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Author(s):
Hertwig, Christian; Meneses, Nicolas; Mathys, Alexander

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Review

Cold atmospheric pressure plasma and low energy electron beam as alternative nonthermal decontamination technologies for dry food surfaces: A review

Christian Hertwig[a], Nicolas Meneses[b], Alexander Mathys[c,*]

[a] Leibniz Institute for Agricultural Engineering and Bioeconomy, Max-Eyth-Allee 100, D-14469 Potsdam-Bornim, Germany
[b] Bühler AG, Corporate Technology, Gupfenstrasse 5, CH-9240, Uzwil, Switzerland
[c] ETH Zurich, Institute of Food, Nutrition and Health, Laboratory of Sustainable Food Processing, Schmelzbergstrasse 9, CH-8092 Zurich, Switzerland

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ABSTRACT

Background: Dry food products are often highly contaminated, and dry stress-resistant microorganisms, such as certain types of *Salmonella* and bacterial spores, can be still viable and multiply if the product is incorporated into high moisture food products or rehydrated. Traditional technologies for the decontamination of these products have certain limitations and drawbacks, such as alterations of product quality, environmental impacts, carcinogenic potential and/or lower consumer acceptance. Cold atmospheric pressure plasma (CAPP) and low energy electron beam (LEEB) are two promising innovative technologies for microbial inactivation on dry food surfaces, which have shown potential to solve these certain limitations.

Scope and approach: This review critically summarizes recent studies on the decontamination of dry food surfaces by CAPP and LEEB. Furthermore, proposed inactivation mechanisms, product-process interactions, current limitations and upscaling potential, as well as future trends and research needs for both emerging technologies, are discussed.

Key findings and conclusions: CAPP and LEEB are nonthermal technologies with a high potential for the gentle decontamination of dry food surfaces. Both technologies have similarities in their inactivation mechanisms. Due to the limited penetration depth of both technologies, product-process interactions can be minimized by maintaining product quality. A first demonstrator with Technology Readiness Level (TRL) 7 for LEEB has already been introduced into the food industry for the decontamination of herbs and spices. Compared with LEEB, CAPP is at the advanced development stage with TRL 5, for which further work is essential to design systems that are scalable to industrial requirements.

1. Introduction

Low moisture or dry food ingredients and products have been a significant component of the human diet for thousands of years. These food ingredients have a low moisture content, such as spices, nuts and cereals, or they undergo a drying process, e.g., nuts, herbs and fruits. Dry products can be contaminated with food-spoiling bacteria, pathogens and bacterial spores, especially herbs and spices, which can often be highly contaminated with viable counts $> 10^7$ CFU g^{-1} (Schweiggert, Carle, & Schieber, 2007). The contamination source is often related to the pre-harvest environment and can be transferred during post-harvest processing, which can also be conducted in an open-air environment. Contamination may also occur as a consequence of recontamination issues during processing and poor manufacturing practices. Dry products are often inappropriately assumed to be microbiologically safe since the low moisture content represents a significant barrier to the growth of microorganisms (Gurtler, Doyle, & Kornacki, 2014). However, dry stress-resistant microorganisms, such as bacterial spores or some types of *Salmonella*, are still viable and can multiply if the product is rehydrated or incorporated into high moisture products with a sufficient amount of available nutrients. For instance, the contamination of nuts and nut products with pathogens is a reoccurring concern in the food industry. Between 2001 and 2016, approximately 82 recalls of nuts and nut products were reported in the United States, with quantities of up to 148.000 tons of products, due to pathogen contamination (Palumbo, Beuchat, Danyluk, & Harris, 2015). *Salmonella* strains are responsible for most of the reported outbreaks associated with the consumption of nuts and nut products (Gurtler

* Corresponding author.
E-mail address: alexander.mathys@hest.ethz.ch (A. Mathys).

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et al., 2014). Furthermore, in recent decades, cases of food-borne diseases and intoxicants have increased due to contaminated herbs and spices, which were incorporated in different food products (Buckenhüskes & Rendlen, 2004). Moreover, the use of spoiled herbs and spices can also drastically reduce the shelf-life of food products. The inactivation of microorganisms on dried products is challenging, mostly due to the increased resistance of microorganisms in low-water activity foods in comparison with water-rich foods (Fine & Gervais, 2005). Common technologies for the decontamination of dry products are fumigation with ethylene or propylene oxide, saturated steam or dry heat, microwave treatment, high energy electron beams and Gamma-rays (Pan, Bingol, Brandl, & McHugh, 2012; Schweiggert et al., 2007). However, these technologies have certain limitations and drawbacks, such as alterations of the aroma, odor and color (Schweiggert et al., 2007). Irradiation with Gamma-rays can only be applied in authorized facilities and in controlled doses. Moreover, this process has a poor consumer acceptance in the EU. The main drawbacks of Gamma-irradiation are difficulties in transporting sources and licensing new facilities. Fumigation with ethylene oxide and propylene oxide is widely used in the USA; however, it is prohibited in many countries due to its carcinogenic potential to humans (Farkas & Mohácsi-Farkas, 2014; Schweiggert et al., 2007).

Such drawbacks and limitations, as well as consumers' demand for high quality and safe foods, require the development and application of emerging nonthermal technologies for the gentle decontamination of dry food products. Cold atmospheric pressure plasma (CAPP) and low energy electron beam (LEEB) are two promising innovative technologies for microbial inactivation on dry food surfaces. This review will discuss and compare the application of both nonthermal technologies for microbial inactivation; dealing with bacteria, molds, and bacterial spores; proposed inactivation mechanisms, product-process interactions, current limitations and upscaling potential; and future trends and research needs.

2. Cold atmospheric pressure plasma

2.1. Cold atmospheric pressure plasma technology

Plasma can be characterized as an at least partially ionized gas and is a complex mixture of different components, such as charged particles (electrons & ions) and neutral species (atoms & molecules), in addition to radicals, UV photons and irradiated heat. In general, plasma can be classified according its temperature into thermal and nonthermal plasmas. A thermal plasma is an almost completely ionized gas, whereby the temperatures of the charges and neutral species are approximately equal, with temperatures typically reaching at least 15,000 K (Eliasson & Kogelschatz, 1991). In comparison to thermal plasma, nonthermal ones are only partially ionized, indicating that the number of neutral species is much higher than the number of charged species, whereby the temperature of the different particles is not equal. The temperature of electrons is still in the range of several thousand Kelvin, but the temperature of the neutral species and ions can be close to ambient temperature. Thus, nonthermal plasmas are also termed cold plasmas.

For the generation of cold plasma energy need to be supplied to a gas, electric energy sources have been shown to be the most convenient. The lifetime of the particles inside the plasma is quite small due to energy loss by collision processes, and therefore energy must be supplied continuously for plasma applications. The generation of cold plasma can be achieved under atmospheric pressure and/or lower pressure conditions. Hence, plasma generation at atmospheric pressure allows continuous processing, which is a clear advantage for food process applications. A manifold number of different plasma systems are available because of various possible electrode configurations (geometry, number, location). Fig. 1 schematically illustrates two of the most commonly used plasma systems: plasma jet and dielectric barrier discharge (DBD) systems. However, CAPP technology is not yet standardized. Research groups are often using plasma systems, which are available in their country and/or custom built or in-house manufactured systems. Furthermore, these plasma systems are designed mostly for laboratory applications.

2.2. Microbial inactivation mechanisms of cold atmospheric pressure plasma

There is continued and growing interest in CAPPs for microbial inactivation in the food sector, whereby the possible applications are manifold, such as the treatment of fresh and dry products, or the in package treatment of food (Bourke, Zuizina, Han, Cullen, & Gilmore, 2017; Niemira, 2012a; Schlüter et al., 2013). As stated in chapter 2.1, CAPP is a complex mixture of different generated components, such as UV photons, charged particles, radicals and other reactive nitrogen, oxygen and hydrogen species (RNS, ROS & RHS), such as nitrogen oxides ($NO^{\bullet}$ and NO_x), peroxynitrite ($ONOO^-$), atomic oxygen (O), ozone (O_3), singlet oxygen ($^1O_2$), superoxide anion (O_2^-), hydrogen radicals ($H^{\bullet}$), hydroxyl radicals ($OH^{\bullet}$) and/or hydrogen peroxide (H_2O_2). These reactive species, which can act individually and/or synergistically, are responsible for the antimicrobial effect of CAPP. The plasma system and the applied operating parameters (e.g., process gas, moisture, and energy input) affect the composition of the generated plasma and consequently also the antimicrobial efficiency of the plasma treatment (Bourke et al., 2017; Niemira, 2012a). Whereby, the plasma chemistry is complex and can involves hundreds of different species and thousands of possible reactions (Lu & Wu, 2013; Sakiyama, Graves, Chang, Shimizu, & Morfill, 2012). Another aspect that influences CAPP-based microbial inactivation is the manner in which the plasma will be applied to a surface, such as direct, semi-direct or indirect application (Table 1). Direct plasma application to a surface results in an interaction of possible generated UV photons, charged particles, and radicals, among others, with microorganisms on the surface. Compared with direct CAPP treatment, the antimicrobial effect is based on long-lived reactive species if CAPP is applied semi-directly or indirectly.

The mechanisms responsible for microbial inactivation by CAPP have not been elucidated in detail. Fig. 2 illustrates the mechanisms that could be involved in CAPP-mediated microbial inactivation. However, the inactivation mechanisms can vary between microorganisms, e.g., bacterial spores are more resistant than bacteria (Hertwig, Reineke, Ehlbeck, Knorr, & Schlüter, 2015a). During the treatment, microorganisms are exposed to continuous bombardment with different reactive components of the generated plasma. Thereby, atomic and molecular radicals and excited molecules can cause an erosion of the microbial cell, atom by atom, through etching (Moisan et al., 2001). The reactive components will be adsorbed onto the microorganism surface, causing the formation of volatile compounds via chemical reactions and openings and lesions in the cell membrane. Consequently, Gram-negative bacteria are believed to be more easily inactivated by etching due to their thinner membrane structure compared with Gram-positive bacteria (Stoffels, Sakiyama, & Graves, 2008). Fröhling, Baier, Ehlbeck, Knorr, and Schlüter (2012) showed that Gram-negative *Escherichia coli* are more sensitive to CAPP than Gram-positive *Listeria innocua*. Bacterial spores can also be inactivated by etching caused by oxygen atoms and radicals (Park et al., 2004). Microbial support structures, such as biofilms, can also be degraded by the erosion of organic material due to the breakage of chemical bonds (Bourke et al., 2017). Different reactive species inside the CAPP, such as RNS and ROS, can also interact with various cellular macromolecules, such as membrane lipids, proteins and DNA. Oxidative DNA damage caused by ROS can lead to microbial inactivation (Li, Sakai, Watanabe, Hotta, & Wachi, 2013). Oxidative damage of membrane lipids affects their ability to regulate mass transport in and out of the cell (Laroussi & Leipold, 2004). Furthermore, the generated reactive species may also diffuse into the microbial cell, which can result in a decrease in intracellular

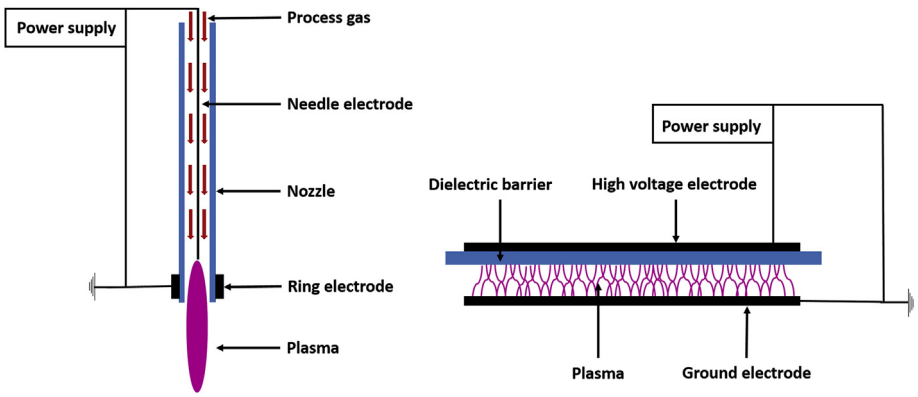

Fig. 1. Schematic of a plasma jet system (left) and dielectric barrier discharge system (right).

pH. If a bacterial cell cannot maintain pH homoeostasis, it will be inactivated (Booth, 1985; Padan & Schuldiner, 1987).

