

# **A STUDY ON COLD PLASMA FOR FOOD PRESERVATION**

## **ABSTRACT:**

Cold plasma is an electrically energized matter composed of highly reactive species which includes molecules of charged and gas particles in the form of positive and negative ions, photons electrons, free radicals at room temperature. It is an emerging technology in non-thermal food preservation in the application of sterilization. An increase in the plasma-based treatment for food is employed to inactivate the food borne pathogens seen in the recent years. The present study reviews the action of plasma agents on the microbial population, surface decontamination of the raw produce in the food processing and future novelty in food technology.

**Key words:** cold plasma, Food, preservation, sterilization.

## **Introduction**

The matter is any entity that has weight/mass and occupies space. The matter is known to exist in three distinct phases. However, there is a fourth phase called plasma. So, Plasma is hence referred to as the fourth state of matter. The term 'Plasma' was first employed by Irving Langmuir in 1928 to define this fourth state of matter which is partially or wholly ionized state of gas and discovered plasma oscillations in ionized gas. Matter changes from solid to liquid and further to gas occurs as we increase the energy input likewise increasing the energy input beyond a certain level in gas state causes ionization of molecules to the plasma state. D Agostino et al. reported that plasma could be obtained either at low temperature, non-equilibrium glow discharge or high temperature, equilibrium thermal plasma.

From the properties of plasma, it is used in various fields such as textile, electronics, life sciences, packaging etc. Roth et al. Application of the plasma technology as a surface cleaning tool has been commercially adopted for the removal of disinfection chemicals applied to medical devices manufactured from heat sensitive plastics. Moisan et al. In the biomedical sector plasma technology used for cold sterilization of instruments and prostheses

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30 as well as many thermolabile materials used in the biomedical technology sector for its  
31 particular advantages, such as including its moderate or negligible impact on substrate  
32 materials and use on nontoxic compounds. Conventionally, sterilization methods such as  
33 heat, chemical solutions are used for the surface disinfection of fruits, seeds, and spices etc.,  
34 which are often time-consuming and damaging or have toxic residues. Van de Veen et al.  
35 reported that the effect of cold plasma on bacterial spores is more than the conventional  
36 techniques like heat, and chemicals and UV treatment. One of the important challenges  
37 associated with cold plasma technology is ensuring high microbial inactivation while  
38 maintaining sensory qualities and that ensure their fresh appearance |

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39 The three conventional states of matter are solids, liquids, and gases; plasma has been  
40 described as the fourth state of matter, an unfamiliar designation that warrants explanation.  
41 As materials acquire energy (such as by heating), they change state, from solid (lowest  
42 energy) to liquid and then ultimately to gas. The melting points and boiling points of  
43 materials widely vary. For all materials, however, at each phase transition, the interactions  
44 and structures between molecules become loose and ultimately breakdown entirely (Niemira  
45 2012). Gases are collections of molecules (e.g., N<sub>2</sub>, O<sub>2</sub>, CO<sub>2</sub>) or single atoms (e.g., He,  
46 Ne, Ar) without large-scale structure. |

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47 At still higher energies, the intra molecular and intra-atomic structures break down,  
48 liberating free electrons and ions. The plasma may be thought to be an ionized gas consisting  
49 of neutral molecules, electrons, and positive and negative ions. Plasmas generated in  
50 conventional devices do not ionize all of the atoms in a gas, even for hot (i.e., thermal)  
51 plasmas, such as welding arcs and spark plugs (Fridman *et al.* 2005). Within these hot  
52 plasmas, all species are extremely reactive. Within cooler (i.e., nonthermal) plasmas, such as  
53 those found in neon signs and plasma display screens, some of the chemical species are more  
54 reactive than others. For this reason, the chemical composition of the feed gas becomes a  
55 determining factor in the types of reactions that the plasma can initiate (Lieberman  
56 & Lichtenberg 2005, Niemira & Gutsol 2010).

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57 The energy required to ionize gases into plasma can come from a variety of sources,  
58 such as heat, electricity, laser light, radiation, and extremely rapid compression. As a cloud of  
59 active particles, the plasma retains the imparted energy for a period. When the active particles  
60 recombine with each other, the energy is released as visible and UV light in the process of  
61 recombination (Lieberman & Lichtenberg 2005, Niemira 2012). Of more interest to food |

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processors, the active particles in the plasma can react with the food substrate, releasing the stored energy into the bacteria or viruses to be targeted. How much energy a plasma has to impart will depend on its chemical composition, density, and temperature.

## Plasma Science (Plasma- Definition, Physics, and Chemistry)

In 1922, the American scientist Irving Langmuir proposed that the electrons, ions and neutrals in an ionized gas could be considered as corpuscular material entrained in some kind of fluid medium and termed this entraining medium "plasma", similar to the plasma, introduced by the Czech physiologist Jan Evangelista Purkinje to denote the clear fluid which remains after removal of all the corpuscular material in blood. However, it emerged that there was no "fluid medium" entraining the electrons, ions, and neutrals in ionized gas (Bellan 2015), nevertheless the name prevailed.

The term "plasma" refers to a partially or wholly ionized gas composed essentially of photons, ions, and free electrons as well as atoms in their fundamental or excited states possessing a net neutral charge. The plasma possesses a net neutral charge because the number of positive charge carriers is equal to the number of negative ones (Kudra and Mujumdar 2009). Electrons and photons are usually designated as "light" species in contrast to the rest of the constituents designated as "heavy" species. Due to its unique properties plasma is often referred to as the fourth state of matter according to a scheme expressing an increase in the energy level from solid to liquid to gas and ultimately to plasma.

