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Review on cold plasma technology: The future of food preservation

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Abstract
Cold plasma technology is an emerging food processing treatment which devises wide range of applications especially in various food processing sectors. Cold plasma is known to have greater effects towards the microbial decontamination of various food products to ensure the food safety and shelf life to consumers. Cold plasma uses several reactive gaseous species which are likely less ionised for the inactivation of microbe's present in meats, poultry, fruits, and vegetables. This review paper will cover the concepts and underlying principles, applications in food, critical parameters, advantages and limitations when this technique is employed. Though, cold plasma technology holds inordinate ability towards decontamination of foods, the chemistry of reactive gaseous species with foods during cold plasma treatment, effects of CPT (Cold Plasma Technology) on environment and its economic impact has to be studied evidently through further research to gain its importance among consumer.

Keywords: Cold plasma, food safety, reactive gaseous species, food preservation, shelf life

Introduction
Food wastage is the major problem worldwide due to lack of farming practices and post-harvest processing. Higher production and growth with the lack of handling practices lead to the huge food losses. In which microbes and pathogens are the major constraint which causes food spoilage at altered times from farm to fork (Alexander *et al*., 2017) [5]. Increasing concern towards food safety among consumers, makes the food industries and regulation agencies to focus more on food quality factors which are responsible for spoilage. Hence, there is a vigorous efforts taken towards the food safety consideration and it's led to a solution for using many non-thermal preservation techniques in place of thermal food preservation techniques. Cold plasma technology is an emerging non-thermal food processing treatment which has wide range of applications in food sectors particularly in decontamination of foods (Afshari and Hosseini, 2014; Mahendran *et al*., 2017) [1, 2]. The term plasma is the Greek word (meaning "moldable substances) was first described by the chemist Irving Langmuir in 1920's (Rajvanshi, 2008) [3]. Over the years plasma found its usage in electronics and polymer industries for the surface modification of various polymers. However this technology attracted pervasive attention of many researchers and food industry practitioners for its application in food sector (Pankaj *et al*., 2014) [4]. CPT is found to be a powerful technique for the decontamination of food products including sporulating and pathogenic organisms (Niemira, 2012) [58].

Plasma is referred as the fourth state of matter as the properties of plasma is different from all states. Plasma is a very hot ionised gas which is made up of equal numbers of positively charged particles (protons) and negatively charged particles (electrons). When some external energy is applied to the atoms the high energy causes the electrons to strip away from the atomic nuclei and produces various reactive plasma products such as electrons, ions, neutrons, protons and reactive oxygen, atomic oxygen (O), ozone (O_3), hydroxyl radicals (OH^+) and nitrogen species (N_2, NO, NO_2, nitric oxide radical NO^+). The external energy sources can be electrical energy, magnetic current, radiofrequency waves, intense ultraviolet or laser light (Mishra *et al*., 2016) [6]. It is found that more than 99% of the visible matter in the universe is said to be in the plasma state. For an example, most of the substance present in stars, sun, beam emitting pulsars, interstellar clouds and supernovas are in the plasma state. The flash light during lightning occurs due to the electric current superheating of gases in the atmospheric state, creating a channel of ionised plasma. Manmade plasma applications are found in the production of fluorescent lights, neon lights, plasma television etc.

(https://www.livescience.com/54652-plasma.html, 2019; https://fstjournal.org/features/28-1/cold-plasma, 2019) [7, 8].

Plasma can exists in various states from extreme no equilibrium to almost complete thermal equilibrium state. Based on the method of generation, pressure and the relative temperature, plasma can be classified in to two different groups they are 1) Non-thermal plasma 2) Thermal plasma. Thermal plasma contains contain gas species and electrons having same temperature of around 10000°K under high pressure are thermodynamically equilibrium in nature. Hence, Thermal plasma has found its use in effectively treating the hazardous metal wastes (Pankaj *et al*., 2018; Mizuno 2009) [9, 10]. Non-thermal plasma are also called as cold plasma or non-equilibrium plasma is partially ionised gas which are produced under atmospheric/vacuum temperature of about 30-60°C. Cold plasma contains various gaseous species possess same energy of above moderate room temperature but the electron poses higher temperature of 20,000K with higher energy (Misra *et al*., 2011) [11]. Cold plasma could be possible method for food applications to inactivate microorganisms to keep the food safe throughout its shelf life. Fast growing demand for fresh produce poses the food industries to supply minimally processed food in a safe manner to the consumer. As a result, cold plasma technique can be a promising technique for preserving food by destroying microorganisms without affecting its quality (Mishra *et al*., 2016) [6].