The cell membrane can also be damaged via electrostatic disruption due to the accumulation of charged particles on the microorganism surface (Laroussi, 2002). Such electrostatic forces caused by charge accumulation on an outer membrane surface can overcome the tensile strength of the membrane, resulting in its rupture, as shown by Mendis, Rosenberg, and Azam (2000).

UV photons emitted by CAPP are able to induce the dimerization of thymine bases in bacterial DNA strands, which impedes the ability of bacteria to replicate (Laroussi, 2002). Furthermore, UV photons can also damage the DNA of bacterial spores (Setlow, 2007). Faster spore inactivation with increasing UV emission of the used CAPP has been reported by Boudam et al. (2006) and Reineke, Langer, Hertwig, Ehlbeck, and Schlüter (2015). Hertwig et al. (2015b) showed an increase in DNA damage in spores with increasing UV emission. UV photons can also induce intrinsic photodesorption, an erosion of the cell, atom by atom, due to the breakage of chemical bonds (Moisan et al., 2001).

Bacterial spores can also be inactivated by CAPP due to damage to the inner spore membrane and key germination proteins (Wang, Doona, Setlow, & Li, 2016). Hertwig, Reineke, Rauh, and Schlüter (2017a) showed that different spore properties, the outer spore coat, dipicolinic acid (DPA) level inside the spore core and the DNA saturation with small acid soluble proteins (SASPs), contribute to the resistance of *Bacillus subtilis* spores to different generated plasma components. Nevertheless, identifying the main mechanism responsible for microbial inactivation is challenging, since the different interactions can occur simultaneously and also post mortem.

The humidity of the process gas also has an effect on CAPP-based inactivation, whereby the specific impact is still part of ongoing research. Muranyi, Wunderlich, and Heise (2008) treated *B. subtilis* and *Aspergillus niger* spores with air-plasma up to 8 s and increased the relative process gas humidity up to 80%. For *A. niger* spores an improved inactivation effect was observed, with maximum inactivation at 70% relative humidity. This effect was enhanced after prolonged CAPP treatment. However, *B. subtilis* spores showed slightly lower inactivation with increased relative humidity, especially for shorter treatment times. In contrast, Jeon, Klaempfl, Zimmermann, Morfill, and Shimizu (2014) and Patil et al. (2014) reported an increased inactivation for *Geobacillus stearothermophilus* and *Bacillus atrophaeus* spores with increasing process gas humidity. Both research groups suggested that the effect was probably due to a higher generation of ROS, such as hydrogen peroxide and hydroxyl radicals. In a humid environment reactive CAPP components, such as ROS and RNS, are able to initiate various chain reactions. This can results in the generation of a larger diversity of reactive species (Surowsky, Schlüter, & Knorr, 2014).

2.3. Cold atmospheric pressure plasma treatment of dry food surfaces

The application of CAPP is a promising technology for the decontamination of dry food products. A comprehensive overview of studies investigating microbial inactivation on dry food products by CAPP is presented in Table 2, which briefly summarizes the plasma sources and process gases used to inactivate various microorganisms and the obtained results. The summarized literature overview shows the high diversity in plasma processing with respect to the plasma source, energy input, process gas and kind of application. The large number of CAPP process variables involved, as well as food matrix-related parameters and methodological differences, complicate the comparability of these studies. The results showed that CAPP is capable of decontaminating plant and animal-based products; however, the largest proportion of the work was performed using plant products. Choi, Puligundla, and Mok (2016) investigated the application of CAPP to dried, shredded Alaska pollock, a typical snack food in China, Korea and other Asian nations. For the treatment, they used a corona discharge plasma jet driven by air and reported an inactivation of the native microbial flora of 2.5 $\log_{10}$ after a 20-min treatment. Bußler et al. (2016) focused on an alternative and sustainable food source: edible insects. They treated mealworm larvae flour with air-plasma generated by a surface DBD up to 15 min

Table 1
Overview of different types of cold plasma applications. (Adapted from Schlüter et al., 2013).

Type	Description	Examples
Direct	Plasma is in direct contact with the substrate	Plasma jet
	Interaction based on irradiation (ultraviolet (UV), vacuum UV (VUV)), charged molecules, radicals, and reactive particles	Dielectric barrier discharges (DBD)
Semi-direct	Distance between plasma and substrate much larger than the mean free particle path	Surface DBD with gap
	No interaction with charged particles	Sterrad process with
	Antimicrobial effect based on irradiation, long-lived radicals, metastable and inhibitory substances	plasma-activated hydrogen peroxide
Indirect	Irradiation with VUV, UV	UV lamps
	No reaction with plasma particles	Ozone generator
	Plasma is used to treat gas or liquids	Plasma-processed air (PPA) Plasma-processed water (PPW)

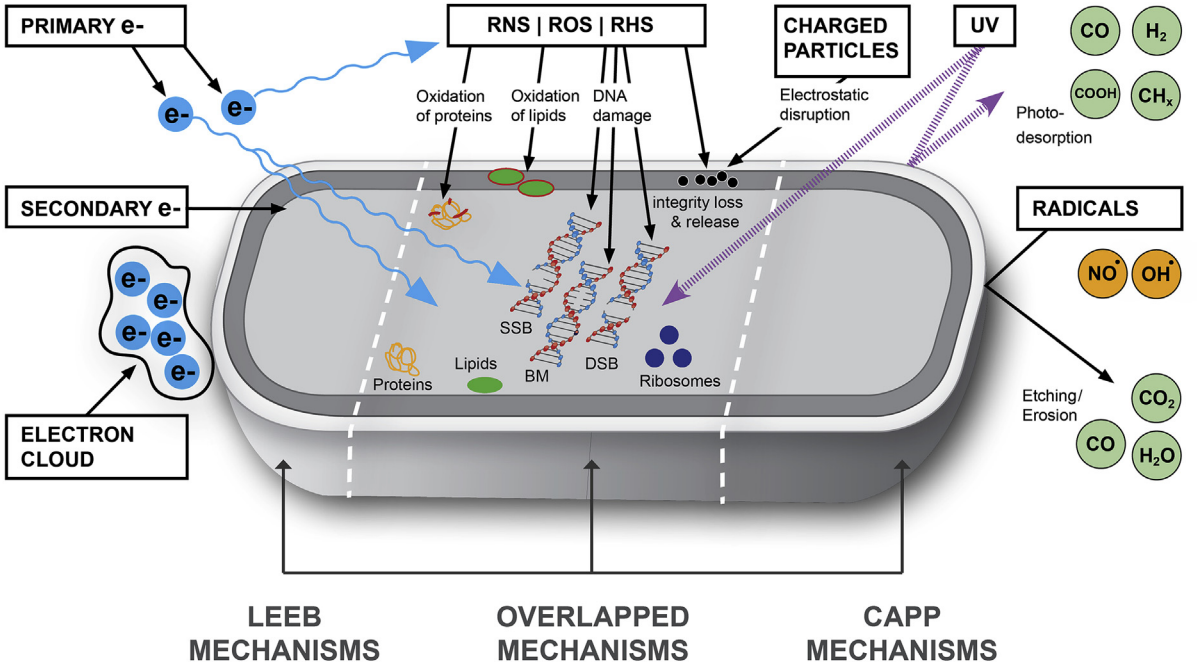

Fig. 2. Schematic overview of cold atmospheric pressure plasma (CAPP) and low energy electron beam (LEEB) effects versus inactivation mechanisms based on available literature in Chapter 2.2 and 3.2. (BM: Base modification; DSB: Double strand break; SSB: Single strand break).

and inactivated the native microbial load by 3.0 $\log_{10}$. Sun, Anderson, and Keller (2014) inoculated black peppercorns with a cocktail of different *Salmonella* strains and reported an inactivation of 5.5 $\log_{10}$ following 80 s of air-plasma treatment. The antimicrobial efficiency of a direct argon plasma jet and indirect air-plasma treatment was compared by Hertwig et al. (2015a). Direct plasma treatment resulted in much lower inactivation, probably due to the involvement of different inactivation mechanisms. The indirect plasma treatment inactivated *Salmonella enterica*, *B. subtilis* and *atrophaeus* spores by 4.1, 2.4 and 2.8 $\log_{10}$, respectively, after a 30-min treatment. Hertwig et al. (2015c) studied the inactivation of the native microbial flora of black peppercorns, crushed oregano and paprika powder following an indirect treatment with air-plasma. The authors reported an inactivation of > 4.0 and > 3.0 $\log_{10}$ for black peppercorns and paprika powder, respectively, after 60 min. A lower inactivation of 1.6 $\log_{10}$ was determined for crushed oregano after 90 min, probably due to the much lower initial native microbial load. Deng et al. (2007) treated almonds inoculated with *E. coli* and showed an inactivation of 5.0 $\log_{10}$ after 30 s of DBD plasma treatment. Niemira (2012b) inoculated almonds with different types of *Salmonella* and *E. coli* O157:H7 and used a plasma jet with the process gases air and N_2. The treatment with air-plasma was more efficient, the highest inactivation was achieved for *E. coli* O157:H7 after 20 s with 1.2 $\log_{10}$. Air-plasma was also more efficient in inactivating *Salmonella* Enteritidis PT30 on almonds compared with plasma generated with N_2, O_2, CO_2 and CO_2 in an admixture with argon (Hertwig et al., 2017b). After a 15-min treatment between two diffuse surface barrier discharge plasma plates, > 5.0 $\log_{10}$ *S.* Enteritidis PT30 was inactivated.