## Definition of Technology

Thermal plasma, operating at many hundreds or thousands of degrees above ambient, would be immediately detrimental to the quality of food products. A nonthermal plasma is therefore the focus. For the sake of clarity, however, a distinction must be made between what nonthermal means to a plasma physicist and what the same term means to a food processor. To the physicist, non-thermal means that the plasma has a distinctly non uniform distribution of energy (a non-equilibrium) among the constituent particles. Electrons are likely to transfer energy via collisions with heavier particles, exciting the larger particle in to a state of reactivity (Fridman et al. 2005, Niemira & Gutsol 2010). To a food processor, non-thermal means that the mode of action of the antimicrobial process does not rely on thermal kill for inactivation of associated pathogens. As a practical matter, non-thermal processes are regarded as those that cause little or no thermal damage to the food product being treated.

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93 There are three primary mechanisms by which cold plasma inactivates microbes (Moisanetal.  
94 2002).

- 95 • The first is the chemical interaction of radicals, reactive species, or charged particles  
96 with cell membranes.
- 97 • The second is by damage to membranes and internal cellular components by UV  
98 radiation.
- 99 • Finally, DNA strands may be broken by UV generated during recombination of the  
100 plasma species.

101 While on a given commodity, one mode of action may be more significant than another, the  
102 greatest sanitizing efficacy results from plasma with multiple antimicrobial mechanisms  
103 (Moisan et al. 2002, Laroussi 2003). As a food processing technology, cold plasma is new  
104 enough that the terminology is still evolving. The terms cold plasma (Noriega *et al.* 2011),  
105 cool plasma (Tran et al. 2008), atmospheric pressure plasma(Chirokovetal.2005),cold  
106 atmospheric gas plasma (Moisanetal.2001),and other comparable terms have been used in  
107 recent publications. In other cases, the plasma is described by the generative technology, e.g.,  
108 dielectric barrier discharge (Fridman *et al.* 2006), plasma jet (Lu etal.2009), uniform glow  
109 discharge plasma(Gadrietal.2000),gliding arc discharge (Burlicaetal. 2010), etc.

## 110 **Types of Plasma**

111 Two classes of plasma, namely thermal and Non-thermal plasma (NTP) can be  
112 distinguished on the basis of conditions in which they are generated. This classification of  
113 plasma is based on the relative energetic levels of electrons and heavy species of the plasma.  
114 NTP (near ambient temperatures of 30-60°C) is obtained at atmospheric or reduced pressures  
115 (vacuum) and requires less power. NTPs are characterized by an electron temperature much  
116 above that of the gas (macroscopic temperature) and consequently, do not present a local  
117 thermodynamic equilibrium. NTP is also generated by an electric discharge in gas at lower  
118 pressure or using microwaves.

119 Typical illustrations for plasma generation at atmospheric pressure include the corona  
120 discharge, Dielectric barrier discharges (DBD), Radio-frequency plasmas (RFP) and the  
121 gliding arc discharge. To the contrary, thermal plasmas are generated from higher pressures  
122 and require high power. Thermal equilibrium may exist in between the electrons and the

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heavy species. Plasma generation at atmospheric pressure is of interest, both technically and industrially for the food industries because this does not require extreme conditions.

**A thermal plasma** is characterized by the existence of a thermodynamic equilibrium between the electrons, ions and neutral particles. The temperatures of a thermal plasma at an atmospheric pressure generally are above 6000 K. This corresponds to a mean kinetic energy of less than 1 eV.

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**A non-thermal plasma** has significantly different electron and gas temperatures. For example, the electron temperature may be several 10,000 K, which corresponds to a mean kinetic energy of more than 1 eV, whereas the gas temperature can be close to ambient. In spite of their low temperature, such plasmas can trigger chemical reactions and excitation states via electron impact. In Contrast thermal plasma, non-thermal plasma can also be applied directly to thermally sensitive surfaces.

### Plasma Sources

Usually, plasma treatments was carried out under vacuum conditions, but researchers have developed atmospheric pressure plasma system, resulting reduced cost, increased treatment speed, and industrial applicability (Yoon and Ryu 2007; Yun *et al.* 2010). The ability to generate non-thermal plasma discharges at atmospheric pressure makes the decontamination process easier and less expensive (Kim *et al.* 2011). Nevertheless recently, most of the cold plasma devices available commercially was developed for research to aim at biomedical applications. Therefore, for food applications, these devices may need to be customized or tailor made. The barrier glow discharge generated between two parallel electrodes is a widely employed NTP system.

Food may be conveyed through the discharge to achieve microbial decontamination in most of the industrial scale. Another configuration is the plasma pen or jet, in which a stream of gases can be directed at the object to be treated. Biozone, a Scientist has developed the new process for the generation of the cold oxygen plasma (COP) by using air to high- energy deep UV light with a effective radiation spectrum between 180 nm & 270 nm. This cold gas plasma, composed of several species of negative and positive ions, free radical molecules, electron, UV-photons and ozone (Terrier *et al.* 2009). Duo-Plasma line is linearly extended plasma source excited using microwaves of 2.45 GHz at a pressure <1000 Pa (Petasch *et al.* 1997) and several other plasma treatment systems have evolved based on this principle. The

154 Plasmodul is a microwave sustained low pressure plasma reactor with a modular concept  
155 based on the Duo-Plasmaline principle which provides an easy up scaling for industrial  
156 applications (Schulz *et al.*).

