriae, flagella, exopolymers, peptidoglycan in Gram-positive bacteria, and lipopolysaccharides in Gram-negative bacteria) contribute to the adhesion of microorganisms to the substrates. For example, the fimbriae contain a high proportion of hydrophobic amino acids, which leads to the establishment of hydrophobic interactions with the material (Donlan 2002). The flagella allow the bacterium to be mobile and play an important role in the early stages of adhesion by counteracting electrostatic repulsion forces (Pratt and Kolter 1998). Lipopolysaccharides (LPS) are present in the wall of Gram-negative bacteria, and more specifically the carbohydrate part (O antigen) of these LPS, give the cell hydrophilic properties. As a result, mutants of Pseudomonas fluorescens unable to produce LPSs adhere in greater numbers to hydrophobic substrates (Williams and Fletcher 1996). The teichoic acids, specific components of Gram-positive bacteria, affect their adhesion mechanism since they give the cell a negative surface charge. Indeed, Gross et al. (2001) demonstrated that a mutant of Staphylococcus aureus, whose teichoic acids do not contain D-alanine, was unable to adhere to the polystyrene, due to the increased negative surface charge compared to the D-alanine-containing strain. Surface proteins, frequently referred to as “adhesins,” are also strongly involved in the bacterial adhesion to surfaces via hydrophobic interactions (Flint et al. 1997). For example, Cucarella et al. (2001) identified in Staphylococcus aureus a protein called BAP (“Biofilm Associated Protein”). They have shown that the bacteria producing this protein strongly adhered to plastic surfaces (polystyrene, polyvinyl chloride) while mutants BAP-deficient adhered poorly to the two tested surfaces. Moreover, several studies showed that polysaccharides, present on the bacterial surface, are involved in their initial attachment. Polymers excreted by bacteria (EPS) induce a reinforcement of adhesion to the support, making it irreversible (Atabek and Camesano 2007). Moreover, regarding physico-chemical properties of the material affecting the adhesion, the hydrophobicity, and the surface charge of the cell wall play also a major role in the adhesion mechanism (Palmer et al. 2007). Indeed, those properties are linked to the composition of the cell surface that are influenced by the growth rate and the physiological state of the bacterial strain. The adhesion of a bacterial strain to a receptor substrate permits the development of a different perception of its environment (Kimkes and Heinemann 2020). Specific genes are then over- or under-expressed according to the new bacterial needs. Thus, the genes coding for flagella are inhibited, since the microorganism turned to the sessile state (Kuchma and O’Toole 2000). Otherwise, the expression of genes involved in quorum sensing or EPS production or parietal proteins increases (Prigent-Combaret et al. 1999; O’Toole et al. 2000).

Environmental characteristics affect directly the bacterial adhesion to a substrate. The increase in contact time between the microorganism and the support induces a reinforcement of established linkages (Nejadnik et al. 2008).

The temperature of the surrounding environment influences microbial colonization, because growth temperature is maximal for a so-called optimal temperature, specific to each microorganism. In addition, numerous studies have proven the influence of the ionic strength of the medium on microbial adhesion, through the electrostatic interactions established between the microorganism and support (Bos et al. 1999; Poortinga et al. 2002).

The pH of the surrounding environment has an influence on bacterial growth and on their surface physico-chemical properties. The pH value has also an impact on the surface charge of the substrate, especially in the case of metals such as stainless steel, for which the oxidation state depends on the pH (Palmer et al. 2007). The presence of surfactants in the bacterial environment modifies the solid/liquid and microorganism/liquid interfaces, influencing cell adhesion and detachment, as noted by McEldowney and Fletcher (1986). Moreover, the presence of nutrients, such as carbon and nitrogen, affects the bacterial metabolism and has an influence on the bacterial surface properties, thus on the initial adhesion (Strevett and Chen 2003; Nitschke and Silva 2018). Hydrodynamic conditions influence bacterial adhesion. Indeed, when the flow regime is laminar or slightly turbulent, the boundary layer at the material/liquid interface is thick and the adhesion of microorganisms depends on their ability to penetrate it according to their mobility and size and the flow velocity. Moreover, depending on the Brownian motion, gravity or convection movements can favor the initial adhesion of microorganisms. Otherwise, when the regime is turbulent, the numerous eddies can facilitate contact between bacteria and surface. However, the decrease in the thickness of the hydrodynamic boundary layer reduces the interaction time between the cell and the substrate. Indeed, the establishment of weak bonds hinders the irreversible attachment of the bacteria (Donlan 2002; Palmer et al. 2007; Nejadnik et al. 2008). In conclusion, microbial adhesion on surfaces is a multifactorial phenomenon constantly evolving over time. It involves many different parameters linked to the substrate, the microorganisms, and the suspending medium. Controlling all of these parameters
is a challenge, for both industrial and biomedical fields, in which the microbial colonization of surfaces is at the origin of particularly negative impacts.

