Production of antibacterial coatings through atmospheric pressure plasma: A promising alternative for combatting biofilms in the food industry

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Abstract

One of the main problems in the food industry is the formation of biofilms on food contact surfaces. These bacterial communities show high resistance against the commonly used disinfectants, which makes them difficult to eradicate causing economic losses and threatening the quality of the products and the health of consumers. Several studies have reported the use of atmospheric pressure plasma technologies to provide antibacterial properties to a wide range of materials through the deposition of coatings that either avoid the initial attachment of bacteria to the surface or kill the attached bacteria before the mature biofilm is formed. These technologies avoid the use of extreme pressures and temperatures during the deposition process, thus preserving the properties of the substrate, which makes them interesting for their potential application in the production of anti-biofilm food contact materials. This paper reviews different approaches that use atmospheric pressure plasma technologies to combat bacterial colonization and biofilm formation on materials of relevance for the food industry. Three types of approaches are identified and their suitability in the food industry is discussed.

Keywords: Antimicrobial coatings; Biofilm; Atmospheric Pressure Plasma; Food industry

1. INTRODUCTION

Microbial persistence, defined as the prolonged survival of microorganisms colonizing certain habitats, constitutes an important matter of concern for food industries. It can lead to important economic losses due to the corrosion of metal surfaces and cross contamination of the food products, which may have a negative impact on their sensory properties and pose the human health at risk because of the presence of pathongens on the food (Larsen et al. 2014). A frequent way of microbial persistence in food processing environments is the formation of biofilms on surfaces and equipments. Biofilm formation comprises several stages: (i) initial attachment, (ii) proliferation/maturation, and (iii) dispersion. Firstly, planktonic bacteria adhere to a conditioned surface and produce an exopolysaccharide matrix that facilitates bacterial aggregation and colonization. Then the biofilm undergoes maturation, thereby acquiring a complex three-dimensional structure. Bacteria within biofilms are more difficult to eradicate because they have greater resistance to antimicrobials and to inactivation techniques than bacteria in planktonic state. Furthermore, their proliferation and dispersion is facilitated because bacteria from mature biofilms can detach, return to a planktonic state and spread to other areas of the production facility, leading to biofilm dispersal and continuous contamination of new habitats and foods (Chen et al. 2013; Dong et al. 2007; Ermolaeva et al. 2015; Kovalova et al. 2016). For instance, long-term persistence (more than ten years) has been reported for *Listeria monocytogenes* on food contact surfaces in the processing environments of several products, such as cheese (Fox et al. 2011; Lomonaco et al. 2009), fish (Wulff et al. 2006) or meat (Nesbakken et al. 1996; Ojeniyi et al. 2000).

In food industries, biofilm control strategies usually involve the use of chemical treatments such as detergents and disinfectants. However, those chemical agents have several drawbacks, as they are usually not environmentally friendly, may pose a risk to the health of the consumer if they contaminate the food, and can damage the surfaces and materials where they are used. Therefore, their use must be strictly controlled and there are maximum allowed concentrations in food that must not be exceeded (Gabriel et al. 2016; Li 2016). In addition, since biofilms are very difficult to be completely removed, the best solution to biofilm-related contamination events is, in many cases, the disposal of the contaminated equipment and products, which involves a high economic cost to the industry (Guo et al. 2013; Smet et al. 2018).

For all these reasons, great efforts are being dedicated to the development of novel alternative strategies to prevent biofilm formation on food products and food contact surfaces, either by avoiding the initial adhesion of bacteria or by killing them before the mature biofilm is generated.

Regarding the sanitation of food products, some authors have proposed the combined use of different sanitizers as a way to improve their antibacterial effects while reducing their required concentrations. For instance, Chen et al. (2019) studied the disinfection of organic broccoli sprouts using a combination of lactic acid and a low concentration of sodium hypochlorite. They observed that such combination achieved greater reductions in the colony forming units of *Listeria innocua* than using either of the two sanitizers individually. They concluded that, whereas lactic acid might induce oxidative damage to the bacteria and disrupt their cytoplasmic membrane, its sanitizing effect could be synergistically promoted by sodium hypochlorite.