157 This type of microwave excited plasma sources are well suited for large area plasma  
158 treatment (Petasch *et al.* 1997) and can probably be employed for surface treatment of foods  
159 or processing surfaces at industrial scale. More recently, Kim et al. (2010) developed a cold  
160 plasma jet operating at 20 kHz Alternating Current (AC) under atmospheric pressure. The  
161 most changeable feature of most plasma systems is the freedom to select a gas or gas mixture.  
162 Improvements in the existing plasma systems and newer equipment directed for treatment of  
163 real food systems are likely to draw attention of researchers and engineers in near future.

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164 Recently a novel approach which shows significant potential for the treatment of  
165 various foods has been reported. The approach is based on a dielectric barrier discharge with  
166 the food package in contact with high voltage electrodes. Only 40-50 W of power is needed  
167 to ionize air inside a 4 L re-sealable plastic (LDPE) bag (Klockow and Keener 2009). The  
168 high voltage process ionizes any gas within the electric field contained within the package.  
169 Ionization can generate significant amounts of reactive molecules with little increase in  
170 product surface temperature. Particular treatment times for targeted spore or bacterial  
171 reductions are dependent on product loading, packaging material, gas composition and  
172 package/electrode configuration. The in-package ionization process has been demonstrated in  
173 a number of common packaging materials including cardboard, glass, LDPE, HDPE, PETE,  
174 polystyrene, rubber, tygon, and others. Scale-up of the system has facilitated treatment of air  
175 filled packages with an electrode gap of up to 10 cm with rapid processing times (Keener et  
176 al. 2010).

## 177 **Types of Cold Plasma Systems**

178 There is a rapidly expanding array of technologies used to generate cold plasma.  
179 These can operate at atmospheric pressure or at some degree of partial vacuum. The gas  
180 being ionized may be as simple as air or nitrogen, or it may be a more exotic mixture  
181 containing some proportion of noble gases, such as helium, argon, or neon. The driving  
182 energy may be electricity, microwaves, or lasers. This wide array of design elements is an  
183 indication of the flexibility of cold plasma systems and the extent to which new forms of cold  
184 plasma systems continue to be built and evaluated.

185           However, all cold plasma systems intended for use in food processing fall generally  
186 into one of three categories. These categories are defined by where the food to be treated is  
187 positioned with respect to the cold plasma being generated: at some significant distance from  
188 the point of generation, relatively close to the point of generation, or within the plasma  
189 generation field itself. Conceptually, these categories are derived from the nature of cold  
190 plasma chemistry, with delineations having to do with the half-life and reactivity of charged,  
191 active species within the plasma (Niemira & Gutsol 2010).

192           The first category is remote treatment cold plasma systems. The plasma is generated  
193 using one of a variety of methods and moved onto the surface to be treated. The plasma may  
194 be driven by a flow of the feed gas or (less commonly) manipulated through the use of  
195 magnetic fields. This type of system has the advantage of placing the surface to be treated at a  
196 physically separate point of generation (Chirokov et al. 2005). This simplifies the design and  
197 operation of the device, and increases the flexibility with respect to the shapes and sizes of  
198 objects to be treated. However, the most reactive chemical species are also those that have the  
199 shortest half-life. During the time of flight, free electrons may recombine with other plasma  
200 products, such as heavy ions or atomic species. By the time the quenched plasma reaches the  
201 target surface, the composition is secondary chemical species, i.e. lower activity, long-living  
202 chemical species resulting from chemical recombination within the plasma (Gadri et al.  
203 2000).

204           The lower concentration of ions that exist in this afterglow plasma generate UV light  
205 and activate chemical species upon reaction with the target, but their concentration is much  
206 lower than in active plasma (i.e., plasma supported by electric field) (Fridman & Kennedy  
207 2004). These Cond category is known as direct treatment cold plasma systems. In this  
208 systems, plasma generation instrument supplies active plasma directly to the object to be  
209 treated. As with the first category, the plasma is moved via the flow of the feed gas or by a  
210 comparable means. So the target is relatively close to the site of cold plasma generation &  
211 exposed to the plasma before active species recombine and are lost, these systems provide  
212 higher concentrations of active agents (Laroussi&Lu2005). Systems of this type can operate  
213 in pulsed mode, with plasma generated at pulse frequencies of hundreds or thousands of  
214 times per second.

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## 217 **Cold-Plasma Generators**

218 In this section, several methods that have been used to generate relatively large volumes  
219 of non-equilibrium cold plasmas, at or near atmospheric pressure (sometimes referred to as  
220 “high” pressure) are presented. This is not a comprehensive list of all existing methods. The  
221 methods presented here were chosen for two main reasons.

222 1. They have been used extensively to study the germicidal effects of cold, high-  
223 pressure plasmas; and

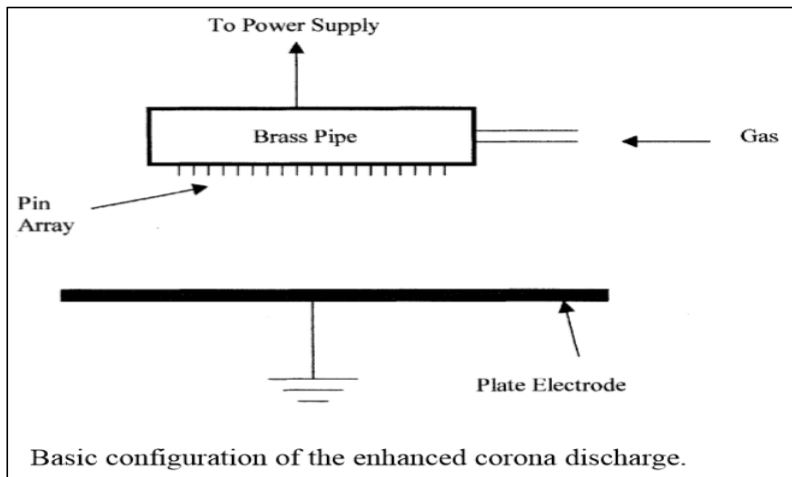
224 their potential use in various other industrial plasma processing applications (lighting,  
225 surface modification, etching, deposition).