2. Principle and mechanism of decontamination

Plasma sterilisation effect was first documented and patented in the year 1968 by Menashi. When the food surface contaminants are exposed to reactive species produced by plasma there will be an accumulation of electrostatic forces at place where the high energy flows. The energy flow further induce radical bombardment action and hence cell lysis occurs. The impact of radical bombardment causes the lesions on the surface makes the microbial cell impotent to repair quickly which results in cell destruction. This phenomenon is termed as "plasma etching". Plasma etching causes DNA and chemical bonds denaturation, thus produces an antimicrobial effect on the cell (Menashi, 1968) [12]. The mechanism and action of plasma on bacterial cell is given below in Figure 1.The efficiency of plasma treatment against contamination is based on the several factors mentioned below.

2.1. Factors affecting plasma decontamination

- Power level to generate the plasma
- Gaseous mixture and Intensity of gases species
- Length of exposure
- Flow rate, Pressure and design of the system
- Milleu factors - Relative humidity, pH and nature of sample (Ekezie *et al.* 2017; Cullen *et al*., 2018) [13, 14].

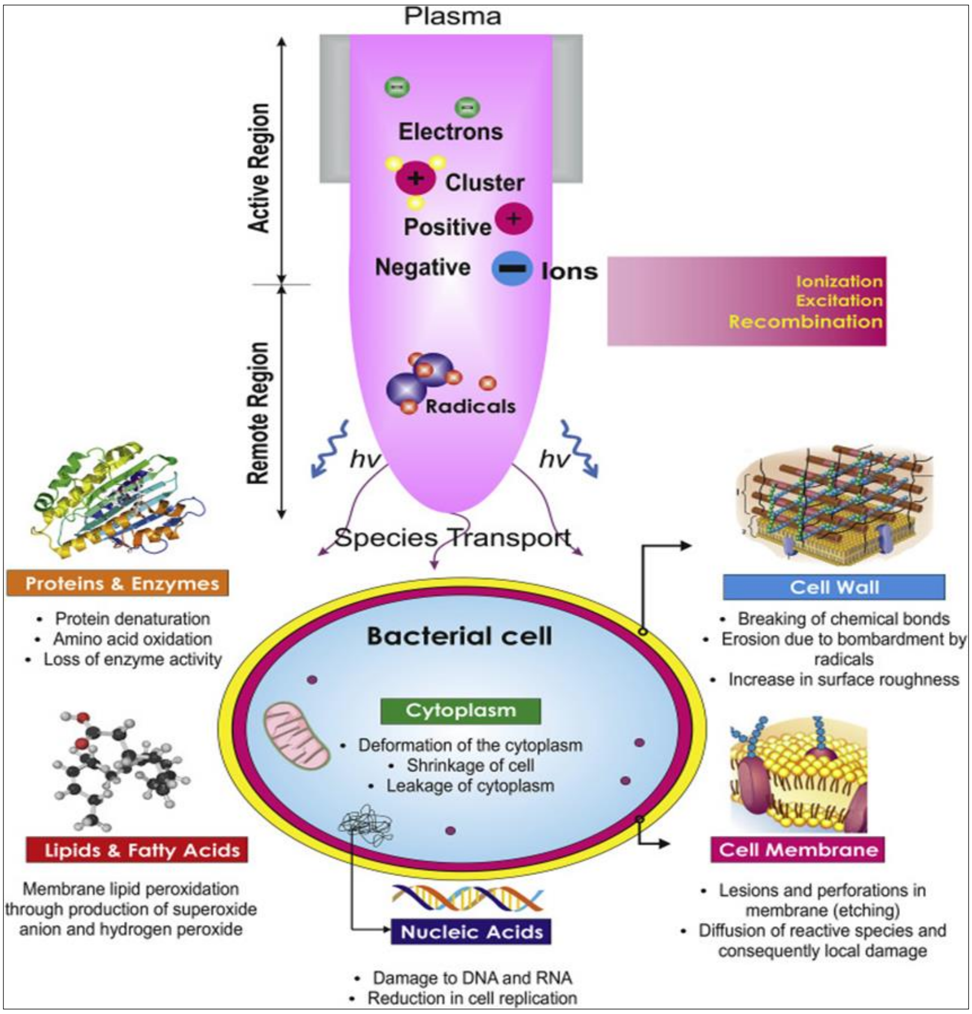

Fig 1: Action of Cold plasma on Bacterial cell (Misra and Jo, 2017) [31].

3. Methods of generation of Cold plasma

Plasma target chamber consist of simple gas such as air or nitrogen or the system with mixture of noble gases such as helium, argon and neon to attain plasma state. Ionised gas will be generated with the application of electric field or any external energy. For atmospheric cold plasma generation

DBD method, atmospheric plasma jet discharge, corona discharge and gliding arc discharge are commonly used as it requires mild conditions for processing operation (Niemira, 2012; Mehmood *et al*., 2018) [58, 15].

3.1 Dielectric barrier discharge