Low concentration electrolyzed water has also been proposed as a proper sanitizer for food products and food contact surfaces because of its disinfection efficacy, quick production, low corrosivity and low cost (Liu et al. 2018). The main factors that determine the sanitizing effects of electrolyzed water are pH, oxidation-reduction potential (ORP) and free available chlorine (FAC) concentration (Zhang et al. 2018b). According to the literature, an increase in the pH is related with a decrease in the FAC concentration and in the ORP (Rahman et al. 2010). On the one hand, high ORP can inflict oxidative damage to the bacterial cells, leading to necrosis. On the other hand, the regulations establish limits for the FAC concentration of chlorine-based sanitizers, which is why electrolyzed water can be used only in low concentrations (Zhao et al. 2019).

Zhang and Yang (2017) studied the effects of electrolyzed water (4 mg/L FAC), citric acid (0.6% w/v) and hydrogen peroxide (H_2O_2, 1% w/v) in the sanitation of fresh-cut lettuce, using the three sanitizers individually and in different combinations. They concluded that the combination of electrolyzed water and H_2O_2 was a promising approach for the sanitation of fresh-cut lettuce because it achieved an efficient reduction in the microbial loads without compromising the sensory properties and the quality of the product. In a more recent study on the sanitation of fresh organic lettuce, Zhao et al. (2019) analyzed the bactericidal efficacy of low concentration acidic electrolyzed water (4 mg/L FAC) and levulinic acid (3% v/v). They found that combining both sanitizers was more effective at reducing the microbial load than using either of them individually. Furthermore, the lettuces that were treated with a combination of electrolyzed water and levulinic acid showed no significant change in their quality during 7-day storage.

Aiming at the sanitation of food contact surfaces, Zhao et al. (2017) studied the effect of neutralized electrolyzed water (pH: 7, 4 mg/L FAC) and ultrasound (37 kHz, 80W) on stainless steel coupons. They used both sanitation approaches individually and in combination, and evaluated their effectiveness for the inactivation and detachment of *Escherichia coli*, *Pichia pastoris* and *Aureobasidium pullulans*. The combined method was the most effective approach of the study, showing greater antibacterial effect than using either the electrolyzed water or the ultrasound. While the electrolyzed water showed strong bactericidal effect, the ultrasound promoted bacterial detachment, improved the dispersion of HClO in the aqueous media and could facilitate the penetration of HClO into the detached cells.

Among other novel anti-biofilm approaches for food contact materials, surface modification and coating deposition are particularly interesting. They can change the characteristics of a surface without altering the main properties of the substrate, therefore limiting microbial adhesion and biofilm formation (Bhatt et al. 2015; Chen et al. 2013; Dong et al. 2007; Guo et al. 2013; Moreno-Couranjou et al. 2018; Sardella et al. 2016).

Surface modification plasma-based techniques are a promising approach to control biofilm formation. When energy is provided to a gas, it can be ionized and generate plasma, which is composed of ions, neutrals and electrons. Depending on the conditions in which the ionization is performed, plasmas with different characteristics and suitable for various applications can be generated (Bárdos and Baránková 2010). According to the temperature of their components, plasmas can be classified as equilibrium (or thermal) and non-equilibrium plasmas. In an equilibrium plasma, the electrons and the heavy particles (ions and neutrals) are at the same temperature (of around 10^4 K). In a non-equilibrium plasma, the temperature of the heavy particles (300 – 1000 K) is at least one order of magnitude lower than the temperature of electrons (Winter et al. 2015). Therefore, it is possible to generate relatively cold plasmas near to room temperature that can be applied to highly temperature sensitive substrates (Merche et al. 2012). Surface modification plasma-based techniques have become popular because they are capable of depositing homogeneous coatings on a great variety of substrates by means of a solvent-free process that only requires a reduced amount of chemical precursors. This fact makes them economically and environmentally beneficial in comparison to other traditional surface modification techniques

(Hernández-Orta et al. 2018) and makes it possible to tune the surface properties by varying the parameters of the plasma generation process. Regarding agriculture and food applications, Bourke et al. (2018) noted that the ability of cold plasma technology to operate at low temperatures and its high antimicrobial efficacy, make it an advantageous technology for the treatment of raw materials, end-products and equipment, showing a great capacity of enhancing the microbiological safety of foods while maintaining their quality characteristics. Usually, the generation of cold plasma for surface treatment is performed under low pressure conditions. However, such conditions involve the use of a complex and expensive vacuum technology. Recently, plasma systems operating under atmospheric pressure have received a great deal of attention because they can generate cold plasma without using a coupled vacuum equipment, which facilitates their implementation for in-line operation and reduces costs (Lin et al. 2010; Merche et al. 2012; Ramkumar et al. 2017; Ramkumar et al. 2018).