## 226 **The Corona Discharge**

227 Siemens (2005) was the first to suggest the use of a corona discharge to generate  
228 ozone in order to disinfect water supplies. This was the first recorded use of plasma toward  
229 the inactivation of micro-organisms. Menashi(1972) used a pulsed RF-driven corona  
230 discharge to create a plasma at atmospheric pressure. He reported that up to microbial spores  
231 could be inactivated in less than 1 s. Garate(1978) et al. used an “Enhanced Corona  
232 Discharge” to destroy concentrations of up to  $4.10^6$ /ml of *Escherichia coli*, and spores of  
233 *Bacillus subtilis* in less than 15 min. A schematic of the enhanced corona discharge is shown  
234 in Fig. This discharge consists of a line of pins fastened to a hollow pipe at one end and  
235 protruding from the other end through tiny holes.

236 The feed gas escapes through the holes and provides a local atmosphere around the  
237 corona points. The feed gas, a non-electronegative gas such as helium or argon, replaces the  
238 air around the corona points and therefore enhances the discharge by removing the electron-  
239 attaching electronegative-oxygen molecules. The pin array can be biased by a dc or ac high-  
240 voltage supply, or by a pulsed power supply.





**Fig:1:The Glow Discharge at Atmospheric Pressure**

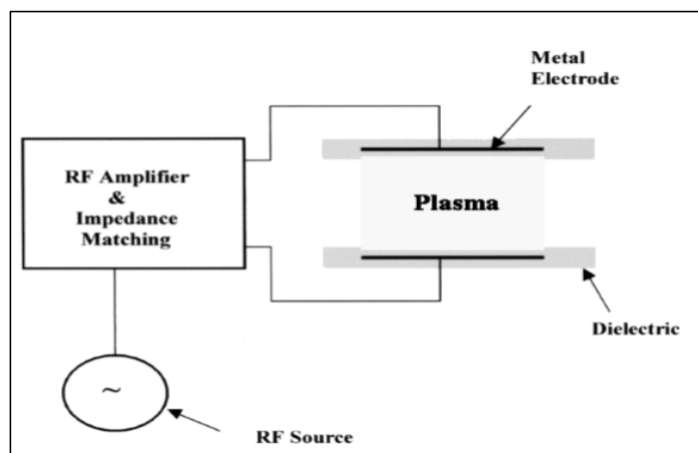
One of the early developments of diffuse glow discharge plasma at atmospheric pressure was reported by Donohoe. Donohoe(1979) used a large gap (cm) pulsed-barrier discharge in a mixture of helium and ethylene to polymerize ethylene. Later, Kanazawa et al(1979). reported their development of a stable glow discharge at atmospheric pressure by using a dielectric-barrier discharge (DBD) configuration. They claimed that to obtain a diffuse discharge (as opposed to a filamentary discharge, which is traditionally produced by DBDs), helium had to be the major constituent of the gas mixture, and the frequency of the applied voltage had to be in the kilohertz range. Schematic of the DBD-based glow discharge at atmospheric pressure. At least one of the two electrodes must be covered by a dielectric material. After the ignition of the discharge, charged particles are collected on the surface of the dielectric.

This charge build-up creates a voltage drop, which counteracts the applied voltage, and therefore chokes the discharge current. The discharge subsequently extinguishes. As the applied voltage increases again (at the second half cycle of the applied voltage) the discharge reignites. This process is repeated over and over during each full cycle of the applied voltage. Laroussi(2003), reported the use of the glow discharge at atmospheric pressure to destroy cells of *Pseudomonas* fluorescence. He used suspensions of the bacteria in Petri dishes placed on a dielectric-covered lower electrode. The electrodes were placed within a chamber containing mostly helium with an admixture of air. He obtained full destruction of concentrations of  $4.10^6/\text{ml}$  in less than 10 min. Using a similar discharge, Kelly-Win

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tenberg(2000). reported the inactivation of *B. subtilis* spores using an air gap. *E. coli*, *B. subtilis*, and a variety of other gram-negative as well as gram-positive bacteria were inactivated successfully by many researchers using the DBD-based diffuse-glow discharge.

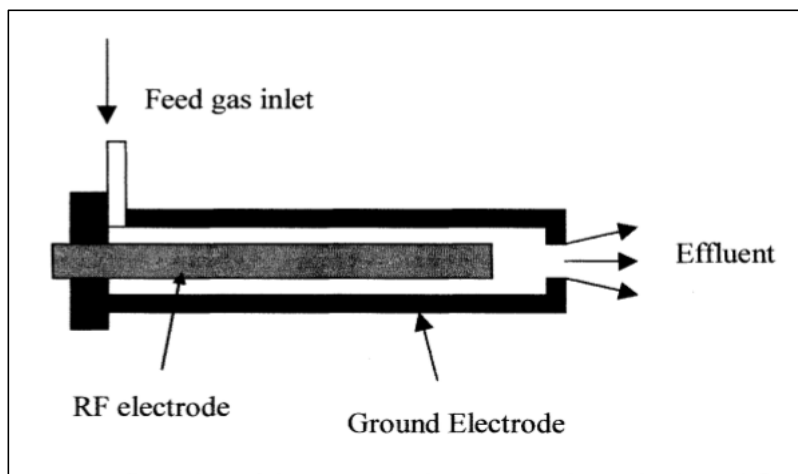


**Fig.2: Configuration of the DBD-based diffuse glow discharge at atmospheric pressure.**

### **The Atmospheric-Pressure Plasma Jet**

The atmospheric-pressure plasma jet (APPJ) is a capacitively coupled device consisting of two coaxial electrodes between which a gas flows at high rates. The outer electrode is grounded, while the central electrode is excited by RF power at 13.56 MHz. The free electrons are accelerated by the RF field and enter into collisions with the molecules of the background gas. These inelastic collisions produce various reactive species (excited atoms and molecules, free radicals) that exit the nozzle at high velocity. The reactive species can therefore react with a contaminated surface placed in proximity (cm) of the nozzle.