This method uses two flat metal electrodes which are covered with dielectric material. Neutral gas or any noble gaseous mixture moves between two electrodes in a closed target chamber and is ionized to generate plasma products as indicated in the Figure 2 (a). One electrode is connected to high voltage circuit and the other is grounded. The power consumption ranges between 10 and 100 W is used for its operation (Shimizu *et al*., 2018) [16].

3.2 Jet discharge
Plasma jet devices are made up of two concentric electrodes. The outer electrode is grounded and the inner electrode is connected to external energy source such as radio frequency source and creates RF (Radiofrequency) energy. Thus interacts with the working gas in the target chamber causes ionization and exits through nozzle and gives 'jet-like' appearance (Zhang, 2015) [17] as shown in Figure 2 (b).

3.3 Gliding Arc discharge
Gliding arc discharge follows periodic phenomenon that produces an auto-oscillating plasma species between two electrodes submerged in a laminar or turbulent flow. Plasma discharge starts from narrow end (termed as equilibrium stage), where the connecting electrodes of opposite polarity are joined together and it grows between lengths of the inter-electrode. The non-equilibrium phase starts when the arc exceeds its critical value. Plasma column undergoes heat loss when begins to exceeds the energy supplied by the power source. At that point, plasma rapidly cools and produces cold plasma (https://www.advancedplasmasolutions.com/what-is-plasma/plasma-discharges/,18; Khalili *et al*., 2018) [18, 19]. Figure 2 (c) represents the gliding arc discharge of plasma onto the almonds.

3.4 Corona Discharge plasma
In this process, plasma is produced by non-uniform electric field strength under atmospheric pressure. Corona discharge appears near sharp points and along thin wires and it is represented in Figure 2 (d) shown below. In highly non-uniform electric field, gases exceeds its breakdown strength and produces weakly ionised plasma with some luminosity. Corona discharges are best suited for food sterilization applications (https://www.advancedplasmasolutions.com/what-is-plasma/plasma-discharges/,18; Antao, 2009) [18, 20].