Some authors have reported that atmospheric pressure plasma treatments are able to inactivate biofilm-associated microbial cells, showing a synergistic effect when combined with different biocides for biofilm reduction. Indeed, Koban et al. (2013) used an atmospheric pressure plasma jet in combination with several agents (i.e. chlorhexidine (CHX), octenidine (OCT), polyhexanide (PHM), sodium hypochlorite (NaOCl), hydrogen peroxide and ethylenediaminetetraacetic acid (EDTA)), which were applied before or after the plasma treatment to reduce biofilms cultivated on titanium discs. They found that, regardless of the treatment sequence, the plasma/biocide combinations had increased antimicrobial effects in comparison to using only the plasma or the biocide agents. Similar results were obtained by Gupta et al. (2017), who studied the antimicrobial effect of an atmospheric pressure plasma jet combined with CHX in two-step treatments for the sterilization of the biofilms formed by *Pseudomonas aeruginosa* on titanium surfaces. Although these studies suggested that surface disinfection could be achieved by using lower and safer concentrations of biocides in combination with atmospheric pressure plasma, this type of approaches still rely on using chemical agents that can pose a risk to human health and the environment.

The present review focuses on the potential of atmospheric pressure plasma technologies to produce safer materials where biofilm formation is limited or controlled. As summarized in Table 1, it compiles different approaches to prevent biofilm formation: the deposition of composites containing embedded biocidal agents, the immobilization of antimicrobial agents on surfaces and the deposition of coatings that modify the surface physicochemical properties.

2. ATMOSPHERIC PRESSURE PLASMA SYSTEMS AND COATING DEPOSITION

As observed in the bibliography (Table 1), dielectric barrier discharge (DBD) systems and atmospheric pressure plasma jets (APPJ) are the most widely used sources of atmospheric pressure plasma for the production of antibacterial surfaces. In order to understand how these sources work, Figs. 1a,b depict usual set-ups of APPJ and DBD systems. Nevertheless, it is worth mentioning that these are general schematics and some variations of these models can be found in each particular study in the bibliography. Generally, both configurations employ two electrodes. One of the electrodes is grounded and the other one is excited by a high voltage source to generate an electric field between them. A flow of gas passes between the electrodes and is ionized in the electric field, thus generating plasma.

The usual set-up of APPJ systems (Fig. 1a) consists of a gun that contains two cylindrical and coaxial electrodes. The gas passes along the gun and generates a plasma jet that extends to the open air from the exit of the gun. In DBD systems (Fig. 1b), the electrodes are usually two parallel plates that are covered by a dielectric material and confined in a chamber. The space between the electrodes is fed with a flow of gas that generates plasma inside the chamber. The dielectric barrier that covers the electrodes allows the generation of a stable and uniform plasma.

The most common way to deposit coatings by means of atmospheric pressure plasma systems is through plasma polymerization. This process consists in feeding the plasma flow with an atomized liquid precursor that is based in a monomer. The interaction between the precursor and the plasma leads to the fragmentation of the molecules of the precursor and to the formation of the functional groups that determine the properties of the coating (Bhatt et al. 2015). The products of the reactions between the precursor and the plasma, as well as non-reacted precursor and reactive species from the plasma, land on the surface of the substrate where they adsorb and surface reactions take place. As shown in Fig. 1c, these reactions can result in desorption of volatile byproducts from the surface and diffusion processes (Borer 2005). Difussion is related with an increase in the mobility of the deposited material on the surface of the substrate (Amirzada et al. 2016). As a result, the material can nucleate forming clusters that coalesce into greater features, thus increasing the roughness of the coating (Fig. 1d).

The formation of the functional groups, plasma species and activation sites involved in the plasma polymerization, as well as the growth and physicochemical properties of the coatings, depend on many factors such as the characteristics of the substrate, the chemical compositions and flow rates of the precursor and the process gas, and the power set at the plasma generator. For instance, plasma polymerization of siloxane monomers is widely used for producing coatings with structural stability and flexibility (Stallard et al. 2012). Ramamoorthy et al. (2009) studied the influence of several process parameters on the characteristics of siloxane coatings obtained by plasma polymerization of tetraethyl orthosilicate using an APPJ system. As the flow rate of the precursor increased, thicker and more organic coatings were obtained. This increase in the organic content of the coatings was associated to a lower fragmentation of the precursor in the plasma when high flow rates of precursor were used, which resulted in the incorporation of higher amounts of carbon into de coatings. According to their observations, increasing the plasma power can be associated with an increase in the roughness of the coatings. They suggested that this could be due to the deposition of higher amounts of particulates that are formed by excess gas phase reactions when high power levels are used.