In addition to bacteria, CAPP can also be applied to inactivate molds on nuts. Dasan, Boyaci, and Mutlu (2017) used an atmospheric pressure fluidized bed plasma generated with air and N_2 for the treatment of dehulled hazelnuts. The authors reported an inactivation for both process gases for *A. flavus* and *parasiticus* spores of > 4.0 $\log_{10}$ after 5 min. Similar results were obtained by Dasan, Boyaci, and Mutlu (2016) by treating maize using the same plasma setup and mold spores. The application of CAPP is also able to reduce the native microbial flora of sprout seeds. Contaminated seeds are one of the major concerns related to sprout-associated outbreaks. The native microbial flora of chickpea seeds could be inactivated by 2.0 $\log_{10}$ following 5 min of air-plasma treatment (Mitra et al., 2014). Butscher, Van Loon, Waskow, von Rohr, and Schuppler (2016a) treated *E. coli*-inoculated alfalfa, onion, radish and cress seeds using argon-plasma and reported an inactivation between 1.4 and 3.4 $\log_{10}$, depending on the seeds, after 10 min. Air-plasma generated with a corona discharge plasma jet inactivated 2.3 $\log_{10}$ of the broccoli seed native microbial flora after 3 min (Kim, Puligundla, & Mok, 2017). The same research group also investigated the antimicrobial efficiency of different CAPP treatments on a dried laver. A DBD air-plasma treatment of 10 min could inactivate the native microbial flora up to 2.5 $\log_{10}$.

In addition to microbial inactivation, CAPP can also be applied to degrade mycotoxins on dry food products. Siciliano et al. (2016) inoculated aflatoxins onto dehulled hazelnuts and treated them with DBD N_2-plasma. After a 12-min CAPP treatment, the aflatoxin concentration was reduced by up to 70%. Similarly, a > 80% reduction of aflatoxins on corn following a 10-min air-plasma treatment with RH of 40% was reported by Shi, Ileleji, Stroshine, Keener, and Jensen (2017). Furthermore, direct and indirect CAPP treatment were equally effective for the degradation of aflatoxin. Ten Bosch et al. (2017) studied the degradation of different mycotoxins produced by *Fusarium*, *Aspergillus* and *Alternaria* species using DBD air-plasma and showed a varied degradation rate depending on the mycotoxin structure.

CAPP affects not only microorganisms, but it also interacts with the treated surface, which could lead to plasma-product interaction. However, due to the limited penetration depth of the plasma components, such plasma-product interactions are commonly restricted to the product surface. Bußler et al. (2016) reported an increased oil-binding capacity after air-plasma treatment of mealworm flour, whereas the water binding capacity decreased. In contrast, Thirumdas, Deshmukh, and Annapure (2016) observed an increase in water holding and binding capacity in basmati rice flour after an air-plasma treatment. CAPP could also lead to undesirable plasma-product reactions. Hertwig et al. (2017b) investigated the impact of different process gases on the almond surface color. The use of process gases containing nitrogen (air and N_2) resulted in a significant browning of the almonds, whereas the other applied process gases (O_2, CO_2, 90% CO_2 + 10% Ar) did not considerably alter the color. Indirect air-plasma treatment of black

Table 2
Overview of studies investigating the decontamination of dry food products by cold atmospheric pressure plasma (CAPP). Some values/numbers have been approximated from graphs provided in the cited literature. (CFU: Colony forming unit; DBD: Dielectric barrier discharge; DCSBD: Diffuse coplanar surface barrier discharge; RH: Relative humidity).

Reference	Product	Plasma source	Process gas	Organism	Comments
Deng et al. (2007)	Almond	DBD (16–30 kV, 1.2–2.4 kHz)	Air	*Escherichia coli* (1.1×10^6–4.8×10^5 CFU g^{-1})	• 5.0 $\log_{10}$ reduction, 30 s at 30 kV and 2 kHz
Niemira (2012a,b)	Almond	Plasma jet (549 W, 47 kHz)	Dry air, N_2	*Salmonella* Anatum, *Salmonella* Stanley, *Salmonella* Enteritidis PT30, *Escherichia coli* O157:H7	• Air generally more effective as N_2 • Greatest reduction ~1.4 $\log_{10}$ for *E. coli* at 6 cm distance and 20 s
Mitra et al. (2014)	Chickpea seed	FlatPlaSter 2.0 (surface micro-discharge plasma)	Air	Native microbial flora	• 2.0 $\log_{10}$ reduction after 5 min
Sun et al. (2014)	Black peppercorn	Dyne-A-Mite variable chemistry plasma (arc discharge plasma)	Dry air	Mixed cocktail of *Salmonella* Enteritidis PT30, *Salmonella* Oranienburg, *Salmonella* Tennessee, *Salmonella* Anatum	• 4.5–5.5 $\log_{10}$ reduction after 60–80 s
Hertwig et al. (2015a)	Black peppercorn	Direct: RF plasma jet (27.12 MHz, 30 W) Indirect: microwave torch (2.45 GHz, 1.2 kW)	Direct: argon Indirect: dry air	*Bacillus subtilis* and *atrophaeus* spores, *Salmonella enterica* (~10^7 CFU g^{-1}), native microbial flora	• Direct: 0.8, 1.3, 2.7 and 0.7 $\log_{10}$ reduction for *B. subtilis* and *atrophaeus*, *S. enterica* and native microbial flora after 15 min • Indirect: 2.4, 2.8, 4.1 and 2.0 $\log_{10}$ reduction for *B. subtilis* and *atrophaeus*, *S. enterica* and native microbial flora after 30 min
Hertwig et al. (2015c)	Black peppercorn (BP), crushed oregano (OR), paprika powder (PP)	Microwave torch (2.45 GHz, 1.2 kW) indirect application of exhaust gas	Dry air	Native microbial flora	• BP 4.0 $\log_{10}$ reduction after 60 min • PP 3.0 $\log_{10}$ reduction after 60 min • OR 1.6 $\log_{10}$ reduction after 90 min
Kim et al. (2015a)	Dried laver	DBD (30 kV, 30 kHz)	Air	Native microbial flora	• 2.5, 1.5 and 1.0 $\log_{10}$ reduction for aerobic bacteria, marine bacteria and molds after 10 min
Kim et al. (2015b)	Dried laver	Corona discharge plasma jet (20 kV, 58 kHz)	Air	Native microbial flora	• 2.0, 1.5 and 1.2 $\log_{10}$ reduction for aerobic bacteria, marine bacteria and molds after 20 min
Bußler et al. (2016)	Mealworm larvae flour	Surface DBD (8.8 kV, 3 kHz)	Air	Native microbial flora	• 3.0 $\log_{10}$ reduction after 15 min
Butscher et al. (2016a)	Alfalfa, onion, radish, cress seeds	DBD (10 kHz, 8 kV)	Argon	*Escherichia coli* (10^7 CFU g^{-1})	• Onion: 1.4 $\log_{10}$ reduction after 10 min • Radish: 2.0 $\log_{10}$ reduction after 10 min • Alfalfa: 3.0 $\log_{10}$ reduction after 10 min • Cress: 3.4 $\log_{10}$ reduction after 10 min
Butscher et al. (2016b)	Wheat grain	DBD (8 kV, 10 kHz)	Argon	*Geobacillus stearothermophilus* (10^7 CFU g^{-1})	• 3.0 $\log_{10}$ reduction after 60 min
Choi et al. (2016)	Dried Alaska pollock shreds	Corona discharge plasma jet (20 kV, 58 kHz)	Air	Native microbial flora	• 2.5, 1.5, > 1.2 and ~1.0 $\log_{10}$ reduction for aerobic bacteria, marine bacteria, *Staphylococcus aureus* and molds after 20 min
Lee et al. (2016)	Brown rice	DBD (250 W, 15 kHz)	Air	*Bacillus cereus*, *Bacillus subtilis*, *Escherichia coli* O157:H7 (4.1 $\log_{10}$ CFU g^{-1})	• 2.3 $\log_{10}$ reduction after 20 min
Dasan et al. (2016)	Maize	Atmospheric pressure fluidized bed plasma (655 W, 25 kHz)	Air, N_2	*Aspergillus flavus*, *Aspergillus parasiticus* spores (1–1.3 $\times$ 10^7 CFU g^{-1})	• Air: 5.5 and 5.2 $\log_{10}$ reduction for *A. flavus* and *parasiticus* after 5 min • N_2: 4.6 and 4.7 $\log_{10}$ reduction for *A. flavus* and *parasiticus* after 5 min
Dasan et al. (2017)	Hazelnut	Atmospheric pressure fluidized bed plasma (655 W, 20 & 25 kHz)	Air, N_2	*Aspergillus flavus*, *Aspergillus parasiticus* spores (4.7–5.8 $\times$ 10^6 CFU g^{-1})	• Air: 4.5 and 4.2 $\log_{10}$ reduction for *A. flavus* and *parasiticus* after 5 min • N_2: 4.2 and 4.1 $\log_{10}$ reduction for *A. flavus* and *parasiticus* after 5 min
Hertwig et al. (2017b)	Almond	DCSBD (350 W, 15 kHz)	Air, N_2, O_2, CO_2, CO_2/Ar (90/10%)	*Salmonella* Enteritidis PT30 (5.5 $\times$ 10^6 CFU g^1)	• Air: > 5.0 $\log_{10}$ reduction after 15 min • N_2: 2.0 $\log_{10}$ reduction after 15 min • O_2: 4.8 $\log_{10}$ reduction after 15 min • CO_2: 2.3 $\log_{10}$ reduction after 15 min • CO_2/Ar: 3.0 $\log_{10}$ reduction after 15 min
Kim et al. (2017)	Broccoli seed	Corona discharge plasma jet (20 kV, 58 kHz)	Air	Native microbial flora	• 2.3, 2.0,1.8, 1.5 and 1.2 $\log_{10}$ reduction for aerobic bacteria, *E. coli*, *Salmonella*, molds & yeast and *B. cereus* after 3 min

peppercorns, oregano and paprika powder resulted in a significant loss of paprika powder redness, but the color of the other two products was only slightly affected (Hertwig et al., 2015c). The impact of direct and indirect CAPP treatment on quality parameters of black peppercorns, volatile oil content and the main aroma compound piperine, has been shown by Hertwig et al. (2015a). Neither plasma treatment substantially affected the surface color or the quality parameters. The impact of different CAPP applications on dried seafood products after microbial inactivation has also been investigated (Choi et al., 2016; Kim et al., 2015a, 2015b). The researchers reported no significant changes in physicochemical properties, such as color, total phenolic content or radical scavenging activity, after the CAPP treatment.

CAPP can interact with the surface of seeds to enhance germination and seedling growth without affecting physico-chemical and sensory characteristics of the grown sprouts (Butscher et al., 2016a; Dobrin, Magureanu, Mandache, & Ionita, 2015; Kim et al., 2017; Mitra et al., 2014; Zahoranová et al., 2016). CAPP treatment can change the surface characteristics of the seeds, which increases their wettability, as shown by a decreased contact angle and improved water uptake of the seeds (Randeniya & De Groot, 2015).