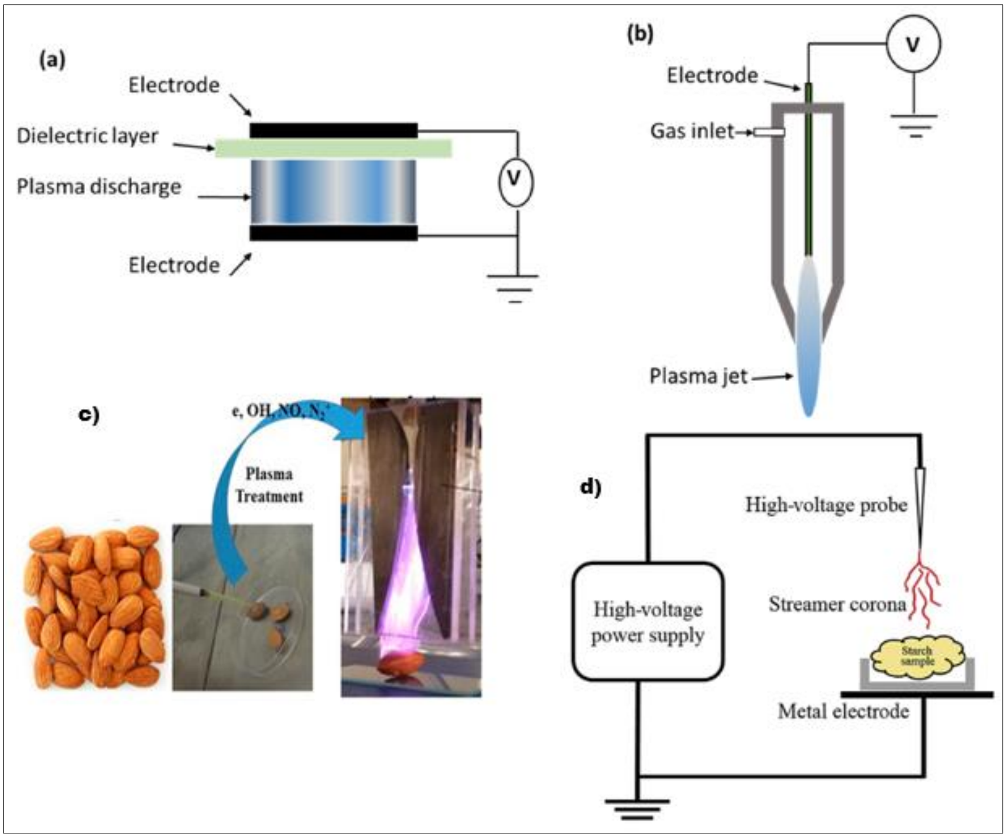

Fig 2: Methods of Discharge of plasma a) Dielectric discharge [59] b) Jet discharge [59] (Pankaj *et al*., 2017) c) Gliding arc discharge (Khalili *et al*., 2018) [19] d) Corona discharge (Wu *et al*., 2018) [24].

4. Exposure methods
There also different exposure methods studied by Cullen *et al* (2018) to deliver the plasma species to the defined target (Sarangapani *et al*., 2018) [21].

- Direct exposure
- Indirect or remote exposure
- Plasma-activated water

4.1 Direct exposure:
This method involves the direct exposure of plasma discharge onto the food surface by jet plasma or DBD method. It maximises the food interaction with the short-lived reactive gas species, UV (Ultraviolet rays) and electrons. However this method is not suitable for the sensitive food products which are complex in nature (Sarangapani *et al*., 2018; Okazaki *et al*., 2014) [21, 22].

4.2 Indirect exposure
In this approach, food products are placed in the target chamber which are some distance away from the plasma discharge. This method is suitable for sensitive food products which are fragile or contain susceptible tissues. By this method, one can achieve the uniform plasma discharge over certain produce (Sarangapani *et al*., 2018; Misra *et al*., 2014) [21, 23].

4.3 Plasma-activated water
In this technique water is activated for a period of time with several metastable species of cold plasma which results in the activation of relatively long-lived reactive species such as hydrogen peroxide, nitrates, and nitrites in the water. The resulted plasma water is then used for treatment of certain fresh produce by immersion, spraying or frozen as an active ice (Sarangapani *et al*., 2018; Wu *et al*., 2018) [21, 24].