As in Duday et al. (2013), using an aminosilane as the precursor provides amino groups and a silica skeleton to the coatings, which favour their adhesion and stability. Furthermore, these authors compared the plasma polymerization of (3-Aminopropyl)trimethoxysilane (APTMS) by a direct DBD system with two flat parallel electrodes and a DBD afterglow plasma reactor, which is similar to an APPJ system, using N_2 as the process gas in both systems. According to their observations, the lower confinement of the DBD afterglow reactor led to a higher presence of water and oxygen-based contaminants in the deposition zone around the substrate, which caused an increase in the decomposition of the precursor and in the growth rate of the coatings. In other work by Vartiainen et al. (2005), amino groups were provided to the studied surfaces by adding NH_3 to the N_2 flow used for the generation of plasma, whereas adding CO_2 to the N_2 flow provided carboxyl groups. The activation sites corresponding to the amino and carboxyl groups immobilized the enzyme glucose oxidase when glutaraldehyde and carbodiimide were used respectively as linkers.

The nature of the process gas has an influence on the power that is required to generate the plasma, which is also related to the degree of fragmentation of the precursor during the plasma polymerization. As reported by Da Ponte et al. (2011), higher power was required to ignite the plasma using nitrogen than using helium. The authors suggested that this may have been the reason why they observed more fragmentation of their lactic acid precursor, as well as a higher loss of oxygen functionalities from their coatings, when they used nitrogen than when they used helium.

Reactive oxygen and nitrogen species that are generated in the plasma process can form compounds with antimicrobial activity, such as hydroxyl radicals (OH*), nitrogen dioxide (NO_2), peroxynitrites ($ONOO^-$ or ONO_2^-) and nitrous anhydride (N_2O_3). These species can be provided by the process gas, being oxygen and nitrogen among the most commonly used gases for the generation of atmospheric pressure plasma (Table 1), and from the surrounding atmosphere in open air plasma processes even if the process gas contains none of them (Cullen et al. 2018). For instance, hydrogen peroxide and superoxide radicals can generate hydroxyl radicals in the presence of transition metals (Yost and Joshi 2015). On the other hand, the reactions between nitric oxide (NO) and oxygen or superoxide radicals can form nitrogen dioxide, peroxynitrites and nitrous anhydride (Lu et al. 2014).

Despite the potential beneficial effects of these reactive species, they can also affect the quality of the food products. Few studies have assessed their impact in the nutritional and sensory properties of food. Furthermore, there are discrepancies among the available information, which are probably due to the variability in the experimental conditions and products used in each study (López et al. 2019). Also, according to a review by Cullen et al. (2018), the sparse data available regarding toxicity and the lack of legislation or regulatory guidance related to the use of plasma for food-related applications still pose a challenge for the implementation of plasma technologies in the food industry. Therefore, more research is necessary in order to determine which process conditions are suitable to provide antibacterial protection to specific food products without compromising their safety and quality.

3. COMPOSITES CONTAINING AND RELEASING BIOCIDAL AGENTS

One of the approaches to obtain antibacterial surfaces, therefore limiting biofilm formation, involves the deposition of a composite coating. This coating comprises a polymeric or inorganic matrix containing an embedded biocidal agent that is released when the coating comes into contact with the surrounding medium (Sardella et al. 2016) as shown in Fig. 1e. Although many different materials can be used as the matrix of the composite, plasma polymers have the advantages of showing a good adhesion onto the surface and the ability to be deposited forming low thickness coatings without altering the mechanical properties of the substrate. It is even possible to control the release rate of the antibacterial agent by tuning the characteristics of these polymeric layers (Vasilev et al. 2011). Thus, a quick loss of the antibacterial activity of the coatings due to a too fast release rate can be avoided.