**Cold plasma technologies**

The treatment of surfaces with plasma techniques permitted to develop surfaces according to desired properties. Plasma has been defined as a gas that is partially or fully ionized into charged particles and neutral molecules (Moreau et al. 2008). It is regarded as the fourth state of matter and obtained when gases are excited into energetic states by radiofrequency, microwaves, or electrons from a hot filament discharge (Bogaerts 1999; Chu et al. 2002). Concerning the types of plasmas, they are divided into two main categories: Thermal plasmas and non-thermal plasmas also called atmospheric cold plasma, cold atmospheric plasma, or simply cold plasma (Mandal et al. 2018). The thermal and cold plasmas can be defined according to the conditions in which they are created.

Cold plasma technologies provide a uniform modification of the whole surface with less material degradation than several wet chemical treatments (Karam et al. 2013). In fact, the imparted functionalization type can be controlled by plasma gas selection like Ar, N₂, O₂, H₂O, CO₂, or NH₃, and by experimental conditions such as pressure, power, time, or gas flow rate (Kang and Neoh 2009). Plasma-surface modification (PSM) allows changing the chemical composition and properties such as wettability, hardness, chemical inertness, and biocompatibility of material surfaces (Nedøla et al. 2017). There are two very interesting things about cold plasma technology. Firstly, cold plasma is source of elevated temperature electrons at ambient conditions. Secondly, the cold plasma, when interacting with an atmospheric or controlled environment, elaborates many reactive components. Indeed, cold plasma is produced at low levels of power and pressures, with absence of localized thermodynamic equilibrium; it is indeed defined as non-equilibrium plasma. The provided energy breaks the gas into a several reactive species, following other reactions such as ionization, excitation, and de-excitation. Moreover, the specific procedure carried out during cold plasma production determines the orientation for application alongside composition of reactive species (Taccogna and Dilecce 2016). Those reactive species can be applied for many chemical reactions in different domains of science (Gorbaney et al. 2016). Such plasma is of particular interest technically and industrially because they do not require extreme conditions that might change the material properties (Wiesemann 2014).

There are options concerning the delivery of the generated plasma species to the substrate. Firstly, the direct exposure in which the substrate is directly exposed to the plasma discharge itself. It can be the splash of a plasma jet or the field between two electrodes. The other option is the indirect or remote exposure that requires placing the surface at a distance from the plasma discharge. The long-lived components interact with the surface after recombination with several induced species (Sarangapani et al. 2018).

It is therefore important to first define the technological trajectory for cold plasma generation, which comprise those developed under reduced pressure and induced by atmospheric pressure.

Cold plasma generated under reduced pressure is known as microwave plasma directed by electromagnetic waves generated at frequencies of hundreds of MHz. In contrast to methods presenting electrodes, the microwave discharges are produced via a magnetron supplying microwaves, guided by a coaxial cable, into a process chamber. The irradiation is then absorbed and heat is produced (Fig. 3A) (Isbary et al. 2013). The inelastic collisions generate ionization reactions. The absence of electrodes in microwave plasma is considered beneficial and can be easily restarted in air. Moreover, the gas required in this technique is low comparing to the large quantities of reactive species released. In addition, this plasma is limited in space and its application to wide zones is non-workable in comparison with plasma jets. Plasma jet is a particular configuration discharge. In general, the active region is characterized by a flow of auxiliary gas, producing a burning small jet of ionization waves and active particles. High power and local practicability are profitable in those plasma types called jet, plasma torch, plasma needle, or plasma pen (Fig. 3B) (Scholtz et al. 2015).

Cold plasma induced at atmospheric pressure includes dielectric barrier discharges (DBD) (Fig. 3C), corona discharge (Fig. 3D), radio frequency plasma (Fig. 3E), and gliding arc discharge (GAD) (Fig. 3F). DBD plasma is induced by an alternating current emitted when two metallic electrodes are retained apart using a dielectric material at a discharge gap ranging from 100 mm to a few centimeters. The dielectric impedes the generation of sparks due to charges movement. The DBD technique encloses the application of different gases, reduction of gas flow rate, a uniform discharge activation over several meters, and is characterized by a good adaptability since the electrode geometries employed can be varied. However, prevention and security measures are requested since DBD needs high ignition voltages of 10 kV (Cullen et al. 2018; Fg et al. 2017).