As in the case of the diffuse DBD, the stability of the APPJ plasma (as well as its nonthermal characteristic) depend on using helium as a carrier gas. Herrmann et al(1999). used the APPJ to inactivate spores of *Bacillus globigii*, a simulant to Anthrax (*Bacillus anthracis*). Herrmann et al(1999). reported the reduction of seven orders of magnitude of the original concentration of *B. globigii* in about 30 s.

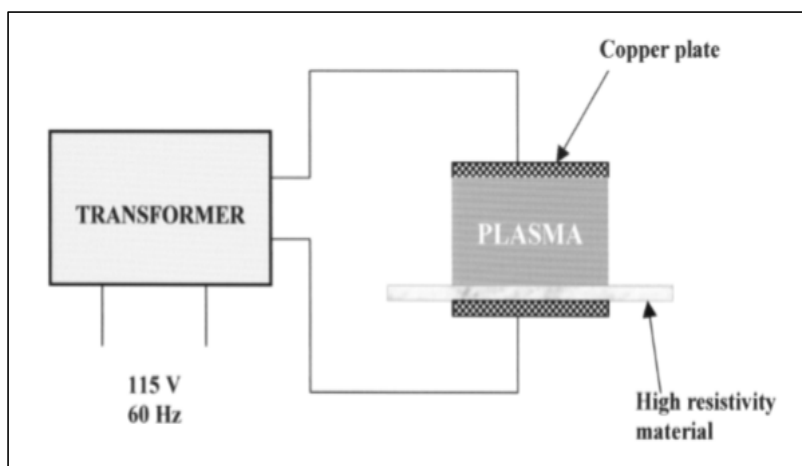


**Fig:3: Configuration of the atmospheric-pressure plasma jet (APPJ)**

### **The Resistive Barrier Discharge**

The concept of the resistive-barrier discharge (RBD) is based on the DBD configuration. However, instead of a dielectric, a high-resistivity material is used to cover at least one of the electrodes. The high-resistivity layer plays the role of a distributed ballast which limits the discharge current and therefore prevents arcing. The advantage of the RBD over the DBD is the possibility to use dc power (or low-frequency ac, 60 Hz) to drive the discharge.

Using helium, large-volume diffuse cold plasma at atmospheric pressure can be generated. Richardson et al. and Laroussi et al. reported a fourorders-of-magnitude reduction in the original concentration of vegetative *B. subtilis* cells in about 10 min. They also reported that the RBD-inactivated endospores of *B. subtilis*, but not as effectively as the vegetative cells. In these experiments, they used a gas mixture of 97%–3% helium-oxygen, respectively.



**Fig4 : Configuration of the resistive-barrier discharge (RBD)**

#### **Action of Plasma on microorganisms**

##### **Action on cell components and functions**

The use of sterilizing properties of plasma was first introduced towards the end of 60s, patented in 1968 (Menashi 1968) and first works with plasma made from oxygen were proposed in 1989. Thereafter, considerable research has been performed on the mechanism of microbial inactivation by plasma agents. The plasma agents contribute to the lethal action by interacting with the biological material. Nelson and Berger (1989) have shown that O<sub>2</sub> plasma could be a very efficient biocidal against bacteria.

Plasma treatment can effectively inactivate a wide range of micro-organisms including spores (Kelly-Wintenberg et al. 1999; Feichtinger et al. 2003; Lee et al. 2006) and viruses (Terrier et al. 2009). Effect of plasma can be quite selective, meaning tuneable between damage to pathogenic organisms without damage to the host, or activation of different pathways in different organisms (Dobrynin et al. 2009).

Low-pressure oxygen plasma has been shown to degrade lipids, proteins, and DNA of cells (Mogul et al. 2003). The reactive species in plasma have been widely associated to the direct oxidative effects on the outer surface of microbial cells. As an example, commonly used oxygen and nitrogen gas plasma are excellent sources of reactive oxygen-based and nitrogen-based species, such as O<sup>•</sup>, O<sub>2</sub>, O<sub>3</sub>, OH<sup>•</sup>, NO<sup>•</sup>, NO<sub>2</sub>, etc. Atomic oxygen is

337 potentially a very effective sterilizing agent, with a chemical rate constant for oxidation at  
338 room temperature of about 106 times that of molecular oxygen (Critzter et al. 2007).

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339 These act on the unsaturated fatty acids of the lipid bilayer of the cell membrane,  
340 thereby impeding the transport of bio-molecules across it. The double bonds of unsaturated  
341 lipids are particularly vulnerable to ozone attack (Guzel-Seydim et al. 2004). Membrane  
342 lipids are assumed to be more significantly affected by the reactive oxygen species (ROS)  
343 due to their location along the surface of bacterial cell, which allows them to be bombarded  
344 by these strong oxidizing agents (Montie et al. 2002). The proteins cells and the spores are  
345 equally vulnerable to the action of these species, causing denaturation and cell leakage.  
346 Oxidation of amino acids and nucleic acids may also cause changes that result in microbial  
347 death or injury (Critzter et al. 2007).