2.4. Limiting factors

The structure of a surface is one main parameter limiting the antimicrobial effect of a CAPP treatment due to a non-uniformity of the treatment. Hertwig et al. (2015a) and Butscher, Zimmermann, Schuppler, and Rudolf von Rohr (2016b) reported that a well-structured surface, i.e., a corrugated surface, can negatively affect microbial inactivation. Hertwig et al. (2015b) showed how the microbial inactivation of a CAPP is significantly reduced by the treatment of surfaces with a complex structure, such as on black peppercorns. The researchers investigated the inactivation of *B. subtilis* spores inoculated with a comparable spore density of 4×10^6 spores cm^{-2} on a simple flat glass surface, a spherical model (glass beads) and a real food matrix (black peppercorns). Black peppercorns have a well-structured surface characterized by cracks, grooves and pits, which might cause shadow effects for the different generated components of the plasma. Components such as UV photons and radicals cannot interact with the microorganisms, thus reducing the inactivation efficiency of the CAPP treatment. Another parameter affecting the plasma-based inactivation is the microbial load on the treated surface. Yu et al. (2006) investigated the inactivation of *E. coli* with surface densities between 10^7 to 10^{11} CFU cm^{-2} and reported a decreasing inactivation with an increasing cell surface density. Deng, Shi, Shama, and Kong (2005) and Fernández, Shearer, Wilson, and Thompson (2012) reported a slower inactivation with an increased initial microbial load for the treatment of *B. subtilis* spores and *Salmonella* Typhimurium. The decreased inactivation at a higher cell surface density probably results from stacking and aggregating of the microorganisms. The top layer of multilayered cell structures, even if the microorganisms are inactivated, could form a physical barrier to shield the microorganisms beneath the top layer from generated reactive plasma components and inactivation (Deng et al., 2005). This process can be attributed to the limited penetration depth of plasma components, which depends on their half-lives and the treated material, e.g., the penetration depth of UV photons can exceed 1 μm depending on the wavelength and material (Lerouge, Wertheimer, Marchand, Tabrizian, & Yahia, 2000). In the case of cell agglomeration, the inactivated top cell layer must be decomposed due to processes such as etching and photodesorption, so that the reactive plasma components can inactivate cell layers beneath the top one.

The surface-to-volume ratio of a product could also influence the inactivation efficiency. Hertwig et al. (2015c) reported that for a CAPP treatment of products with a high surface-to-volume ratio, such as powdered products, the plasma is more likely to interact with the food surface itself rather than with the microorganisms on that surface.

3. Low energy electron beam

3.1. Low energy electron beam technology

Electron beam (EB) technology belongs to the group of ionizing radiation, such as X-rays and γ-rays. All three technologies depend on the generation of high energy ionizing radiation to inactivate microorganisms. However, compared with X- and γ-rays, for which inactivation is due to energetic photons, EB utilizes high energy electrons for microbial inactivation. Unlike radiation with γ-rays, EB does not use radioisotopes. The electrons are generated via electricity and can be switched on or off as needed. Depending on the kinetic energy of the electrons, EB can be distinguished between high energy (> 300 keV) and low energy electron beams (< 300 keV). The penetration depth of the electrons is controlled and determined by their energy and mostly by the density of the treated material. After the electron penetrates the product, it will lose its kinetic energy due to collisions with the product particles. The higher the kinetic energy, the deeper the electron penetrates into the product (Urgiles et al., 2007). High energy electrons can effectively penetrate food products up to several centimeters, whereas the penetration depth of LEEB is limited to the micrometer scale (Urgiles et al., 2007). High energy EB already proved to be an efficient technology for the decontamination of dry products and shelf life extension of fresh produce, such as melons and berries (Pillai & Shayanfar, 2018). Whereby, a uniform treatment can be ensured by an optimized process configuration and by an optimized packaging of the product to be treated (Pillai & Shayanfar, 2015). Furthermore, the high penetration depth of the high energy electrons mitigate a non-uniformly dose distribution inside the product. The dose uniformity within a batch is controlled with dosimeters placed in several different locations of the batch (Pillai & Shayanfar, 2015).

LEEB technology deposits electron energy close to the surface where microorganisms are present, resulting in an extremely high efficiency for surface decontamination (Urgiles et al., 2007). LEEB was introduced in 2002 in the agricultural sector as a replacement for the chemical dressing of seeds. Currently, LEEB systems for seed treatment with a capacity of up to 30 tons per hour are available (Röder et al., 2009). In 2012, Tetra Pak introduced LEEB for the sterilization of food packaging material as replacement for hydrogen peroxide (Comet Group, 2012a). Recently, the first demonstrator of LEEB was introduced in the spice and herb industries (IIA, 2017). A sealed EB lamp is used as a LEEB source (Fig. 3). The low energy electrons are generated by a cathode made of tungsten filaments, sitting at negative high voltage electrodes inside an ultrahigh vacuum. If a high voltage is applied, the electrons start to “boil off” the tungsten wires, and electrostatic optics focus and guide them to the window (Comet Group, 2012b). Such LEEB systems offer different advantages, such as a low or negligible thermal impact on the treated product, compact size and simple integration into existing production processes (Chalise, Hotta, Matak, & Jaczynski, 2007; Comet Group, 2012a). Compared with other technologies using ionizing radiation, the shielding infrastructure of LEEB machinery can be maintained at a minimum level due to the lower penetration depth of the electrons and reduced X-ray scattering.

3.2. Microbial inactivation mechanisms with low energy electron beams

The mechanisms of microbial inactivation with LEEB are not completely understood, but it is believed that they are similar to those associated with other types of ionizing radiation (X- and γ-rays) (Chalise et al., 2007). The main target of ionizing radiation is DNA (Moeller et al., 2008; Nicholson, Munakata, Horneck, Melosh, & Setlow, 2000). LEEB can inactivate microorganisms due to direct or indirect interactions, whereas the final inactivation is an outcome of both processes. The mechanisms involved in microbial inactivation by LEEB are illustrated in Fig. 2. The direct interactions result from energy transfer of the electrons to target molecules, e.g., DNA, RNA, enzymes and

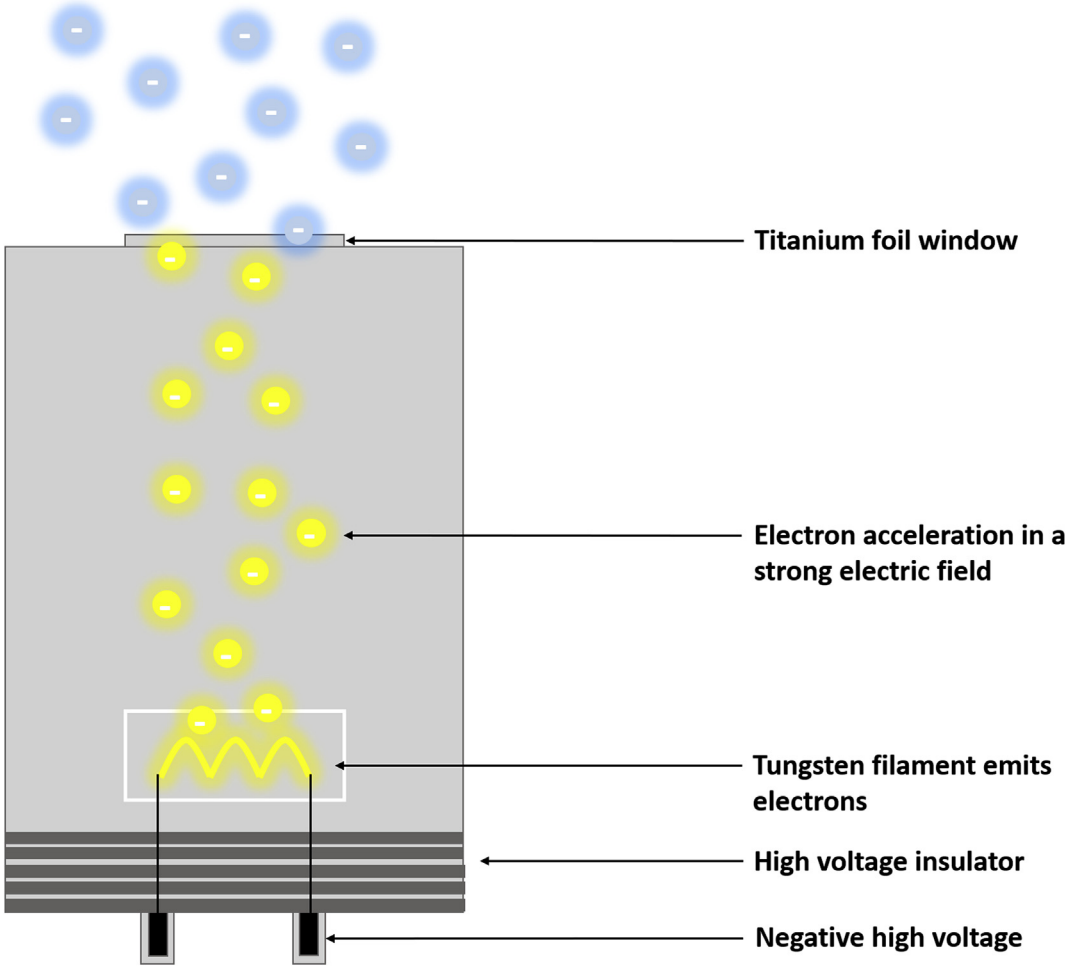

Fig. 3. Schematic of a low energy electron beam lamp. (Adapted from Comet Group, 2012b).

membrane proteins. Thus, the cellular DNA is damaged due to disintegration, such as single or double-strand breaks, as well as base modifications. The induced DNA damage is the main mechanism responsible for microbial inactivation by direct effects (Tahergorabi, Matak, & Jaczynski, 2012). Indirect effects of LEEB are caused by the ionization of water and/or oxygen molecules, leading to the generation of different reactive species. Since bacterial cells consist of up to 70% water, the generated electrons will be absorbed, generating highly reactive radicals, such as hydroxyl and superoxide radicals, affecting inter alia DNA and cell membranes (Chalise et al., 2007; Tahergorabi et al., 2012). Ghomi et al. (2005) treated *E. coli* with a pulsed LEEB of 5 μs generated by a secondary emission electron gun and suggested that cell death occurred by irreversible electroporation of the bacterial cells. However, spectroscopic and scanning electron microscopy analyses indicated no damage to the outer cell structure. The antimicrobial efficiency can vary between microorganisms, e.g., bacterial spores are more resistant than bacteria. Gram-negative *E. coli* is less resistant to LEEB than the Gram-positive *B. subtilis* (Chalise et al., 2004; Rahman et al., 2006).