Corona discharge plasma is elaborated surrounding sharp pointed electrodes that contain substantial electric field for developing the ionization energy of arbitrarily created electrons to the expedition for gas, molecules, or atoms. High voltage is needed to generate this discharge. It is not expensive and simple to employ. Corona discharges
are carried out for surface treatment and fighting microbial contamination. However, it is constrained to heterogeneous diminutive areas. Otherwise, radiofrequency plasma is generally produced when a gas is localized within an oscillating electromagnetic field, achieved by distinct electrodes maintained outside the reactor or by an induction coil. Comparable to microwaves, this class of plasma are produced at frequencies ranging hertz to megahertz (Scholtz et al. 2015; Fg et al. 2017; Mandal et al. 2018).

GAD are elaborated in a reactor comprising two or more diverging metallic electrodes working at a high potential difference. In this technique, an inlet gas, composed of humid air, is pumped into the discharge gap between the electrodes. This leads to the formation of an arc in between the narrowest inter-electrode area, which is directly blown away into the diverging area by the inlet gas. Generally, GAD develops both thermal and cold plasmas, depending on the conditions. This technique is applied for both liquid and surface treatments. It is employed for chemical contaminant (e.g., organic solvents, industrial wastes) degradation and for antibacterial effect (Patil et al. 2016; Dasan et al. 2017).

Cold plasma is considered in this review since its suitable for surface coating elaboration. Indeed, cold plasma technology applied for deposition and coating production is divided hereafter in three approaches that will be highlighted: the plasma functionalization, polymerization, and plasma-induced grafting.

### Approaches for cold plasma surface modification

Plasma functionalization approach concerns a plasma treatment leading to the incorporation of new functionalities on the material surface. In fact, different reactive and inert gases are used alone or in combination in order to generate active plasma species on polymers (Karam et al. 2013). The active plasma species bombard the atom surface and break the covalent bonds between them, conducting to hydrogen abstraction and creation of surface radicals. Radicals have the potency to react with the gas-phase species to form several chemically active functional groups on the surface (Bogaerts et al. 2002). The type of the formed functional groups rely on the gas used for functionalization as well as the experiment conditions such as the excitation type, reactor geometry, applied power, time, temperature, flow rate, and gas pressure (Chan et al. 1996; Chu et al. 2002). Oxygen
plasma guides to the set-up of a variety of oxygen functional groups like carboxylic acid groups, peroxide groups, and hydroxyl groups on the polymer surfaces (Fig. 4) (Chan et al. 1996; Sanchis et al. 2006). Otherwise, carbon dioxide plasmas can form hydroxyls, ketones, aldehydes, esters, and carboxyl groups on a selected surface (Desmet et al. 2009). Nitrogen and ammonia plasmas introduce primary, secondary, and tertiary amines, as well as amides on the material surface (Tušek et al. 2001; Kull et al. 2005). However, the plasma functionalization technique is believed to be disadvantageous regarding its inability to form a single functional group and the instability of the changes induced to the surface. This ageing procedure is due to post-plasma oxidation, reorientation of polar groups on the surface towards the bulk, diffusion of molecules with low molar mass to the polymer, and the environmental conditions like humidity that causes the absorption of water molecules by hydrophilic coating, resulting in the disturbance of the surface properties (Schönherr et al. 2000; Upreti et al. 2006; Siow et al. 2006; Tsougeni et al. 2009) (Fig. 5).

Another approach is the induced-plasma chemical grafting. It is a technique where there is an association of plasma functionalization and classic chemistry (Karam et al. 2013). In this technique, a polymer surface is exposed to a cold plasma of a gas such as oxygen, helium, or argon to activate the surface and create free radicals (Bogaerts et al. 2002). The material is then exposed to atmospheric air that oxidizes the radicals, resulting in the formation of peroxide functions that will allow the grafting of monomers in a further step (Legeay et al. 2006; Ma et al. 2007). The immersion in the monomer solution is carried out under heating (50 °C). Indeed, the heating enhances the decomposition of peroxide and oxygen is avoided in the solution since it can stop the reactions (Gupta et al. 2001; Chu et al. 2002; Legeay et al. 2006). This method prevents the ageing effects since grafting chemicals onto the surface increases the stability of this treatment (Kang et al. 1996; Goddard and Hotchkiss 2007).

In addition, the plasma polymerization (Fig. 6) is essentially a plasma-enhanced chemical vapor deposition (PE-CVD) procedure which is an effective technique to elaborate organic thin coatings on a material, and offering
proper control over the film character (Hamedani et al. 2016). This approach uses electrical energy for generating a plasma that turn on the reaction by transmitting the energy of its compounds to the precursors leading to free radical creation followed by polymerization process (Vasudev et al. 2013). This polymerization is chemically and physically different from conventional polymerization involving radicals and ions even if the same monomers are used in both polymerization techniques (Chu et al. 2002).