Deng et al. (2015) used a three-step process to coat polyethylene terephthalate (PET) fabrics by embedding silver nanoparticles between two organosilicon layers that were deposited through atmospheric plasma polymerization of tetramethyldisiloxane (TMDSO). They observed that the coatings showed antimicrobial activity against *P. aeruginosa*, *Staphylococcus aureus* and *Candida albicans*. As the thickness of the second organosilicon layer (barrier layer) increased, lower bacterial reductions were obtained. This suggested that the increase in thickness caused a reduction in the amount of cracks and pores of the barrier layer that could reduce the release of silver ions from the nanoparticles to the medium. Thus, this characteristic could be used as a possible way to control or modulate the antimicrobial efficacy of the coatings and the durability of their antibacterial activity.

Deng et al. (2014) employed a single-step process to deposit nanocomposite films on silicon wafers by generating an APPJ in nitrogen and feeding the plasma with oxygen, TMDSO and a powder of silver nanoparticles. They identified that the metallic silver nanoparticles were embedded inside the films, whereas at the film surface the silver was present as AgO. The coatings showed antibacterial activity against *E. coli* (Gram-negative) and *S. aureus* (Gram-positive), being more effective against Gram-negative bacteria. A single-step process was also presented by Liguori et al. (2016), who used an APPJ to deposit a plasma-polymerized matrix of polyacrylic acid with embedded silver nanoparticles onto polyethylene substrates by injecting two precursors in the plasma region: (1) acrylic acid for the matrix and (2) a dispersion of silver nanoparticles in ethanol. Agar disk diffusion tests against *E. coli* revealed the formation of growth inhibition zones. These zones were attributed to silver ions that may have been released by the silver at the surface of the coatings or through the cracks that were observed by scanning electron microscopy.

Shi et al. (2011) reported a synergistic effect of electrons and UV radiation in atmospheric plasma that favours the reduction of silver and the formation of silver nanoparticles dispersed in polymer solutions. They applied atmospheric plasma treatments to polyacrylonitrile (PAN) and silver nitrate ($AgNO_3$) solutions before electrospinning them into nanofibers. After 7 days of silver release in water, the nanofibers obtained from plasma-treated solutions maintained a high antibacterial activity against both *E. coli* (Gram-negative) and *Bacillus cereus* (Gram-positive), whereas the nanofibers obtained from untreated solutions maintained only a slight antibacterial activity.

As pointed by Gupta and Xie (2018) in a recent review about the applications, toxicity and regulations of nanoparticles, these can be more toxic than lager particles because of their greater mobility. Their small size (< 100 nm) makes them able to enter the human body by ingestion or inhalation, transfer to the blood stream and reach the organs or the central nervous system. Furthermore, their release in industrial waste poses a risk of environmental contamination. Since the toxicity of nanomaterials may be affected by their specific properties, there is still limited information in this regard and their regulation is still challenging. For this reason, efforts are being dedicated to classify nanomaterials, as well as to understand and control their potential risks. For instance, the European Food Safety Authority (EFSA) is working on a guidance on risk assessment of the application of nanoscience and nanotechnologies in the food and feed chain (EFSA Sicentific Comittee et al. 2018), which gives suggestions regarding the available methods for the characterization of nanomaterials and the key parameters that should be measured. Furthermore, although silver is used as an effective biocide with activity against bacteria in a wide range of fields including biomedicine- and food-related applications, its toxicity to higher organisms such as humans is a matter of concern (Marambio-Jones and Hoek 2010).

Instead of using silver or other metallic nanoparticles, safer and more environmentally friendly alternatives can be produced by embedding organic antibacterial agents in the matrix. Natural essential oils derived from plants (Bazaka et al. 2015) and biomolecules that are naturally produced by living organisms (e.g., lysozyme or nisin) can be used to provide the composite coating with antimicrobial activity (Sardella et al. 2016). In fact, edible composite films and coatings with antimicrobial efficacy have been produced using biomolecules and essential oils (Dhumal and Sarkar 2018). Palumbo et al. (2015) used atmospheric pressure plasma generated in a DBD reactor to deposit bio-composite coatings on silicon wafers in a single step-process. These coatings consisted of an organic matrix that was obtained by plasma polymerization of ethylene, containing embedded lysozyme that was provided forming an aerosol. The authors verified the release of the embedded lysozyme into water. They also observed by an agar test against *Micrococcus lysodeikticus* that lysozyme kept its antibacterial activity after having interacted with the plasma.