5. Food applications of cold plasma treatment
Cold plasma technology shows promising dimensions for various sectors of food processing. It includes

5.1 Grain science and processing sector
Food grains and legumes were investigated for *Aspergillus spp.* and *Penicillum spp.* before and after treatment with plasma products showed significant log reduction after exposure for 15min (Selcuk *et al*, 2008) [25]. Depending upon the method of generation, treatment time and type of starch present in the food grains, cold plasma species are able to alter the starch properties. Cold plasma reactive species acts on food grains and causes surface modification, molecular degradation/granular etching or corrosion. Application of cold plasma was effectively studied on various food grain starches such as banana starch, Rice starch, zein, pea protein isolates, Brown rice and Basmati rice to improve its functional properties by surface and molecular modification of starch (Ezeh *et al*., 2018) [26]. CP helps in improving the swelling capacity, decreasing the cooking temperature, pasting viscosity, water solubility and water holding capability of food grains. When oxygen containing cold plasma are used for food rich in fats it may induce lipid oxidation and reduce the acceptability (Gavahian *et al*, 2018) [27]. Plasma when treated on banana starch at different voltages (30kV, 40kV, 50kV) for 3min time interval, it does not show any changes in the level of resistant starch and amylose content but increased the relative crystallinity and gelatinization temperature. Hence it was concluded that plasma could be a righteous tool to modify the characteristics of banana starch and other types of starches (Wu *et al*., 2018) [24]. Non-thermal fluidised bed plasma were used for the decontamination of maize grains, significant log reduction was found and well established (Dasan *et al*, 2016) [28]. Bacterial counts of *Bacillus cereus*, *Bacillus subtilis* and *E. Coli* were tested on brown rice using plasma. High antioxidant activity were observed on brown rice when treated with plasma and that could probably increase the nutritional value of the consumer (Chen *et al*., 2016) [29].

5.2 Food packaging
Originally cold plasma technology was employed for improving the surface modification and printability for packaging materials. Currently, CP treatments are utilised for the food packaging and biofilms treatments to enhance its antimicrobial and mechanical properties. It is proved that when plasma products acts on food packages it causes the surface functional group activation/addition or surface energies production to make positive impact on the various packaging properties include glazing, sealability, moisture/gas barrier property etc. It is considered to be reliable and cost effective technology (Rajvanshi, 2008; Miemira, 2012) [3, 5]. Recently cold plasma was utilised for in package processing to avoid the post packaging contamination. It is concluded that in- package treatment would be a possible method for industrial scale manufacturing (Misra *et al*., 2014) [23]. Indirect cold plasma treatment on RTE (Ready to Eat) meat inside PE (Polyethylene) bags were studied and stated that plasma reduced the counts of *Listeria innocua* (Rød *et al*., 2012)[30]. However, studies should be conducted to study the mechanism of action, since it involves thousands of chemical reactions when treated.

5.3 Meat and Egg processing sector
In meat sector, CP application was reported on beef, pork and chicken meat quality, microbial decontamination and shelf life extension. The result states that the cold plasma species are effective against *E. coli*, salmonella species, *L.monocytogenes*, yeast and mold species on meat surface (Rød *et al*., 2012; Misra and Jo, 2017; Lee *et al*.,2011) [30-32]. CP decreases the immobilisation of water in protein myofibrillar network and changes its functional properties of packed meat (Wang *et al*., 2016) [33]. CP technology application is found to have positive effects on surface decontamination of egg shell membrane against *S. enteritidis* and *S. typhimurium* microorganism (Ragni *et al*.,2010)[34]. Atmospheric plasma jets were checked on the surface of sliced ham and chicken meat. The result showed a significant log reduction of 6.52 when treated with nitrogen and oxygen mixture type (Song *et al*., 2009) [35]. Thus CP helps in achieving the better quality meat products in the market.