Plasma polymerization technique presents remarkable properties, like chemical stability, because of its highly cross-linked nature, the variety of monomers and materials that can be used in this technique, and the film uniform thickness. Moreover, plasma polymerization technology has diversity of potential applications that make it a spot of interest for industrials and researchers (Chu et al. 2002; Hamedani et al. 2016). With this depositing process, the selected substrates can be covered with various types of coatings starting with gaseous precursors. In general, a short-chain monomer is cross-linked, fragmented, rearranged, and polymerized under the influence of the plasma to generate a long-chain polymer (Dessaux et al. 1998) (Fig. 7).

Plasma polymerization method permits the elaboration of thin films using organic monomers that polymerize on the surface, thanks to this technique, while other conventional methods do not permit their polymerization.

Plasma physico-chemical modification to surface

In this section, the main physico-chemical changes introduced to the surface after plasma surface modification (PSM) are mentioned. Biomaterial surface properties are usually described in terms of surface energy (wettability), chemistry, topography, roughness, and electrostatic charge. PSM with reactive gas leads to the introduction of active chemical function species and then, the modification of chemical properties (Károly et al. 2019).

The surface chemistry controls the charge and the hydrophobicity of a material. Thus, it has a direct effect on the cell...
adhesion to plasma-modified surfaces. The nature of introduced chemical groups depends on the gas used to generate plasma (Amani et al. 2019).

The surface wettability is an important parameter that significantly changes after PSM. Surface wettability before and after PSM is usually characterized by water contact angle (θ) measurements. θ is used to measure the surface hydrophilicity using organic (non-polar) or polar solvents by placing a droplet of liquid on a dry surface (Iqbal et al. 2019). Generally, the lower the θ, the more hydrophilic is the surface. Surface energy can be altered by PSM techniques to strongly influence cell adhesion (Rezaei et al. 2014). The free radicals generated during the plasma process react with the environmental O₂ leading to the formation of polar groups (such as -OH, -COOH). Those functions provide more hydrogen bonding, resulting in a decreased contact angle, and hence lowering of θ (Goddard and Hotchkiss 2007). Furthermore, it was found that PSM increased the hydrophilicity and surface energy without altering the bulk properties of the materials (Sharma et al. 2002; Govindarajan and Shandas 2014; Jaganathan et al. 2015).

Surface topography and roughness are considerably modified after PSM on the micron and nanometer scale, resulting in microorganisms' behavior modification that could interact with plasma-treated materials (Jing et al. 2007a, 2007b). In fact, it has been reported that PSM of biomaterials have the ability to regulate cell functions such as proliferation, differentiation, and apoptosis (Ito 1999). Oxygen plasma treatment has been used by Ha et al. (1997) to modify the surface characteristics of polyether ether ketone (PEEK). They showed that PSM created spherulitic surface irregularities of PEEK characterized by an increased roughness. Moreover, it has been shown that plasma-treated polyurethane (PU) had homogeneous surfaces after treatment, which did not lead to significant changes in PU film topography (Sanchis et al. 2007).

Furthermore, plasma treatment affects the hardness and elastic modulus of treated polymer surfaces owing to the effects of densification and cross-linking (Shi et al. 2001; Powles et al. 2005). Shi et al. (2001) study showed that the nano-hardness and elastic modulus of plasma-treated ultrahigh molecular weight polyethylene UHMWPE doubled, whereas the wear resistance coefficients was significantly enhanced by a factor of three compared with the untreated samples. They concluded that improvement of wear resistance can be mainly attributed to ion bombardment–induced cross-linking, and thus surface hardening. Surface charge is determined mainly by zeta potential measurements based on the quantification of electrophoretic mobility of materials in solution, depending on the polarity (charge) of the absorbed counter ions in the electric double layer, and the ionic concentration of the solvent (Khorasani and Mirzadeh 2007). Basically, PSM results in the introduction of different charged species (anionic and cationic), functional groups, and free radicals, on the surfaces of materials. These created species are directly involved in the modification of the original zeta potential of initial surface. In fact, many studies have demonstrated the impact of PSM on material surface charge. Shao et al. (2017) used atmospheric-pressure dielectric barrier discharge for the modification of epoxy material surface and refine the dissipation of surface charge aiming to reduce the accumulation of surface charge. Another study demonstrated that cold plasma treatment participates in charging organic surfaces. In this investigation, the surface density of the electrical charge of lentil seeds and pepper and polymers like polystyrene, polyethylene, poly(methyl methacrylate), and polycarbonate was established experimentally (Shapira et al. 2018). Moreover, it has been reported that the electronegativity of PVC showed a high increase from −9 to −22 mV after PSM (Khorasani and Mirzadeh 2007; Khorasani et al. 2008). In addition, in human surgery, plasma surface polymerization has been applied to set up non-thrombogenic cardiovascular implant surfaces based on the electrostatic interaction between the negatively charged plasma proteins and cationic coating due to the introduction of -NH₂ and -COOH groups (Lassen et al. 1992).