4. IMMOBILIZATION OF ANTIBACTERIAL AGENTS

The amount of the active antimicrobial agent that is released with the aforementioned approaches has to be controlled in order to avoid toxicological or sensory issues that could arise by the migration of too high antimicrobial concentrations to the food products (Conte et al. 2006). Alternatively, another possible approach consists in immobilizing the antimicrobial agents onto food contact surfaces without releasing them (Fig. 1f). Thus, the antimicrobial activity against the bacteria in close contact with the surface is achieved avoiding the migration of the antimicrobial agents into the food (Conte et al. 2008). Several authors have used plasma techniques to immobilize antimicrobial peptides (Duday et al. 2013), enzymes (Conte et al. 2008; Thallinger et al. 2016; Vartiainen et al. 2005) and polymers (Tseng et al. 2009) on different relevant surfaces.

For instance, Duday et al. (2013) used organosilicon-based coatings deposited by plasma polymerization as reactive layers for the covalent immobilization of antibacterial nisin peptides on stainless steel substrates. They used APTMS as the precursor for plasma polymerization, in order to generate amino groups capable of reacting with the carboxyl groups of the peptides, thus favouring their immobilization. Two types of atmospheric plasma DBD equipment were used by these authors in order to compare the effects of two processes: (i) direct DBD and (ii) DBD afterglow. Although the coatings which were obtained by the direct DBD process had higher contents of amino groups, they seemed to be unstable when they were immersed in the peptide solutions for the covalent immobilization and showed low bactericidal activity. On the other hand, a more satisfactory compromise between the content of amino groups and the stability of the coatings was obtained through the DBD afterglow process. Thus, a 3.6 log reduction of *Bacillus subtilis* was achieved. In addition, the bactericidal properties of the DBD afterglow coatings remained after aging and washing tests.

Similarly, Vartiainen et al. (2005) used the atmospheric pressure plasma generated in a DBD reactor to activate bi-oriented polypropylene films by creating amino and carboxyl groups for the covalent immobilization of the antimicrobial enzyme glucose oxidase. The formed films completely inhibited the growth of *E. coli* and significantly reduced the growth of *B. subtilis*. Since the antimicrobial activity of glucose oxidase is usually attributed to the generation of H_2O_2, these results were in agreement with the well-known lower resistance of Gram-negative bacteria against hydrogen peroxide than that of Gram-positive bacteria.

Tseng et al. (2009) also grafted nylon fabrics with antimicrobial agents, in this case a chitosan oligomer or polymer, after activation by open air plasma. Different speeds and numbers of scans were used for the activation of the different samples included in the study. Tests using *S. aureus* as a target microorganism suggested that the antibacterial activity of the fabrics generally tended to be greater when a higher speed and a higher number of scans were used for the plasma activation. Furthermore, significantly better antibacterial effects were exhibited by the fabrics that were grafted with the chitosan polymer as compared to those grafted with the chitosan oligomer.

5. COATINGS FOR SURFACE PHYSICOCHEMICAL MODIFICATION

Atmospheric pressure plasma can be also used for the modification of the physicochemical properties of a surface without the deposition of any antimicrobial agent, being a very promising approach to combat biofilms (Fig. 2). As previously mentioned, biofilm formation implies the adhesion of planktonic cells to a surface. Then, if a surface is modified, e.g. by changing its physicochemical properties, bacterial adhesion and, consequently, growth and maturation of a biofilm can be prevented (Li 2016). Throughout the years, an increased body of knowledge about the physical and chemical properties of the materials as key factors influencing cellular adherence and physiology has become available. As a consequence, the modification of surface physicochemical properties has become a topic of interest for the development of several approaches aimed at preventing microbial attachment and biofilm formation (Bazaka et al. 2015).