Low energy electrons generate secondary electrons with lower energy levels on their way to the treated surface due to ionization processes by inelastic collisions. This process can result in the creation of a so-called “electron cloud”, which can also penetrate and cover the pores and cracks of structured surfaces (Bugaev et al., 1994). Nikjoo and Lindborg (2010) proposed that low energy electrons can generate more secondary electrons than high energy ones. Furthermore, it is well known that secondary electrons with lower energy levels damage DNA (Brun, Cloutier, Sicard-Roselli, Fromm, & Sanche, 2009; Folkard et al., 1993; Nikjoo, O’Neill, Goodhead, & Terrissol, 1997).

An important parameter to compare the effects on biological material of different ionizing radiations, such as different energy levels of EB, is the relative biological effectiveness (RBE), which is a dimensionless value that describes the effectiveness of one type of ionizing radiation in comparison to a reference radiation. The higher the RBE of an ionizing radiation type, the more effective it is compared to the other one. Bellamy, Puskin, Hertel, and Eckerman (2015) reported an RBE > 1 for low energy electrons compared with the reference of 1 MeV electron radiation, implying a higher antimicrobial efficiency of low energy compared with high energy EB.

3.3. Low energy electron beam treatment of dry food surfaces

Table 3 provides a comprehensive overview about the application of LEEB for microbial inactivation on dry food products. The first studies were carried out by researchers in Japan at the end of the 20th century (Hayashi et al., 1997, 1998a, 1998b; Hayashi & Todoriki, 1999; Todoriki & Hayashi, 2000). The authors showed that LEEB using electron energies between 60 and 210 keV enables inactivation of the native microbial flora of different kinds of seeds (radish, alfalfa, etc.), varieties of rice and wheat, as well as herbs and spices (black and white peppercorn, coriander, basil). Hayashi et al. (1997) treated different rice varieties, wheat and buckwheat, and they reported a minimum electron energy for microbial inactivation between 75 and 160 keV and a necessary dose to reduce the native microbial load < 10 CFU g^{-1} between 6 and 20 kGy depending on the product. For the treatment of black peppercorns, a dose of 10–15 kGy and electron energy of 210 keV was

Table 3
Overview of studies investigating the decontamination of dry food products by low energy electron beam (LEEB). Some values/numbers have been approximated from graphs provided in the cited literature. (CFU: Colony forming unit).

Reference	Product	LEEB source	Treatment parameter	Organism	Comments
Hayashi et al. (1997)	Rough rice (RR), brown rice (BR), wheat (WH), unhulled buckwheat (BW)	Van de Graaff electron accelerator (Nissin High Voltage Engineering Co. Ltd., Kyoto, Japan)	Acceleration voltage 180–300 kV electron energy at sample surface ~75–210 keV	Native microbial flora RR ~4.7×10^7 CFU g^{-1} BR ~4.1×10^6 CFU g^{-1} WH ~2.7×10^4 CFU g^{-1} BW ~1.4×10^6 CFU g^{-1}	• Necessary dose to reduce microbial load < 10 CFU g^{-1}: RR 15–20 kGy, BR 7–13 kGy, WH 7–12 kGy, BW 6–8 kGy • Necessary electron energy for inactivation: RR 160 keV, BR 75 keV, WH 100 keV, BW 130 keV
Hayashi et al. (1998a)	Brown rice	Van de Graaff electron accelerator (Nissin High Voltage Engineering Co. Ltd., Kyoto, Japan)	Acceleration voltage 170–200 kV electron energy at sample surface ~60–100 keV	Native microbial flora (1.8×10^6–2.8×10^5 CFU g^{-1})	• Dose of 10–15 kGy to reduce the microbial load < 10 CFU g^{-1} • Effect irrespective of electron energy
Hayashi et al. (1998b)	Black pepper (BP), white pepper (WP), coriander (CO), basil (BA)	Van de Graaff electron accelerator (Nissin High Voltage Engineering Co. Ltd., Kyoto, Japan)	Acceleration voltage 180–300 kV electron energy at sample surface ~75–210 keV	Native microbial flora BP ~2.8×10^7 CFU g^{-1} WP ~6.0×10^6 CFU g^{-1} CO ~3.4×10^5 CFU g^{-1}	• Necessary dose to reduce the microbial load < 10 CFU g^{-1} for BP 10–15 kGy • Necessary electron energy for inactivation: BP 210 keV, WP 100 keV, CO 100 keV, BA 100 keV • Increasing electron energy up to 210 keV reduced treatment time by factor 6
Hayashi and Todoriki (1999)	Radish, alfalfa seeds	Van de Graaff electron accelerator (Nissin High Voltage Engineering Co. Ltd., Kyoto, Japan)	Acceleration voltage 170–190 kV electron energy at sample surface ~60–90 keV	Native microbial flora	• Necessary electron energy for complete inactivation: radish 60 keV, alfalfa 75 keV
Todoriki and Hayashi (2000)	Adzuki bean (AB), pot herb mustard (PH), black gram (BG) seeds	Van de Graaff electron accelerator (Nissin High Voltage Engineering Co. Ltd., Kyoto, Japan)	Acceleration voltage 170–200 kV electron energy at sample surface ~60–100 keV	Native microbial flora AB ~7.5×10^3 CFU g^{-1} PH ~1.2×10^4 CFU g^{-1} BG ~6.1×10^5 CFU g^{-1}	• Necessary dose to reduce the microbial load < 10 CFU g^{-1} ≥ 10 kGy • Necessary electron energy for inactivation: AB 170 keV, PH 170 keV, BG 190 keV
Todoriki et al. (2002)	Soybean	Van de Graaff electron accelerator (Nissin High Voltage Engineering Co. Ltd., Kyoto, Japan)	Acceleration voltage 170 kV electron energy at sample surface ~60 keV	Native microbial load (~1.1×10^3 CFU g^{-1})	• 10 min treatment, 7.5 kGy reduced the microbial load < 10 CFU g^{-1}
Baba et al. (2004)	Wheat, brown rice	Soft Electron Processor (EBC-150-50-45) with 500 kg h^{-1}	Acceleration voltage of 130 kV for wheat and 150 kV for brown rice	Native microbial load	• D-values for wheat and brown rice with 14 and 12.5 kGy
Trinetta et al. (2011)	Cantaloupe (CA), tomato (TO), lettuce (LE) seeds	E-beam application development unit (Advanced Electron Beams, Wilmington, MA)	Electron energy 150 keV, dose of 7 kGy	*Salmonella enterica* Poona (CA ~6.14 $\log_{10}$ CFU g^{-1}, TO ~5.86 $\log_{10}$ CFU g^{-1}) *Escherichia coli* O157:H7 (LE ~5.47 $\log_{10}$ CFU g^{-1})	• CA: reduced to ~3.90 $\log_{10}$ CFU g^{-1} • TO: reduced to ~1.50 $\log_{10}$ CFU g^{-1} • LE: reduced to ~3.26 $\log_{10}$ CFU g^{-1}
Fan et al. (2017)	Mung bean (MB), red clover (RC), fenugreek (FE) seeds	REAMODE electron accelerator (Fraunhofer Institute for Electron Beam and Plasma Technology, Dresden, Germany)	Acceleration voltage of 140 kV, dose of 4, 8, 12 kGy	*Escherichia coli* K12 MB: ~5.93 $\log_{10}$ CFU g^{-1} RC: ~7.30 $\log_{10}$ CFU g^{-1} FE: ~7.92 $\log_{10}$ CFU g^{-1}	• Dose of 12 kGy inactivated *E. coli* K12 in more than 80% of seeds
Gryczka, Migdał, and Bułka (2017)	Black peppercorns (BP), onion flakes (OF), bay leaves (BL)	Accelerator ILU-6	Electron energy 200 and 300 keV	Native microbial load BP: ~6.49 $\log_{10}$ CFU g^{-1} OF: ~5.51 $\log_{10}$ CFU g^{-1} BL: ~5.60 $\log_{10}$ CFU g^{-1}	• BP: no reduction after 10 min, reduced by ~2.0 $\log_{10}$ after 30 min • OF: reduced by ~0.6 $\log_{10}$ after 12 min • BL: reduced by ~2.6 $\log_{10}$ after 12 min

needed to reduce the native microbial load < 10 CFU g^{-1}. The decontamination of white peppercorns, coriander and basil required a minimum electron energy of 100 keV, but increasing the electron energy to 210 keV reduced the treatment time by a factor of 6 (Hayashi et al., 1998b). Complete microbial inactivation of alfalfa and radish seeds required an electron energy of up to 75 keV (Hayashi & Todoriki, 1999), whereas an energy of up to 190 keV was necessary for inactivation of the native microbial flora on Adzuki bean, pot herb mustard and black gram seeds (Todoriki & Hayashi, 2000). Baba, Kaneko, and Taniguchi (2004) used a Soft Electron Processor with a throughput of 500 kg h^{-1} for the treatment of wheat and brown rice and reported a required dose of 14 and 12.5 kGy to reduce the microbial load by 90%, respectively. Trinetta, Vaidya, Linton, and Morgan (2011) inoculated *S. enterica* Poona on cantaloupe and tomato seeds and *E. coli* O157:H7 on lettuce seeds and treated them with LEEB (150 keV, 7 kGy). The authors reported an inactivation depending on the treated seeds, with the highest inactivation determined for tomato seeds.

LEEB can also be applied to inactivate seed-borne fungi that cause smut and bunt diseases (Röder et al., 2009). Furthermore, this technology also offers the possibility of inactivating the eggs of insect pests. A treatment dose of 0.48 kGy with an electron energy of 60 keV was sufficient to inactivate the eggs of *Callosobruchus chinensis* on the surface of adzuki beans, whereas the larvae inside the beans were not completely inactivated (Imamura et al., 2004).

The limited and controlled penetration depth of the low energy electrons reduced their effect on the product surface. Thus, it can be assumed that product-process interactions, which could affect the product quality, can be minimized. Hayashi et al. (1997) investigated the viscosity of a grain suspension after LEEB treatment. The researchers reported a decrease in viscosity with an increasing electron energy. However, the viscosity of grain suspensions treated with LEEB doses, necessary to reduce native microbial load < 100 CFU g^{-1}, was higher than those of grains exposed to γ-rays. The results suggested that the low energy electrons degraded only starch molecules near the surface. Todoriki, Kikuchi, Nakaoka, Miike, and Hayashi (2002) compared the gelatinized properties of soymilk of beans decontaminated with LEEB and temperature-treated soymilk, whereas the gelatinized properties were higher for LEEB treated samples. Hayashi et al. (1998a) also reported increasing TBA values for brown rice with an increasing electron energy, indicating lipid oxidation. The germination behavior of LEEB-treated seeds was also investigated, but no effect of the treatment on germination was shown (Fan et al., 2017; Todoriki & Hayashi, 2000; Trinetta et al., 2011).