Cold plasma applications

Researchers are continuously fascinated by the applications offered by plasma science. Thanks to the low heat capacity of cold plasma, its production cost efficiency and the diversity of its applications, a very high interest in plasma technology is prevailing. Figure 8 illustrates the main cold plasma applications.

The potential employment of thermal plasma processing technology comprises a large range of activities, such as the extraction of metals, the refining of metals, the production of fine ceramic powders, spray coatings, and the destruction and consolidation of hazardous wastes (Taylor and Pirzada 1994; Samal et al. 2010).

Otherwise, in material science, cold plasma is applied for surface property modifications, for example, in the production of computer chip (Weltmann et al. 2018). Plasma polymer film application includes anti-adhesion surfaces, humidity sensors, electrical resistors, optical filters, protective coatings, chemical barrier coatings, and scratch resistance coatings that have been successfully applied on optical lenses. In environmental sciences, it finds application in air and water purification (Foster 2017), for example, it can be applied for pesticide degradation in water (Pankaj and Keener 2017).

In biomedicine fields, cold plasma technology is applied for teeth and skin therapy, sterilization of medical equipment, and the development of coatings for antibacterial...
purposes (Popelka et al. 2012; Hoffmann et al. 2013). This technology is also employed for wound healing and disease treatments (Pankaj and Keener 2017). Concerning the application of cold plasma in virus inactivation, a recent study showed that cold atmospheric plasma with argon plasma gas was efficient in the inactivation of coronavirus SARS-CoV-2 on several surfaces like metal, plastic, and cardboard. These results proof the interesting potential of cold plasma in the prevention of virus transmission for different surfaces that are generally in frequent contact with individuals (Chen et al. 2020). SARS-CoV-2 infection involves recognizing and linking to the human angiotensin-converting enzyme 2 receptor on cells via the receptor-binding domain (RBD) of the spike protein, and perturbation of this mechanism can effectively inhibit SARS-CoV-2 proliferation. Plasma-activated water impact on coronaviruses has been investigated by Guo et al. (2021). Indeed, in this study, pseudoviruses with SARS-CoV-2 protein S were employed as a model, and plasma-activated water effectively inactivated pseudovirus infection by inhibiting of the protein S. RBD was employed to investigate the molecular particularities. Results showed that the binding activity of RBD was effectively knocked out by plasma-activated water via modification of RBD. These demonstrations present a new opportunity for the engineering scientific, and medical sectors.

Moreover, non-thermal plasma can be used in food industries for the development of coated surfaces with anti-adhesive and antibacterial properties aiming to fight biofilm formation (Ma et al. 2012). In this field, cold plasma is also used in the packaging process as well as in food production to reduce the risk of bacterial contamination since it can be applied for decontamination and toxins degradation. Indeed, it can help for products shelf-life extension and improve the packaging integrity (Karam et al. 2013). Plasma technology can be carried out to design functional films with different biocidal agents, including quaternary ammonium salts, silver, or antibiotics (Wang et al. 2004; Bruckert and Weidenhaupt 2010). In addition, it is applied in surface modification, functionalization, reticulation, and thin film deposition of polymer surface (Pankaj and Keener 2017). The plasma-based techniques are profitable for different purposes; they can be applied for coating/depositing, cleaning/sterilization, and modification of surface chemistry of substrates. Plasma treatment can also be used as a pretreatment to other surface modification techniques (Sabir et al. 2009; Joshy et al. 2019).

**Cold plasma treatments of materials for preventing bacterial adhesion**

Aiming to fight biofilm formation in medical and food fields, many studies were carried out to produce anti-bacterial and anti-adhesive modified surfaces via cold plasma treatments. Among the several materials applied in those fields, stainless steel is a predominant metal used in various application where hygiene is primordial, including food industry and medical sectors (Fouda and Ellithy 2009; Sun et al.
Moreover, titanium alloy is used for dental implants, medical equipment, and in food and pharmaceutical manufacturing areas (Agripa and Botef 2019). In addition, polyethylene terephthalate (PET) is a polymer commonly used in biomedical and food applications (Perez-Roldan et al. 2014). Other polymers like polyamide, polydimethylsiloxane (PDMS), silicone, and polypropylene can be applied for their physico-chemical properties in those sectors. This section highlights the strategies carried out by researchers to elaborate antiadhesive films by cold plasma treatment on the materials surfaces mentioned above. Figure 9 summarizes the main directions followed in coatings elaboration and Fig. 10 shows the general surface properties modifications after plasma treatment.