348 Micro-organisms in coldplasma are exposed to an intense bombardment by radicals,  
349 most likely provoking surface lesions that the living cell cannot repair sufficiently faster. This  
350 may partially explain the observations where in cells are in many cases destroyed very  
351 quickly. This process is termed “etching” (Pelletier 1992). The cell wall rupture has been  
352 additionally attributed by Laroussi et al., (2003) and Mendis et al., (2002) to electrostatic  
353 forces due to accumulation of charges at the outer surface of cell membranes. The  
354 morphological changes in E. coli cells treated with atmospheric plasma at 75W for 2 min as  
355 observed under an electron microscope by (Hong et al. 2009), clearly revealed that the treated  
356 cells had severe cytoplasmic deformations and leakage of bacterial chromosome.

357 These observations demonstrate the loss of viability of bacterial cells after plasma  
358 treatment. An analogy between plasma and pulsed electric field has also been drawn to  
359 explain the action of plasma on the membranes (Pothakamury et al. 1995; Spilimbergo et al.  
360 2003). It is well established that electroporation of membranes is induced by pulsed electric  
361 fields and it appears that plasma acts on similar lines inducing perforations in the membranes  
362 of micro-organisms (Sale and Hamilton 1967; Pothakamury et al. 1995; Wouters and Smelt  
363 1997). In addition to generating pores, humid air plasma additionally provokes marked  
364 acidification of the medium (Moreau et al. 2005; Moreau et al. 2007).

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## 368 **Role of UV photons and charged particles**

369 The production of UV photons of different wavelengths has been proposed to be  
370 involved in dimerizing the thymine bases of DNA including that of spores (Munakata et al.  
371 1991).

372 The role of UV photons in bacterial death when they are submitted to a plasma  
373 treatment was reviewed in detail by (Boudam et al. 2006). More recently, by exclusion of the  
374 reactive particles and spectral fractions of UV radiation from access to the spores Roth et al.,  
375 (2010) revealed that UV-C radiation is the most effective inactivation agent in the plasma.  
376 Ultraviolet (UV) photons play a less important role in atmospheric pressure glow discharge  
377 (APGD) because they are easily absorbed by gas atoms and molecules at atmospheric  
378 pressure (Vleugels et al. 2005).

379 The role of the charged particles in the bacterial inactivation process was recently  
380 investigated by Lu et al. (2009). Their work revealed that the charged particles play a minor  
381 role in the inactivation process when He/N<sub>2</sub> (3%) is used as working gas than when He/O<sub>2</sub>  
382 (3%) is used. Also, they concluded that heat and UV play no or minor roles in the  
383 inactivation process. Similar results were earlier obtained by (Perni et al. 2007) who  
384 interplayed bacterial inactivation kinetics with optical emission spectroscopy, and identified  
385 oxygen atoms as a major contributor in plasma inactivation with minor contributions from  
386 UV photons, OH radicals, singlet oxygen metastables and nitric oxide. Thus, a contradiction  
387 over the role of UV photons in plasma exists, and future studies must be directed to get a  
388 clear picture.

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## 389 **Effect of process parameters**

390 The concentrations in which the plasma agents occur in plasma depend greatly on the  
391 device set-up (reactor geometry), operating conditions (gas pressure, type, flow, frequency  
392 and power of plasma excitation) and gas composition which affect their efficacy in a process  
393 when employed. To cite an example, the destructive efficiency of various gas plasma sources  
394 and temperatures on *Bacillus* spp. Spores were compared by (Hury et al. 1998).

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395 This group demonstrated that oxygen-based plasma is more efficient than pure argon  
396 plasma. Another deciding criterion is whether the substrate to be sterilized is in direct contact  
397 with the plasma (Direct Exposure) or located remote from it (Remote Exposure) (Moisan et  
398 al. 2001; Laroussi 2005; Boudam et al. 2006). If exposed remotely, the quantum of heat

transmitted to a sample is reduced, the charged particles do not play a role since they recombine before reaching the sample, and many of the short-lived neutral reactive species also do not reach the sample.

Since, the components of the plasma are reactive and self-quenching, with a relatively short half-life, decreased time of flight would be expected to be one of the major factors in antimicrobial efficacy in this case (Niemira and Sites 2008).

By varying the process parameters involved in plasma generation, a multitude of mechanisms can be actuated which may act individually or synergistically. Nevertheless, the details of the interaction of the different plasma agents with the different components of bacterial cells or spores are currently very limited. The interactions which occur between plasma agents and biological materials, ultimately leading to sterilization are still under investigation.

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#### Potential Applications:

#### Results of action of cold plasma on packing materials

Packing materials	Treatment plasma	Applied voltage (dosage)	Results	References
Polypropylene	Air Corona	30 kHz, 1.7 j/cm <sup>2</sup>	Decrease in contact angle, increase in adhesion	D. Dixon, B.J. et.al. (2012)
PET films	Glow discharge	10 W	Decrease in contact angle, increase in roughness, crystallinity and degradation yield	K.N. Pandiyaraj, et.al. (2008)
Potato starch film	Air plasma	15W	Decrease in hydrophilic nature, Increase in tensile strength	F.Starzyk, et.al. (2001)
PP film	Diode plasma discharge	8.3 W	Decrease in contact angle, increase in surface energy	P. Slepicka, et. Al.(2010)
PET film	Jet plasma DCdischarge	35 W	Increase in hydrophobic nature	Y. Akishev, et al., (2008)
PET	Jet plasma	285 V	Increase in weight, decrease in contact angle and wettability	K. Gotoh, et.al.,(2011)
HDPE film	RFAr:O <sub>2</sub> Plasma	150 W	Decrease in crystallinity, contact angle, increase in roughness	I. Banik,K.S. Kim, et al., (2002)

LDPE	RFArgon plasma	25–100 W	Decrease in contact angle and ageing effect, increase in crystallinity	M. Ataefard, et.al.,(2009)
BOPP film	RFAir plasma	10–50 W	Decrease in contact angle, increase, roughness ageing effect	S.M. Mirabedini, et. Al.,(2007)

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414 The above table shows: Variability in chemical nature of cold plasma depends upon the  
415 material with which it is packed. Treatment with disparate electric discharges, cold plasma  
416 tends to amend its chemical properties all together.