3.4. Limiting factors

The main limiting factor of LEEB technology is probably the limited penetration depth of the low energy electrons, as well as limited knowledge in the science and industry community. Ghomi et al. (2005) investigated the antimicrobial efficiency of pulsed LEEB (80 keV) by treating different concentrations of *E. coli*. The authors reported a reduction of bacterial inactivation with increasing initial concentrations, which was attributed to the formation of cell agglomerates for higher concentration. The top microorganism layer was inactivated, but it also worked as an attenuator of the low energy electrons, thus reducing the antimicrobial effect, similarly to other surface decontamination technologies such as CAPP. Urgiles et al. (2007) compared LEEB (100 keV) and EB (10 MeV) by treating *Bacillus pumilus*, *megaterium* and *subtilis* spores. For LEEB, D-values of 1.34, 3.46 and 1.01 kGy were reported, respectively. Applying EB with 10 MeV resulted in much higher D-values of 2.12, 4.11 and 2.05 kGy. These results show the high potential of LEEB for surface decontamination since the electron energy is deposited on the surface. However, 100 keV has been reported to cause complete inactivation of the spores following a dose > 30 kGy for LEEB (Urgiles et al., 2007). The FDA (2017) has recommended a minimum dose of 44 kGy for the sterilization of food products by ionizing radiation used solely for space flight programs.

Hayashi et al. (1997) and Hayashi et al. (1998b) reported different required electron energies and doses for the decontamination of different rice varieties, wheat, buckwheat and black and white peppercorns. Similar results were reported by Trinetta et al. (2011) and Fan et al. (2017) for the treatment of different seeds. These findings indicated an impact of the treated surface morphology on the inactivation efficiency of LEEB, demonstrating that a complex surface structure can lower the antimicrobial efficiency of the treatment. The distance between the point of electron emission to the atmosphere and the product can also impact the inactivation process. The passage of electrons through the atmosphere to the target reduces their kinetic energy due to collisions with atmospheric molecules. Electrons with energies of 80 keV have a maximum “lifetime” in air of 7.1 cm, but during their traversal of this pathway, electrons can lose energy due to collisions (Ghomi et al., 2005).

4. Upscaling, consumer acceptance, regulatory and future research needs

To date, no commercial CAPP process for the decontamination of dry products has been applied in industry because no adequate CAPP systems are currently available that are validated, scalable to industrial requirements and cost-effective. The research community is focusing on process parameters, such as plasma sources and process gases, for maximization of microbial inactivation and minimization of the impact on the product quality, since a minimal impact on product quality is an essential factor for the implementation and acceptance of a novel food processing technology (Pankaj, Wan, & Keener, 2018). However, focusing on the application of air-plasma is probably recommended since the use of air is much more cost-effective than noble gases. Niemira (2012a) evaluated the process gas costs regarding the scale-up to industrial systems and showed the significant difference in process costs of CAPP systems using a noble gas such as helium, or air as well as mixtures of oxygen and nitrogen. The process gas costs for 1000 h of operation, calculating with a gas consumption of 500–4000 l min^{-1}, or 300–2400 m^3 h^{-1}, would be: noble gas helium $636,000-$9,096,000; nitrogen $9000-$72,000; oxygen $18,000-$144,000 and air ~$0. Even though the addition of noble gases as a minor component of the process gas can enhance the inactivation efficiency, the specific application must be verified to justify the higher process gas costs (Niemira, 2012a). In addition, plasma equipment that is tailored for the food industry must be developed and designed, which can be easily integrated into existing process lines. Such equipment must be practical to operate, provide competitive economics compared with established technologies and guarantee safety by adequate insulation, grounding and shielding. An ideal industrial scale plasma system should be able to continuously process products with different characteristics, such as powders, granular and/or flat materials. Furthermore, a uniform treatment even for complex shaped products must be ensured. A key factor in this regard is a homogenous spatial distribution of the reactive plasma components. CAPP applications in the material science and medical sector demonstrated that non-uniformity treatment can be mitigate by the arrangement and geometry of the CAPP sources (Homola et al., 2017; Nie, Cao, Ren, Wang, & Kong, 2009). A primary focus for CAPP process validation is the complex plasma chemistry. Air-plasma can generate more than 75 reactive species with approximately hundreds of simultaneous reactions (Keener & Misra, 2016; Sakiyama et al., 2012), which could lead to potentially undesirable product-process interactions. The regulatory approval for a CAPP process will require a large amount of data and time efforts because the complex plasma chemistry with its possible effects on the product must still be analyzed and assessed. Acceptance of data or conclusions reached from a regulatory review may significantly differ between countries due to the different regulatory requirements for such novel food processes (Keener & Misra, 2016). The most effective way to validate a plasma-

based decontamination process would by a case-by-case assessment of a certain product. However, the manner in which consumers will react to products treated with cold atmospheric pressure plasma technology is still not clear. According to the Technology Readiness Level (TRL), an established system to estimate the technology maturity, CAPP can be rated with TRL 5, for which key elements have been demonstrated in a relevant environment. Moreover, the high variety of CAPP sources and set-ups opens the possibility of potential combination processes, such as simultaneous decontamination and drying processes. Future research should also focus on the promising results for the degradation of mycotoxins on different dry products to understand and optimize the degradation mechanisms.

LEEB belongs to ionizing radiation regulation, and thus this technology is subject to the same legal regulations as other irradiation technologies considering the treatment of food products and labeling. In the United States, a maximum irradiation dose of 30 kGy is allowed for dry and dehydrated aromatic substances such as herbs and spices. In the European Union, the application of irradiation to dried aromatic herbs, spices and vegetable seasoning is harmonized, and a maximum average absorbed dose of 10 kGy is allowed (Pillai & Shayanfar, 2017). Although ionizing radiation has proven to be efficient, environmentally clean and energy-effective, it is rarely used in the EU because of its poor consumer acceptance in Europe (Schweiggert et al., 2007). Erroneously, the main consumer concerns in the EU have focused on the perceived “carcinogenicity” of irradiated foods and the association with ‘radioactivity’ (Frewer et al., 2011). However, a joint FAO/IAEA/WHO Study Group on High-Dose Irradiation (JSGHD, 1999) concluded that, from the perspective of human safety, any food may be irradiated at any dose, as reflected in a revision of the Codex General Standard for Irradiated Foods” (CAC, 2003) The first prototype “Soft Electron Processor” for the decontamination of wheat and rice with a performance of 500 $kg\,h^{-1}$ was constructed by Nissin-High Voltage Co. in Japan (Baba et al., 2004) in 2004. However, to reduce 90% of the native microbial load, a dose of > 10 kGy was necessary, making this LEEB application not suitable for commercial use in the European Union. Nevertheless, it has recently been shown that LEEB is able to fulfill industrial requirements for the spice and herb industries, and a first industrial scale prototype, with TRL 7, was introduced in the second quarter of 2017 on the European market, demonstrating a capacity of one ton per hour (IIA, 2017). Although LEEB is already used on an industrial scale, studies investigating the impact on food products, and the potential for mycotoxin degradation are scarce. However, food products produced under established Good Manufacturing Practice and treated with doses greater than 10 kGy can be considered nutritionally adequate and safe (FAO/IAEA/WHO, 1999).

5. Conclusions

The demand by consumers for safe and high quality food requires the research community and industry to develop new technologies with clear pathways into the market. CAPP and LEEB are emerging nonthermal technologies with high potential for the gentle decontamination of dry food surfaces. Due to the limited and controlled penetration depth of both technologies, product-process interactions can be minimized by maintaining the product quality. LEEB has already been successfully introduced into the seed dressing as well as packaging industries and the first demonstrator for the decontamination of herbs and spices has been installed in the market. Compared with LEEB, CAPP is at an advanced development stage with TRL 5, and scalable systems for industrial requirements are needed. LEEB is at an early demonstration phase where research on several food commodities might be still necessary.

There is an urgent need for the introduction of novel technologies to consumers, since consumers are not only concerned regarding the safety and quality of their products but also regarding the applied technologies. The inclusion of all stakeholders in discussions based on scientific evidence and data could lead to successful market launches of these promising emerging surface decontamination techniques in the very near future.

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References

Baba, T., Kaneko, H., & Taniguchi, S. (2004). Soft electron processor for surface sterilization of food material. *Radiation Physics and Chemistry, 71*(1–2), 207–209.
Bellamy, M., Puskin, J., Hertel, N., & Eckerman, K. (2015). An empirical method for deriving RBE values associated with electrons, photons and radionuclides. *Radiation Protection Dosimetry, 167*(4), 664–670.
Booth, I. R. (1985). Regulation of cytoplasmic pH in bacteria. *Microbiological Reviews, 49*(4), 359–378.
Boudam, M. K., Moisan, M., Saoudi, B., Popovici, C., Gherardi, N., & Massines, F. (2006). Bacterial spore inactivation by atmospheric-pressure plasmas in the presence or absence of UV photons as obtained with the same gas mixture. *Journal of Physics D: Applied Physics, 39*(16), 3494–3507.
Bourke, P., Zuizina, D., Han, L., Cullen, P. J., & Gilmore, B. (2017). Microbiological interactions with cold plasma. *Journal of Applied Microbiology, 123*(2), 308–324.
Brun, É., Cloutier, P., Sicard-Roselli, C., Fromm, M., & Sanche, L. (2009). Damage induced to DNA by low-energy (0-30 eV) electrons under vacuum and atmospheric conditions. *Journal of Physical Chemistry B, 113*(29), 10008–10013.
Buckenhüskes, H. J., & Rendlen, M. (2004). Hygienic problems of phytogenic raw materials for food production with special emphasis to herbs and spices. *Food Science and Biotechnology, 13*, 262–268.
Bugaev, S. P., Korovin, S. D., Kutenkov, D. P., Landi, V. F., Mesyats, G. A., & Sakharov, E. S. (1994). Surface sterilization using low-energy nanosecond pulsed electron beams. *10th international conference on high-power particle beams* (pp. 817–820). .
Butscher, D., Van Loon, H., Waskow, A., von Rohr, P. R., & Schuppler, M. (2016a). Plasma inactivation of microorganisms on sprout seeds in a dielectric barrier discharge. *International Journal of Food Microbiology, 238*, 222–232.
Butscher, D., Zimmermann, D., Schuppler, M., & Rudolf von Rohr, P. (2016b). Plasma inactivation of bacterial endospores on wheat grains and polymeric model substrates in a dielectric barrier discharge. *Food Control, 60*, 636–645.
Bußler, S., Rumpold, B. A., Fröhling, A., Jander, E., Rawel, H. M., & Schlüter, O. K. (2016). Cold atmospheric pressure plasma processing of insect flour from Tenebrio molitor: Impact on microbial load and quality attributes in comparison to dry heat treatment. *Innovative Food Science & Emerging Technologies, 36*, 277–286.
CAC (Codex Alimentarius Commission) (2003). *Codex general standard for irradiated foods.* CODEX STAN 106-1983. Rev. 1-2003.
Chalise, P., Hotta, E., Matak, K., & Jaczynski, J. (2007). Inactivation kinetics of *Escherichia coli* by pulsed electron beam. *Journal of Food Science, 72*(7), M280–M285.
Chalise, P., Rahman, M., Ghomi, H., Hayashi, Y., Watanabe, M., Okino, A., ... Hotta, E. (2004). Bacterial inactivation using low-energy pulsed-electron beam. *IEEE Transactions on Plasma Science, 32*(4), 1532–1539.
Choi, S., Puligundla, P., & Mok, C. (2016). Microbial decontamination of dried Alaska pollock shreds using corona discharge plasma jet: Effects on physicochemical and sensory characteristics. *Journal of Food Science, 81*(4), M952–M957.
Comet Group (2012a). Tetra Pak unveils COMET's innovative e-beam technology at Anuga 2012. Retrieved September 11, 2017, from http://www.comet-group.com/news/2012/03/tetra-pak-unveils-comet-s-innovative-e-beam-technology-at-anuga-2012.
Comet Group (2012b). *e-Beam Technology*. Flamatt, Switzerland: Brochure.
Dasan, B. G., Boyaci, I. H., & Mutlu, M. (2016). Inactivation of aflatoxigenic fungi (*Aspergillus* spp.) on granular food model, maize, in an atmospheric pressure fluidized bed plasma system. *Food Control, 70*, 1–8.
Dasan, B. G., Boyaci, I. H., & Mutlu, M. (2017). Nonthermal plasma treatment of *Aspergillus* spp. spores on hazelnuts in an atmospheric pressure fluidized bed plasma system: Impact of process parameters and surveillance of the residual viability of spores. *Journal of Food Engineering, 196*, 139–149.
Deng, S., Ruan, R., Mok, C. K., Huang, G., Lin, X., & Chen, P. (2007). Inactivation of *Escherichia coli* on almonds using nonthermal plasma. *Journal of Food Science, 72*(2), M62–M66.
Deng, X. T., Shi, J. J., Shama, G., & Kong, M. G. (2005). Effects of microbial loading and sporulation temperature on atmospheric plasma inactivation of *Bacillus subtilis* spores. *Applied Physics Letters, 87*(15), 153901.
Dobrin, D., Magureanu, M., Mandache, N. B., & Ionita, M.-D. (2015). The effect of non-thermal plasma treatment on wheat germination and early growth. *Innovative Food Science & Emerging Technologies, 29*, 255–260.
Eliasson, B., & Kogelschatz, U. (1991). Nonequilibrium volume plasma chemical processing. *IEEE Transactions on Plasma Science, 19*(6), 1063–1077.
Fan, X., Sokorai, K., Weidauer, A., Gotzmann, G., Rögner, F., & Koch, E. (2017). Comparison of gamma and electron beam irradiation in reducing populations of *E. coli* artificially inoculated on mung bean, clover and fenugreek seeds, and affecting germination and growth of seeds. *Radiation Physics and Chemistry, 130*, 306–315.
FAO/IAEA/WHO (1999). *High-dose irradiation: Wholesomeness of food irradiated with doses above 10 kGy.* WHO.