Several studies demonstrated that modifying surfaces with hydrophilic and non-charged polymers resulted in reduced cellular, protein, and bacterial attachment on different surface types (Finch 1994; Du et al. 1997; Sofia et al. 1998; Zhang et al. 2001). Indeed, it has been established that surfaces deposited with poly(ethylene glycol) (PEG) are able to reduce bacterial adhesion and biofilm formation. In effect, an investigation of coated PET and polyamide with PEG of different molecular weights using a SiCl₄ cold plasma treatment via the creation of C–Si–Clₓ functionalities permits the covalent linkage of PEG macromolecules through a condensation reaction mechanism. Indeed, these coating showed significant inhibition of attachment and biofilm formation by Listeria monocytogenes and Salmonella enterica sv. Typhimurium compared to unmodified PET and polyamide (Dong et al. 2011). In addition, a research analyzed the coating of PEG-like compounds, 1,4,7,10-tetraoxacyclododecane ether and tri(ethylene glycol) dimethyl ether, onto stainless steel by a cold-plasma enhanced technique. The coatings were more hydrophilic and less rough than the uncoated stainless steel. Biological testing on a mixed culture of Staphylococcus epidermidis, Salmonella Typhimurium, and Pseudomonas fluorescens revealed a reduction in the bacterial adhesion and biofilm formation (Denes et al. 2001). In another study, the PEG-like compound, di(ethylene glycol) vinyl ether was deposited onto stainless steel surface via radiofrequency–plasma processes. These deposited films...
showed a stable chemistry and a more hydrophilic character and a decrease in roughness values in comparison with bare stainless steel. These new characteristics led to an effective anti-adhesive behavior of the coatings towards *Listeria monocytogenes* strains (Wang et al. 2003). Plasma treatments can be used as preliminary preparation for surface grafting. A research investigated the antifouling characteristics of grafted plasma-modified PET surfaces. In fact, two different gases, oxygen and helium, were employed to create superhydrophilic surfaces with various surface chemistries. Oxygen reactive gas used in plasma treatment increases the oxygen groups and enhances the hydrophilic character on the surface (Krstulović et al. 2006). Researchers demonstrated an antibiofilm activity of 3D printed polyactic acid petri dishes treated surfaces. Atmospheric pressure plasma was employed for the polymerization and deposition of acrylic acid. Plasma polymerization caused an increment of oxygen polar groups (C—O and O-C = O) producing a hydrophilic character of the coatings. This hydrophilic character played an essential role in *Pseudomonas aeruginosa* and *Staphylococcus aureus* biofilm reduction (Muro-Fraguas et al. 2020).

In a study, stainless steel surfaces were treated with (3-aminopropyl) triethoxysilane (APTES), tetraethyl orthosilicate (TEOS) and acrylic acid (AA) via non-equilibrium atmospheric plasma. An anti-biofilm efficient activity was detected against *Listeria monocytogenes* and *Escherichia coli* strains. *Listeria monocytogenes* registered the best results, with surfaces coated with a base of APTES and functionalized by TEOS or AA, reduced biofilm formation by 45% and 74%, respectively, in comparison with uncoated SS. Surface characterization showed that the coating with the highest anti-biofilm activity had higher hydrophilicity and lower surface roughness. This results showed that the development of a hydration layer prevented the bacterial adherence, an effect that seems to be increased by low temperature conditions and when the wettability of the strains is enhanced (Fernández-Gómez et al. 2020). Indeed, the environmental conditions, physicochemical characteristics of the surface, and bacterial cell envelope affects the adhesion behavior.