Polyethylene oxide (PEO) and polyethylene glycol (PEG) polymers are commonly used for this purpose because of their ability to repel proteins, therefore reducing bacterial attachment (Nisol et al. 2010). Although their mechanism of action is not fully understood, it is generally believed that the anti-biofilm behaviour might be related to steric repulsion (Fig. 2a) and to the presence of hydrophilic functional groups on the surface of the polymers. Indeed, these functional groups allow the formation of a layer of water when they are in aqueous environments (Fig. 2b), thus preventing direct contact between the polymers and the bacterial proteins which act as receptor sites for bacterial adhesion and colonization of the surface (Dong et al. 2007; Stallard et al. 2016). Da Ponte et al. (2011, 2012) employed a DBD system for the atmospheric pressure plasma deposition of PEO-like coatings on glass substrates using tetraethylene glycol dimethyl ether (TEGDME) as the precursor. As the chemical characterization by X-ray photoelectron spectroscopy (XPS) revealed, by varying the plasma parameters it was possible to achieve a high retention of the TEGDME monomer structure at the surface of the coatings, which showed an ether content (PEO character) of 70% in the carbon component (i.e., in the C1s signal, wich corresponds to the binding energies in the range of 282 – 292 eV in the XPS spectra). According to the literature, coatings with such degree of PEO character allow no cell adhesion (Sardella et al. 2004). Although the potential biological interest and applications of the coatings obtained by these authors is evident, no protein repelling tests or antimicrobial assays were performed in any of these two studies, which only provided a characterization of the physicochemical properties of the coatings produced.

Venault et al. (2013) designed an atmospheric plasma-induced PEGylation process on expanded poly(tetrafluoroethylene) (ePTFE) membranes for the improvement of their resistance to protein adsorption and bacterial attachment. After incubating the ePTFE membranes in a solution of a 10 wt% poly(ethylene glycol) methyl ether methacrylate (PEGMA) monomer and isopropanol, an atmospheric plasma treatment induced the copolymerization of the ePTFE surface and the PEGMA monomer. As the water contact angle (WCA) measurements revealed, a change in the wettability of the membranes was achieved. Whereas the uncoated ePTFE membranes were hydrophobic (WCA of 105 ± 1°), the PEGylated membranes obtained from a plasma treatment of 120 seconds were very hydrophilic (WCA of 9 ± 1°). Chemical characterization confirmed the efficient PEGylation of the ePTFE membranes and the analysis of surface morphology revealed a decrease in the porosity of the PEGylated membranes. As the plasma treatment time of the PEGylated membranes increased, protein adsorption and bacterial attachment decreased. It was concluded that PEGylated membranes with plasma treatments of 60 seconds or longer reduced the adsorption of fibrinogen in an 80% and completely avoided the attachment of *Staphylococcus epidermidis* and *E. coli*.

Stallard et al. (2012) evaluated the protein adsorption and bacterial attachment onto the surface of siloxane coatings that were deposited on silicon wafers and titanium coupons by using an APPJ system. Coatings with wetting characters ranging from superhydrophilic to superhydrophobic were obtained by using precursors with different chemical properties, as well as different process gases, and tuning process parameters such as the precursor flow rate and the distance between the plasma source and the substrate. Generally, it was observed that the adsorption of fibrinogen and bovine serum albumin tended to be greater on hydrophobic surfaces than on hydrophilic surfaces. Nevertheless, hydrophobic, fluorinated coatings exhibited lower protein adsorption than hydrophilic coatings. This fact may be due to the presence of fluorocarbon groups that cause a decrease in surface energy, thus reducing the interaction between the proteins and the surface. Interestingly, in this study the lowest levels of protein adsorption were exhibited by the superhydrophobic coatings, which were obtained by generating a nanotexture through the formation of surface features of between 10 and 250 nm that caused a significant increase in the roughness of the surfaces. Superhydrophobic, fluorinated coatings deposited on titanium coupons showed resistance to *S. aureus* attachment. It was concluded that a combination of a nanotextured morphology and a low surface energy chemistry (low adhesion) created a barrier to wetting by entrapping air in the morphology (Fig. 2c). Thus, the area available for the diffusion of proteins from an aqueous environment to the surface was reduced, as well as the attachment of bacteria.

The deposition of coatings that modify physicochemical properties of the surface can also be used to obtain a bactericidal effect (Fig. 2d). Sarghini et al. (2011) coated stainless steel substrates with 3-(trimethoxysilyl)-propyldimethyoctadecylammonium chloride (ODAMO) and butylamine through atmospheric plasma deposition in a parallel plate DBD chamber. In this case, the precursor was carried and injected in the plasma as an aerosol by a flow of nitrogen or air. The influence of the precursor, carrier gas, power input and deposition time on the physicochemical properties and bactericidal activity of the coatings was analyzed. The coatings obtained were smooth, thin and, in most cases, very hydrophilic. A strong influence of the carrier gas on the wettability of ODAMO-based coatings was identified, as they were more hydrophobic when the precursor was carried by nitrogen. The antimicrobial activity of the samples was tested against *E. coli*. Bacteria were killed with ODAMO-based coatings obtained by using air as carrier gas at any input power. Although butylamine-based coatings supported lower bacterial proliferation than the untreated substrate, they were not as effective as the ODAMO-based coatings. According to the results of the chemical characterization, it was concluded that the antibacterial activity was due to a compromise between two factors: (i) a substantial content in ammonium groups and (ii) the preservation of long hydrocarbon chains from the molecules of the precursor, which is favored by using low power levels.