417 **Recent findings in the area of non-thermal plasmas for inactivation of microorganisms**  
418 **and spores.**

Organism	Plasma conditions	Treatment surface/ medium	Salient result	Reference
<i>Escherichia coli</i> , <i>Staphylococcus aureus</i>	Atmospheric plasma corona discharge, with high voltage (20kV) DC power supply	On agar plates	Changes of pH levels from alkaline to acid, upon plasma application to bacteria in water, does not play a predominant role in cell death.	Korachi et al. (2010)
<i>Escherichia coli</i> , <i>Bacillus subtilis</i> , <i>Candida albicans</i> , and <i>Staphylococcus aureus</i>	High-frequency capacitive discharge (0.4 torr) and barrier discharge (0.4-0.5 torr) in air excited at commercial frequency of 5.28 MHz	Glass plate and petri dish	The most probable sterilization agents of the plasma generated were established to be "hot" and "cold" OH radicals, the excited electrically neutral N <sub>2</sub> and O <sub>2</sub> molecules, and the UV plasma radiation	Azharonok et al. (2009)
<i>Escherichia coli</i> KCTC1039 <i>Bacillus subtilis</i>	Helium and Oxygen based electric discharge plasma produced at a radio frequency (RF) of 13.56 MHz	Dried cells and endospore suspension on a cover-glass	Treated cells had severe cytoplasmic deformations and leakage of bacterial chromosome. UV from the plasma only slightly affected the viability of the spores.	Hong et al. (2009)
<i>Escherichia coli</i> type 1 <i>Saccharomyces</i>	Cold atmospheric plasma plume generated by an	Inoculated membrane filters and	Efficacy of inactivation was markedly reduced for microorganisms on the cut	Perni et al. (2008a)



<i>cerevisiae</i> <i>Gluconobacter</i> <i>liquefaciens</i> <i>Listeria</i> <i>monocytogenes</i>	AC voltage of 8 kV at 30 kHz	inoculated fruit surfaces	surfaces than on filters due to the migration of microorganisms from the exterior of the fruit tissue to its interior and not quenching of reactive plasma species.	
<i>Escherichia coli</i> <i>Saccharomyces cerevisiae</i> <i>Pantoea agglomerans</i> <i>Gluconacetobacter liquefaciens</i>	The cold atmospheric plasma generated by an AC voltage (variable 12kV and 16kV)	Pericarps of mangoes and melons	<i>S. cerevisiae</i> was the most resistant amongst all test organisms. An increase in the applied voltage led to more efficient production of reactive plasma species (oxygen atoms) which was attributed for better inactivation.	Perni et al. (2008b)
<i>Escherichia coli</i> O157:H7 <i>Salmonella</i> sp. <i>Listeria monocytogenes</i>	One atmosphere uniform glow discharge plasma (OAUGDP) operated at 9 kV power and 6 kHz frequency	Apples, Cantaloupe and Lettuce	Inactivation was observed in all the cases. Extent of log reduction varied with the organisms	Critzer et al. (2007)
Biofilms produced by <i>Chromobacterium violaceum</i>	RF high pressure cold plasma jet using Atomflo 250 reactor with 100 W RF power supply using He and N <sub>2</sub> gas	Biofilms produced in 96-well polystyrene microplates	A 10 min plasma treatment was able to kill almost 100% of the cells. A complex, biphasic model of inactivation was observed.	Abramzon et al. (2006)

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#### 420 Recent finding of microbial inactivation using cold plasma

Microorganism	Substrate	Plasma source	Exposure time and dosage	Results	Reference
<i>S. enteritidis</i> (O1) <i>L. monocytogenes</i>	Table Egg	RBD prototype	90 min & 30kV	4–5 Log reduction	N. Rowan, et. Al., (2007)
<i>E. coli</i> 12955 & <i>Salmonella</i> spp	almonds	Dielectric discharge	30 s & 30 kV and 2000 Hz.	4 log CFU/ml	Deng S, et. Al., 2007
<i>E. coli</i> , <i>C. jejuni</i>	Chicken skin	Pulsed gas plasma discharge	24 s at 45kV	up to 8 Log reduction	E. Noriega, et.al., (2011)

<i>E. coli</i> <i>Saccharomyces cerevisiae</i> <i>Pantoea agglomerans</i> <i>Gluconobacter liquefaciens</i>	Mango & Melon (honeydew)	AC voltage	2.5 s, 5 s and 10s, at 12 to 16 kV	P. agglomerans and G. liquefaciens > 3 log reductions after 2.5 s. <i>E. coli</i> > 3 log reductions after 5 s. <i>S. cerevisiae</i> > 3 log reductions after 10s	Perni et al. (2008)
<i>E. coli</i> O157:H7	apple juice	corona discharges	40 s 100 Hz with 4000 pulses of 9000 V peak voltage	5 log CFU/g	Montenegro et al.
<i>E. coli</i> O157:H7 <i>Salmonella</i> Stanley	Red Apples	Gliding arc	3 min & 18kV	up to 3.7 Log reduction	Niemira and Sites (2008)
<i>Yeast/mouls</i>	Strawberris	DBD	5 min at 16 to 18kV	up to 3 Log reduction	N.N. Misra, et.al.,(2014)
<i>A. hydrophila</i>	Lettuce	COP	5 min at 20kV	5 Log reduction	I.K. Jahid, et.al.,(2014)
<i>S. typhimurium</i>	Tomatoes	DBD	300s at 18kV	3.8 Log reduction	D. Ziuzina, et.al.,(2014)