Moreover, surface cross-linking was carried out; thanks to helium inert gas resulting in the increase in surface wettability, an important factor influencing the bacterial adhesion (Gheorghiu et al. 1997; Papakonstantinou et al. 2007). Surfaces were then grafted with PEG, Pluronic F108, Pluronic F68, mixed solutions of Pluronic and surfactant like sodium taurodeoxycholate nonaethylene glycol, monodecyl ether, and hexadecyltrimethyl ammonium bromide. Those coated surfaces showed effective antifouling properties (Perez-Roldan et al. 2014). Moreover, in another study using radiofrequency plasma polymerization, stainless steel surface was deposited with ethylenediamine (EDA), a hydrophilic monomer, in different glow discharge parameters (radiofrequency discharge power of 20–80 W with exposure time of 10 min). The modification of plasma conditions showed different efficiencies of the anti-adhesive character of the coatings tested towards *Enterobacter sakazakii*. The optimal condition showing 99.74% of attachment reduction was plasma modification by EDA at 45 W and for 10 min (Sen et al. 2012). Another coating on stainless steel by plasma technique was elaborated aiming to obtain antiadhesive properties. Effectively a silver nanoparticle component film was coated onto stainless steel to weaken the adhesion power of the model yeast *Saccharomyces cerevisiae*. The coating was done under cold-plasma parameters, mixing silver sputtering and RF glow discharge. The anti-adhesive properties of the coating were attested with shear-flow-induced detachment trial (Guillemot et al. 2008). Plasma material treatment is a strategy to improve coating quality and affect biological response at the surfaces of biomedical devices specifically polymeric materials. A research shows the time-dependent effects of a non-thermal plasma on the surface of polypropylene polymeric implants. Findings suggest that plasma exposure enhanced resistance to *Escherichia coli* adhesion. Bacterial adhesion decreased after 1 min of plasma treatment (\(p > 0.048\)) whereas after 10 min and 20 min of plasma treatment, the bacterial attachment rate, in comparison with the 1-min rate, was reduced by half (\(p < 0.001\)). These results imply that the time exposure of a surface to plasma treatments affects its chemical properties and behavior towards microorganisms (Gd et al. 2020). Furthermore, Lin et al. (2020) elaborated effective anti-adhesive coatings towards *Escherichia coli*. In fact, PDMS polymeric surfaces were modified via an atmospheric plasma-induced polymerization with polyvinyl alcohol (PVA) and then immobilized by a zwitterionic polymer (2-methacryloyloxyethyl phosphorylcholine, MPC). Those surfaces were developed for wound dressing application in biomedical fields. The super-hydrophobic character of those modified surfaces inhibited bacterial adhesion. Titanium (Ti) alloys, often used in medical fields, do not repel bacterial attachment. In an investigation, the production of radicals was carried out via non-thermal atmospheric pressure plasma jet on Ti surfaces aiming to modify its chemical properties. Bacterial adhesion of *Streptococcus sanguinis* to Ti was significantly inhibited after plasma treatment (\(p < 0.05\)) compared to unmodified surfaces. In this work, the anti-adhesive effect was generated by carbon cleaning that was dependent on the gas type used on the titanium surfaces (nitrogen > ammonia and air, \(p < 0.05\)) (Lee et al. 2017). In a recent study, an acrylate-containing coating was elaborated on titanium surfaces through atmospheric pressure plasma treatment of 2-hydroxyethyl methacrylate, a liquid precursor. The obtained hydrophilic coatings decreased *Staphylococcus aureus* and *Escherichia coli* adhesion. These surfaces were produced for dental implant antiadhesive effectiveness (Buxadera-Palomero et al. 2021).
Moreover, in another recent study, superhydrophobic surfaces with antibacterial properties were developed. Surfaces were elaborated using trichloro(1H,1H,2H,2H-perfluorooctyl)silane (TPFOS) and titanium dioxide nanoparticles (TiO$_2$-NPs) as chemical modifiers. The virgin PVDF membrane was pre-treated using PEG-co-PMAA, followed by plasma treatment, to increase the __COOH and __OH groups on the outer layer and enable coordinate bond formation on the membrane surface to TiO$_2$. TPFOS was selected to impart a superhydrophobic character to the titanium surface. Results showed that the PVDF/PP-PT/Ti/Si (polyvinylidene difluoride/coated PEG-co-PMAA-plasma treated/titanium nanoparticles/perfluoroctyl silane) developed membrane registered a larger contact angle of ~152° and a better cleanability than that of the pristine PVDF membrane. Plasma treatment caused the increase in membrane porosity via the polymer ablation mechanism. Treated surfaces showed excellent antibacterial properties when tested against \textit{Staphylococcus aureus} and \textit{Escherichia coli} (Sinha Ray et al. 2021).

Plastic polymers elaborated using monomeric silicone-based chemicals provide excellent chemical and thermal resistance and remarkable optical, electrical, and biomedical properties (Inagaki et al. 1985; Schwarz et al. 1998; Bashir and Bashir 2015). The plasma polymerized organosilicon films can be employed as protective coatings in microelectronics (KRYSZESKI et al. 1979). Organosilicon are also selected for the protection of metals from corrosion (Fracassi et al. 2003). The most employed organo-silicone monomers include TetraMethyldiSilOxane (TMDSO) (Deng et al. 2015), TetraMethylSilane (TMS) (Fonseca et al. 1993), VinylTriMethylSilane (VTMS) (Bonnar et al. 1999), HexaMethyldiSilOxane (HMDSO) (Morent et al. 2009), and HexaMethylDiSilAZane (HMDSZ) (Huang et al. 2015) containing Si, H, C, O, or N atoms (Gaur and Vergason 2000). Organo-silicon monomers are used in industries because they are non-toxic components and they do not generate harmful species during processing. Thus, they can be applied without any special safety considerations (European Commission, Directorate General for Health & Consumers 2014).