Farkas, J., & Mohácsi-Farkas, C. (2014). Spices and seasonings. In Y. Motarjemi, G. Moy, & E. Todd (Eds.). *Encyclopedia of food safety* (pp. 324–330). (1st ed.). London: Academic Press.

FDA (2017). *CFR - code of federal regulations title 21*. Retrieved October 6, 2017, from https://www.accessdata.fda.gov/scripts/cdrh/cfdocs/cfcfr/CFRSearch.cfm?CFRPart=179&showFR=1.

Fernández, A., Shearer, N., Wilson, D. R., & Thompson, A. (2012). Effect of microbial loading on the efficiency of cold atmospheric gas plasma inactivation of *Salmonella enterica* serovar Typhimurium. *International Journal of Food Microbiology, 152*(3), 175–180.

Fine, F., & Gervais, P. (2005). Thermal destruction of dried vegetative yeast cells and dried bacterial spores in a convective hot air flow: Strong influence of initial water activity. *Environmental Microbiology, 7*(1), 40–46.

Folkard, M., Prise, K. M., Vojnovic, B., Davies, S., Roper, M. J., & Michael, B. D. (1993). Measurement of DNA damage by electrons with energies between 25 and 4000 eV. *International Journal of Radiation Biology, 64*(6), 651–658.

Frewer, L. J., Bergmann, K., Brennan, M., Lion, R., Meertens, R., Rowe, G., ... Vereijken, C. (2011). Consumer response to novel agri-food technologies: Implications for predicting consumer acceptance of emerging food technologies. *Trends in Food Science & Technology, 22*(8), 442–456.

Fröhling, A., Baier, M., Ehlbeck, J., Knorr, D., & Schlüter, O. (2012). Atmospheric pressure plasma treatment of *Listeria innocua* and *Escherichia coli* at polysaccharide surfaces: Inactivation kinetics and flow cytometric characterization. *Innovative Food Science & Emerging Technologies, 13*, 142–150.

Ghomi, H., Rahman, M., Chalise, P., Hayashi, Y., Watanabe, M., Okino, A., ... Hotta, E. (2005). Experimental investigation of effect of low-energy pulsed atmospheric electron beam on bacterial cells. *Japanese Journal of Applied Physics, Part 1: Regular Papers and Short Notes and Review Papers, 44*(12), 8698–8701.

Gryczka, U., Migdał, W., & Bułka, S. (2017). The effectiveness of the microbiological radiation decontamination process of agricultural products with the use of low energy electron beam. *Radiation Physics and Chemistry, 39*, 59–62.

Gurtler, J., Doyle, M., & Kornacki, J. (2014). *The microbiological safety of low water activity foods and spices.* New York: Springer.

Hayashi, T., Okadome, H., Toyoshima, H., Todoriki, S., & Ohtsubo, K. (1998a). Rheological properties and lipid oxidation of rice decontaminated with low-energy electrons. *Journal of Food Protection, 6*(1), 73–77.

Hayashi, T., Takahashi, Y., & Todoriki, S. (1997). Low-energy electron effects on the sterility and viscosity of grains. *Journal of Food Science, 62*(4), 858–860.

Hayashi, T., Takahashi, Y., & Todoriki, S. (1998b). Sterilization of foods with low-energy electrons ('soft-electrons'). *Radiation Physics and Chemistry, 52*(1–6), 73–76.

Hayashi, T., & Todoriki, S. (1999). Effects of soft-electrons on the microbial counts and germination of seeds of radish sprout (Daikon) and alfalfa. *Nippon Shokuhin Kagaku Kogaku Kaishi, 46*(11), 754–757.

Hertwig, C., Leslie, A., Meneses, N., Reineke, K., Rauh, C., & Schlüter, O. (2017b). Inactivation of *Salmonella* Enteritidis PT30 on the surface of unpeeled almonds by cold plasma. *Innovative Food Science & Emerging Technologies, 44*, 242–248.

Hertwig, C., Reineke, K., Ehlbeck, J., Erdogdu, B., Rauh, C., & Schlüter, O. (2015c). Impact of remote plasma treatment on natural microbial load and quality parameters of selected herbs and spices. *Journal of Food Engineering, 167*, 12–17.

Hertwig, C., Reineke, K., Ehlbeck, J., Knorr, D., & Schlüter, O. (2015a). Decontamination of whole black pepper using different cold atmospheric pressure plasma applications. *Food Control, 55*, 221–229.

Hertwig, C., Reineke, K., Rauh, C., & Schlüter, O. (2017a). Factors involved in *Bacillus* spore's resistance to cold atmospheric pressure plasma. *Innovative Food Science & Emerging Technologies, 43*, 173–181.

Hertwig, C., Steins, V., Reineke, K., Rademacher, A., Klocke, M., Rauh, C., et al. (2015b). Impact of surface structure and feed gas composition on *Bacillus subtilis* endospore inactivation during direct plasma treatment. *Frontiers in Microbiology, 6*, 1–12.

Homola, T., Shekargoftar, M., Dzik, P., Krumpolec, R., Ďurašová, Z., Veselý, M., et al. (2017). Low-temperature (70 °C) ambient air plasma- fabrication of inkjet-printed mesoporous TiO2 flexible photoanodes. *Flexible and Printed Electronics, 2*, 35010.

IIA (2017). Decontamination of dry food ingredients by low energy electron beam: A world first. Retrieved September 29, 2017, from http://iiaglobal.com/news/decontamination-dry-food-ingredients-low-energy-electron-beam-world-first/.

Imamura, T., Todoriki, S., Sota, N., Nakakita, H., Ikenaga, H., & Hayashi, T. (2004). Effect of "soft-electron" (low-energy electron) treatment on three stored-product insect pests. *Journal of Stored Products Research, 40*(2), 169–177.

Jeon, J., Klaempfl, T. G., Zimmermann, J. L., Morfill, G. E., & Shimizu, T. (2014). Sporicidal properties from surface micro-discharge plasma under different plasma conditions at different humidities. *New Journal of Physics, 16*.

Keener, K. M., & Misra, N. N. (2016). Future of cold plasma in food processing. *Cold plasma in food and agriculture* (pp. 343–360). Elsevier Inc.

Kim, J. W., Puligundla, P., & Mok, C. (2015a). Dielectric barrier discharge plasma for microbial decontamination of dried laver: Effects on physicochemical characteristics. *International Journal of Food Science and Technology, 50*(12), 2630–2638.

Kim, J. W., Puligundla, P., & Mok, C. (2015b). Microbial decontamination of dried laver using corona discharge plasma jet (CDPJ). *Journal of Food Engineering, 161*(2), 24–32.

Kim, J. W., Puligundla, P., & Mok, C. (2017). Effect of corona discharge plasma jet on surface-borne microorganisms and sprouting of broccoli seeds. *Journal of the Science of Food and Agriculture, 97*(1), 128–134.

Laroussi, M. (2002). Nonthermal decontamination of biological media by atmospheric-pressure plasmas: Review, analysis, and prospects. *IEEE Transactions on Plasma Science, 30*(4), 1409–1415.

Laroussi, M., & Leipold, F. (2004). Evaluation of the roles of reactive species, heat, and UV radiation in the inactivation of bacterial cells by air plasmas at atmospheric pressure. *International Journal of Mass Spectrometry, 233*(1–3), 81–86.

Lee, K. H., Kim, H. J., Woo, K. S., Jo, C., Kim, J. K., Kim, S. H., ... Kim, W. H. (2016). Evaluation of cold plasma treatments for improved microbial and physicochemical qualities of brown rice. *LWT - Food Science and Technology, 73*, 442–447.

Lerouge, S., Wertheimer, M. R., Marchand, R., Tabrizian, M., & Yahia, L. (2000). Effect of gas composition on spore mortality and etching during low-pressure plasma sterilization. *Journal of Biomedical Materials Research, 51*(1), 128–135.

Li, J., Sakai, N., Watanabe, M., Hotta, E., & Wachi, M. (2013). Study on plasma agent effect of a direct-current atmospheric pressure oxygen-plasma jet on inactivation of *E. coli* using bacterial mutants. *IEEE Transactions on Plasma Science, 41*(4), 935–941.