To our knowledge, the most recent report on the use of atmospheric plasma for the deposition of coatings that modify the physicochemical properties of a surface for antibacterial purposes is the one by Hernández-Orta et al. (2018). These authors used a DBD reactor operating at room temperature and atmospheric pressure to induce the polymerization and to carry out a subsequent quaternization of 4-vinyl pyridine (4VP) coatings on high-density polyethylene substrates. The polyethylene substrate showed no bactericidal effect against *E. coli*, whereas quaternized coatings subject to 6 and 12 cycles of polymerization achieved a complete bacterial inactivation. These bactericidal properties against Gram-negative bacteria were mostly attributed to the relatively high surface charge density of the coatings, which causes an electrostatic effect that destabilizes the surface of the bacteria and leads to their destruction.

6. CONCLUSIONS AND FUTURE PROSPECTS

Different approaches that make use of atmospheric pressure plasma technologies for the deposition of anti-biofilm coatings have been reviewed in this paper. Since coatings that are effective against both Gram-positive and Gram-negative bacteria have been deposited on different substrates in the literature, it can be concluded that atmospheric pressure plasma technologies are interesting tools for antimicrobial applications in a wide range of fields.

Three types of approaches can be identified depending on how the antimicrobial effect is achieved: (i) composite coatings containing and releasing biocidal agents, (ii) coatings with an immobilized antibacterial agent and (iii) coatings that modify the surface physicochemical properties. Such coatings can prevent biofilm formation by either avoiding the initial bacterial attachment or killing the bacteria once attached to the surface.

From the point of view of the food industry, the deposition of coatings for the physicochemical modification of surfaces seems especially promising because it avoids the main problems that may arise with other alternatives. Surface physicochemical modification makes it possible to combat biofilm in the early stages of its formation by avoiding the initial bacterial attachment, instead of killing the bacteria after attachment. Therefore, the use of biocidal agents is also avoided and non-toxic surfaces with anti-biofilm effects can be obtained, which is very desirable for preserving the quality of food. Furthermore, long-lasting anti-biofilm effects can be obtained through this alternative because it relies on the physicochemical properties of the surface itself, instead of depending on the release of a finite amount of a specific agent as nanocomposites do.

To our knowledge, few studies focused on food-related applications have been published reporting the use of atmospheric pressure plasma technologies for the deposition of anti-biofilm coatings. Furthermore, there is a lack of regulation regarding the implantation of plasma technologies in the food industry and the available information on how food products are affected by plasma-generated reactive species is still limited and variable. Therefore, a wide range of possibilities are available for future research to generate more knowledge and facilitate the regulation and industrial implantation of these approaches by testing them on food contact materials and validating their compatibility with different food products.

As it has been reported, the mechanism of action of some antibacterial coatings is not fully understood. In order to gain a better understanding in this regard, some analysis techniques that have been used in the study of sanitizers may be adequate. For instance, nuclear magnetic resonance can be used for the identification of metabolic variations in bacteria (Liu et al. 2017, 2018), electron spin resonance for the examination of free radicals (Zhang et al. 2018a, 2018b) and sodium dodecyl sulphate polyacrylamide gel electrophoresis for the analysis of proteins (Zhao et al. 2019).

Compliance with Ethical Standards

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Fig. 1 Schematics of plasma deposition processes in (a) an atmospheric pressure plasma jet system and (b) a dielectric barrier discharge system, (c) surface interactions of adsorption, diffusion and desorption, (d) agglomeration and coalescence of the deposited material, and antibacterial action of (e) composite coatings containing and releasing biocidal agents and (f) coatings with immobilized antibacterial agents

Fig. 2 Mechanisms of antibacterial action of different coatings that modify the surface physicochemical properties: (a) steric repulsion, (b) hydration layer on hydrophilic coatings, (c) air entrapment on superhydrophobic coatings, (d) bacterial killing on bactericidal coatings