Among the many monomers which have been employed in plasma polymerization, the organosilicones were recognized to form coatings of special properties. Indeed, organosilanes have at least one carbon-silicon bond, which is very stable and nonpolar. In the procedure of plasma polymerization of organosilicon, the film polymerized on the substrate surface starts to grow when the long-lasting reactive particles, flowing from the microwave discharge, had enough energy to break the chemical bonds and create free radicals implied in the film formation (Callebert et al. 1994; Karam et al. 2013). The deposition zone, where CRNP appears as a yellow afterglow, is a non-ionized zone mostly formed with reactive species like nitrogen atoms in the ground electronic state N(4S), free radicals, and electronically excited N$_2$ triplet states and vibrationally excited N$_2$ in the ground electronic state (Jama et al. 1997; Quédé et al. 2002; Esbayou et al. 2018).

Organosilanes are one of the most versatile molecules that are widely used in coatings and surface modifications cold plasma technologies. Generally, organosilane-coated surfaces exhibit an increase in low surface energy and its hydrophobic characters. These types of films ensure the inhibition of bacterial growth without releasing toxic products of low molecular mass into the environment (Kregiel and Niedzielska 2014). Furthermore, an investigation showed that following plasma-assisted surface silanization, the anti-adhesive properties of coated surfaces increased due to the decrease in roughness properties (Savela et al. 2012; Kregiel et al. 2013). In another research, organosilicone-based films were developed on 316L stainless steel, by atmospheric pressure plasma spraying (APPS) of HMDSO. This plasma coating showed an antifouling character and anti-adhesive properties towards \textit{Staphylococcus aureus} (Zouaghi et al. 2018). Moreover, Kregiel and Niedzielska (2014) developed polyethylene surfaces, activated by plasma processing and modified with active organosilanes. Those coatings exhibited anti-adhesive properties towards \textit{Aeromonas hydrophila}.

Some studies have examined the antimicrobial activity of organosilanes with active biocidal groups chemically linked to their chains. Indeed, Fortuniak et al. (2011) tested the biocidal activities of polysiloxanes linked with antibacterial quaternary ammonium salt (QAS) groups. These polysiloxanes were linear polydimethylsiloxanes with 20% siloxane units substituted at silicon by 3(dimethyl-n-octylammonio) propyl chloride or 3(dimethyl-n-hexadecylammonio) propyl chloride and terminated by silanol functions at both chain ends. Those polymers were cross-linked and added to a silicone substrate. The biocidal test resulted in thousand-fold reduction of \textit{Staphylococcus aureus} after 15 min of contact with the substrate containing 20 wt % of this polymer. This study permitted to conclude that polysiloxane-based surfaces can be used as pretreated substrates to link biocidal agents and develop antimicrobial surfaces. Moreover, another research aimed to study how the modification of silicone elastomer and polyvinyl chloride surfaces, commonly used in the water industry, can reduce the attachment of \textit{Aeromonas hydrophila}, a pathogenic bacterium that has the ability to attach to pipe materials. Silicone elastomer and polyvinyl chloride surfaces were activated via cold plasma with reactive organosilanes by coupling silanes with the native material. Those coated surfaces exhibited higher anti-adhesive and anti-microbial characteristics in comparison to the bare surfaces (Kregiel 2013).
Conclusion
Cold plasma is an innovative technology experiencing an increased popularity since it shows applications at several sectors. In food and medical sectors, pathogenic bacteria adhere on surfaces and form resistant biofilm which are responsible of many infectious diseases. This review presents general aspects of cold plasma surface modifications. It also highlights plasma-coated surfaces designed to inhibit and prevent bacterial attachment on surfaces. However, cold plasma technology requires additional investigations in eco-toxicity, ageing characteristics, coating effectiveness with time, and the interaction mechanisms between the bacteria and plasma coated surface.

Author contribution CJ, NEC, and HA conceived the outline of this review. MH and SK contributed the figures. All authors contributed to literature searches and the writing of the manuscript. All authors have read and approved the manuscript.

Declarations

Ethical approval Not applicable, since the work does not involve any study with human participants or animals.

Consent to participate Not applicable.

Consent for publication All co-authors have given their consent to publish this manuscript.

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