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#### 422 Associated benefits and concerns:

423 Cold plasma treatment to the foods is a promising technology in that which acts  
424 rapidly, does not leave toxic residuals on processed parts or in the exhaust gas and the  
425 temperature rise can be kept to an acceptable level. The viability of grains and legumes had  
426 shown to be preserved post plasma treatment with air and SF6 gases (Selcuk et al. 2008).  
427 Moreover, unlike pulse light and gamma radiation, the shadow effect is minimised  
428 considerably using gas plasma method as reactive species are produced in the whole chamber  
429 (Lassen et al. 2003; Goldman and Pruitt 1998). Contact angle (CA) measurements for  
430 nonthermal oxygen plasma treated lamb's lettuce have shown increased wettability of adaxial  
431 leaf surfaces after plasma exposure (Grzegorzewski et al. 2010a).

432 Further, in this case, a successive degradation of epicuticular waxes and cutin of the  
433 plant's epidermis was indicated using FTIR (ATR) and scanning electron microscopy (SEM).  
434 Above all, it can be conveniently operated in either batch or continuous mode. An aspect of  
435 the future of plasma technology is the possibility of pairing it with other decontamination  
436 processes such as pulsed-light treatment where synergistic effects may be more appreciable.

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437 Studies on the effect of nonthermal plasma on food components are scarce in the  
438 literature. Based on the experiments using low-pressure oxygen plasma it has been observed  
439 that time and structure-dependent degradation can be seen for differently selected model  
440 flavonoids adsorbed on solid surfaces, which was attributed to plasma-immanent reactive  
441 species such as O (3P), O<sub>2</sub> (1Δg and 1Σg+), O<sub>3</sub>, or OH radicals (Grzegorzewski et al.  
442 2010b). It has been observed in lamb's lettuce that pure compounds show a time-dependent  
443 degradation (flavonoids) or remain unchanged (phenolic acids) after exposure to oxygen  
444 plasma (Grzegorzewski et al. 2010a).

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445 Also, for the same model plant based food, a significant increase of protocatechuic  
446 acid, luteolin, and disomitin has been recorded after 120 s treatment time, independent of the  
447 applied plasma driving voltage. The effect of the UV and radical species of plasma on the  
448 lipids and other sensitive constituents of the foods such as vitamins C and E (which are  
449 naturally occurring in most fruits and vegetables and many foods) still remains ambiguous.  
450 Suitability of plasma technology for treatment of high fat/ lipid containing and other sensitive  
451 foods (where chemical changes may be induced) is doubted.

452 Products that have high lipid content would likely be affected by oxidation, resulting  
453 in formation of hydroxyl acids, keto acids, short-chain fatty acids and aldehydes etc. that  
454 cause off-flavours and odours. For these reasons meat products may not be ideal substrates  
455 for treatment with plasma (Critzler et al. 2007). For a full evaluation, additional issues  
456 concerning food quality must be considered and these include changes in nutrient content  
457 colour and textural qualities, toxic residues and other chemical changes (Vleugels et al.  
458 2005). Research efforts must be undertaken to evaluate the projected cost of the treatment for  
459 large quantities of food commodities and also the safety of gases used before direct plasma  
460 techniques will become common in the food industry (Basaran et al. 2008).

461 Therefore, cold plasma technology is an emerging disinfection method that offers an  
462 exciting complementary or alternative, novel non thermal approach for reducing the  
463 microbial populations on the raw or fresh produce surface and packaging materials. There  
464 may be several other applications in relation to food systems, which still remain unexplored.  
465 Various reactive species of plasma interact with the biological cells to cause permanent  
466 changes in them at cellular level and morphology, leading to inactivation.

467 Although cold plasma technology is not yet used commercially on a large scale, the  
468 equipment should be readily scalable. Systems for large scale cold plasma treatment of food

469 and related products using various energy sources and methods (like a multiplicity of  
470 microwave magnetrons) are already under development. This technology is increasingly  
471 finding acceptance among food processors for the surface sterilization and combating biofilm  
472 formation. The effect of cold plasma on the sensitive constituents of foods, mainly lipids,  
473 vitamins etc. have still some issues that need to be addressed and once this is achieved the  
474 technology will find wider applications and adaptation in food industries.

## 475 **Conclusion and Recommendation**

476 Cold plasma treatment proved to be a flexible, efficient, chemical-free antimicrobial  
477 process and it can represent an easy to use sanitizing method for the food industry that does  
478 not require particular temperature, humidity or pressure conditions. The application of plasma  
479 treatment on different commodities represents a relatively new decontamination approach of  
480 this technology, and more research studies are needed if it is to provide commercial  
481 applicability for the food industry.

482 Further development of cold plasma technology will have to be carried out, allowing a  
483 better understanding of the complex interactions during applications, such as food surface  
484 interactions, impact on food composition, optimization of gas composition and other  
485 processing parameters according to the treated sample. Also, additional information regarding  
486 food quality must be considered with respect to the cold plasma treatment, and changes  
487 concerning the nutrient content, toxic residues or textural qualities should be investigated.

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