Lu, X., & Wu, S. (2013). On the active species concentrations of atmospheric pressure nonequilibrium plasma jets. *IEEE Transactions on Plasma Science, 41*(8), 2313–2326.

Mendis, D. A., Rosenberg, M., & Azam, F. (2000). A Note on the possible electrostatic disruption of bacteria. *IEEE Transactions on Plasma Science, 28*(4), 1304–1306.

Mitra, A., Li, Y. F., Klämpfl, T. G., Shimizu, T., Jeon, J., Morfill, G. E., et al. (2014). Inactivation of surface-borne microorganisms and increased germination of seed specimen by cold atmospheric plasma. *Food and Bioprocess Technology, 7*(3), 645–653.

Moeller, R., Setlow, P., Horneck, G., Berger, T., Reitz, G., Rettberg, P., ... Nicholson, W. (2008). Roles of the major, small, acid-soluble spore proteins and spore-specific and universal DNA repair mechanisms in resistance of *Bacillus subtilis* spores to ionizing radiation from X rays and high-energy charged-particle bombardment. *Journal of Bacteriology, 190*(3), 1134–1140.

Moisan, M., Barbeau, J., Moreau, S., Pelletier, J., Tabrizian, M., & Yahia, L. H. (2001). Low-temperature sterilization using gas plasmas: A review of the experiments and an analysis of the inactivation mechanisms. *International Journal of Pharmaceutics, 226*(1–2), 1–21.

Muranyi, P., Wunderlich, J., & Heise, M. (2008). Influence of relative gas humidity on the inactivation efficiency of a low temperature gas plasma. *Journal of Applied Microbiology, 104*(6), 1659–1666.

Nicholson, W. L., Munakata, N., Horneck, G., Melosh, H. J., & Setlow, P. (2000). Resistance of *Bacillus* endospores to extreme terrestrial and extraterrestrial environments. *Microbiology and Molecular Biology Reviews, 64*(3), 548–572.

Nie, Q., Cao, Z., Ren, C., Wang, D., & Kong, M. (2009). A two-dimensional cold atmospheric plasma jet array for uniform treatment of large-area surfaces for plasma medicine. *New Journal of Physics, 11*, 115015.

Niemira, B. (2012a). Cold plasma decontamination of foods. *Annual Review of Food Science and Technology, 3*, 125–142.

Niemira, B. (2012b). Cold plasma reduction of *Salmonella* and *Escherichia coli* O157:H7 on almonds using ambient pressure gases. *Journal of Food Science, 77*(3), M171–M175.

Nikjoo, H., & Lindborg, L. (2010). RBE of low energy electrons and photons. *Physics in Medicine and Biology, 55*(10), R65–R109.

Nikjoo, H., O'Neill, P., Goodhead, D. T., & Terrissol, M. (1997). Computational modelling of low-energy electron-induced DNA damage by early physical and chemical events. *International Journal of Radiation Biology, 71*(5), 467–483.

Padan, E., & Schuldiner, S. (1987). Intracellular pH and membrane potential as regulators in the prokaryotic cell. *Journal of Membrane Biology, 198*, 189–198.

Palumbo, M., Beuchat, L., Danyluk, M., & Harris, L. (2015). Recalls of tree nuts and peanuts in the US, 2001 to present [Table and references]. U.S. Recalls of Nuts, 1–12. Retrieved December 01, 2017, from http://ucfoodsafety.ucdavis.edu/files/43637.pdf.

Pan, Z., Bingol, G., Brandl, M., & McHugh, T. (2012). Review of current technologies for reduction of *Salmonella* populations on almonds. *Food and Bioprocess Technology, 5*(6), 2046–2057.

Pankaj, S., Wan, Z., & Keener, K. (2018). Effects of cold plasma on food quality: A review. *Foods, 7*(1), 4.

Park, J. C., Park, B. J., Han, D. W., Lee, D. H., Lee, I. S., Hyun, S. O., ... Takatori, K. (2004). Sterilization using microwave-induced argon plasma at atmospheric pressure. *Journal of Microbiology and Biotechnology, 14*(1), 188–192.

Patil, S., Moiseev, T., Misra, N. N., Cullen, P. J., Mosnier, J. P., Keener, K. M., et al. (2014). Influence of high voltage atmospheric cold plasma process parameters and role of relative humidity on inactivation of *Bacillus atrophaeus* spores inside a sealed package. *Journal of Hospital Infection, 88*(3), 162–169.

Pillai, S. D., & Shayanfar, S. (2015). *Electron beam pasteurization and complementary food processing technologies.* Cambridge: Woodhead Publishing Limited.

Pillai, S., & Shayanfar, S. (2017). Electron beam technology and other irradiation technology applications in the food industry. *Topics in Current Chemistry, 375*(1), 1–20.

Pillai, S., & Shayanfar, S. (2018). Electron beam processing of fresh produce – A critical review. *Radiation Physics and Chemistry, 143*, 85–88.

Rahman, M., Ghomi, H., Chalise, P., Hayashi, Y., Watanabe, M., Okino, A., ... Hotta, E. (2006). Inactivation of cells and spores of *Bacillus subtilis* using low energy pulsed electron beam. *Japanese Journal of Applied Physics, 45*(33), L881–L883.

Randeniya, L. K., & De Groot, G. J. J. B. (2015). Non-thermal plasma treatment of agricultural seeds for stimulation of germination, removal of surface contamination and other benefits: A review. *Plasma Processes and Polymers, 12*(7), 608–623.

Reineke, K., Langer, K., Hertwig, C., Ehlbeck, J., & Schlüter, O. (2015). The impact of different process gas compositions on the inactivation effect of an atmospheric pressure plasma jet on *Bacillus* spores. *Innovative Food Science & Emerging Technologies, 30*, 112–118.

Röder, O., Jahn, M., Schröder, T., Stahl, M., Kotte, M., & Beuermann, S. (2009). Die e-ventus Technologie - eine Innovation zur nachhaltigen Reduktion von Pflanzenschutzmitteln mit Empfehlung für Bio-Saatgut/E-ventus technology – an innovative treatment method for sustainable reduction in the use of pesticides with recommendation for organic seed. *Journal Für Verbraucherschutz Und Lebensmittelsicherheit, 4*(2), 107–117.

Sakiyama, Y., Graves, D., Chang, H., Shimizu, T., & Morfill, G. (2012). Plasma chemistry model of surface microdischarge in humid air and dynamics of reactive neutral

species. *Journal of Physics D: Applied Physics, 45*, 425201.
Schlüter, O., Ehlbeck, J., Hertel, C., Habermeyer, M., Roth, A., Engel, K., ... Eisenbrand, G. (2013). Opinion on the use of plasma processes for treatment of foods. *Molecular Nutrition & Food Research, 57*(5), 920–927.
Schweiggert, U., Carle, R., & Schieber, A. (2007). Conventional and alternative processes for spice production – a review. *Trends in Food Science & Technology, 18*(5), 260–268.
Setlow, P. (2007). I will survive: DNA protection in bacterial spores. *Trends in Microbiology, 15*(4), 172–180.
Shi, H., Ileleji, K., Stroshine, R. L., Keener, K., & Jensen, J. L. (2017). Reduction of aflatoxin in corn by high voltage atmospheric cold plasma. *Food and Bioprocess Technology, 10*(6), 1042–1052.
Siciliano, I., Spadaro, D., Prelle, A., Vallauri, D., Cavallero, M. C., Garibaldi, A., et al. (2016). Use of cold atmospheric plasma to detoxify hazelnuts from aflatoxins. *Toxins, 8*(5), 1–10.
Stoffels, E., Sakiyama, Y., & Graves, D. B. (2008). Cold atmospheric plasma: Charged species and their interactions with cells and tissues. *IEEE Transactions on Plasma Science, 36*(4), 1441–1457.
Sun, S., Anderson, N. M., & Keller, S. (2014). Atmospheric pressure plasma treatment of black peppercorns inoculated with *Salmonella* and held under controlled storage. *Journal of Food Science, 79*(12), E2441–E2446.
Surowsky, B., Schlüter, O., & Knorr, D. (2014). Interactions of non-thermal atmospheric pressure plasma with solid and liquid food systems: A review. *Food Engineering Reviews, 7*(2), 82–108.
Tahergorabi, R., Matak, K. E., & Jaczynski, J. (2012). Application of electron beam to inactivate *Salmonella* in food: Recent developments. *Food Research International, 45*(2), 685–694.
Ten Bosch, L., Pfohl, K., Avramidis, G., Wieneke, S., Viöl, W., & Karlovsky, P. (2017). Plasma-based degradation of mycotoxins produced by *Fusarium, Aspergillus* and *Alternaria* species. *Toxins, 9*(3), 1–12.
Thirumdas, R., Deshmukh, R. R., & Annapure, U. S. (2016). Effect of low temperature plasma on the functional properties of basmati rice flour. *Journal of Food Science & Technology, 53*(6), 2742–2751.
Todoriki, S., & Hayashi, T. (2000). Disinfection of seeds and sprout inhibition of potatoes with low energy electrons. *Radiation Physics and Chemistry, 57*(3–6), 253–255.
Todoriki, S., Kikuchi, O., Nakaoka, M., Miike, M., & Hayashi, T. (2002). Soft electron (low energy electron) processing of foods for microbial control. *Radiation Physics and Chemistry, 63*(3–6), 349–351.
Trinetta, V., Vaidya, N., Linton, R., & Morgan, M. (2011). A comparative study on the effectiveness of chlorine dioxide gas, ozone gas and e-beam irradiation treatments for inactivation of pathogens inoculated onto tomato, cantaloupe and lettuce seeds. *International Journal of Food Microbiology, 146*(2), 203–206.
Urgiles, E., Wilcox, J., Montes, O., Osman, S., Venkateswaran, K., Cepeda, M., ... Pillai, S. (2007). Electron beam irradiation for microbial reduction on spacecraft components. *IEEE Aerospace Conference Proceedings,* 1–15.
Wang, S., Doona, C., Setlow, P., & Li, Y. (2016). Use of Raman spectroscopy and phase-contrast microscopy to characterize cold atmospheric plasma inactivation of individual bacterial spores. *Applied and Environmental Microbiology, 82*(19), 5775–5784.
Yu, H., Perni, S., Shi, J. J., Wang, D. Z., Kong, M. G., & Shama, G. (2006). Effects of cell surface loading and phase of growth in cold atmospheric gas plasma inactivation of *Escherichia coli* K12. *Journal of Applied Microbiology, 101*(6), 1323–1330.
Zahoranová, A., Henselová, M., Hudecová, D., Kaliňáková, B., Kováčik, D., Medvecká, V., et al. (2016). Effect of cold atmospheric pressure plasma on the wheat seedlings vigor and on the inactivation of microorganisms on the seeds surface. *Plasma Chemistry and Plasma Processing, 36*(2), 397–414.