5.4 Dairy processing sector
Reports shown that the cold plasma has already been tested on various milk products include Whole milk, skim milk, UHT (Ultra High Temperature) milk and sliced cheese. The results of the study forecasted that cold plasma could be an alternative milk processing techniques because it is less likely affected the colour, pH, flavour and nutritional value of the milk products. It also inactivated contaminating microorganisms and alkaline phosphatase enzyme in few seconds (Song *et al*., 2009; Coutinho *et al*., 2018) [35, 36]. Study was conducted using DBD plasma for sterilisation of milk at the voltage of 3kV for 3min at 500Hz frequency, results showed that plasma is a very effective for killing the bacteria completely present in raw milk (Aslan,2016) [37]. In case of sliced cheese more than 8 log reduction were observed (Song *et al*., 2009) [35]. Therefore plasma application on milk and milk products could probably be an effective technique in enhancing their shelf life and keeping quality.

5.5 Fruits and vegetable processing sector (F&V)
Cold plasma treatment is said to be an ingenious technique since its replaces chlorine and water for decontamination of several fruits and vegetables. Cold plasma treatments on fruits and vegetable products includes berries, cherries, Apple, melon, Kiwi etc. were studied. Results proclaimed that CP treatments on the surface of F &V (Fruits & Vegetables) alters the pH and acidity of the food produce. This changes occurs when active species of plasma reacts with moisture on the surface. It is also found that the treated produce shows slight changes in texture (firmness) and colour during their storage period (https://fstjournal.org/features/28-1/cold-

plasma, 2019) [8]. Colour loss was observed on kiwi fruit and orange fruit juice when treated with plasma species (Kovačević *et al*., 2016) [38]. Studies were investigated on fresh and cut produce and blue berries and results demonstrated that CP treatments on F & V was effective against aerobic bacteria (Misra *et al*., 2014; Lacombe *et al*., 2015) [39, 40]. Similarly works on apple surfaces, melons and mangoes shows that there is a significant reduction in the counts of salmonella and *E. coli* after treating with plasma (Tappi *et al*.,2014; Tappi *et al*., 2016) [41, 42]. Therefore, microbial decontamination using plasma on fruits and vegetables is found to have a positive result with some negative impacts during its storage period.

5.6 Waste water (Effluent) treatment

Waste water disposal is now being a major issue faced by food industry since water coming out of food industry is with high concentration of organic loads. So far various thermal, chemical and filtration techniques are used for waste water treatments. ROS (Reactive Oxygen Species) of cold plasma have been reported to cause prompt changes in the degradation or decomposition of liquid waste. UV photons produced during CP treatment causes the pyrolytic effect by electrohydraulic cavitation in an indirect manner (Ekezie *et al*., 2017) [13]. Plasma jet exposure at 25kV for 150sec a were used for treating industrial waste coming out of tomato processing plant and for blackberry & beetroot waste water at the rate of 180 sec exposure showed the significant reduction in bacterial counts. Plasma also reduced *E. coli* counts and endotoxins compounds in the waste water up to 90.22% (Mohamed *et al*., 2016; Sarangapani *et al*.,2016) [43, 44]. Hence CP application is likely considered as potential technique for Industrial effluent treatment.

5.7 Agriculture sector

Agrochemical residues are identified to cause many human health disorder as they contains potentially toxic elements discharged during cultivation and processing. CP treatments are considered to be effective against the agrochemical residues like pesticides, insecticides etc., found on food materials which are applied to control crop infestation and weeds (Sarangapani *et al*., 2016; Bourke *et al*., 2018; Misra *et al*., 2014) [44-46]. In package treatment were done for pesticides residues present in water and strawberries with DBD plasma discharge for 5-8min. Results showed that the high dense pesticide compounds were degraded in to smaller chemical compounds possess less toxicity than parental compounds (Sarangapani *et al*., 2017; Misra *et al*.,2014) [47, 48]. Seed germination is depend on the various intrinsic (moisture content, permeability to gases and water, hardness) and extrinsic (drought, water logging, high salinity, high temperature) factors which adversely affect their performance of growth. Seed germination is enhanced when treated with plasma and found to increase water imbibition capacity of seeds and reduction in the microbial growth (Randeniya and De groot, 2015) [49].

5.8 Miscellaneous

Mycotoxins are the undesirable secondary metabolite produced by fungi which are thermally and chemically stable. Therefore many studies have been done reduce their occurrence in food and feed stuffs. Researchers mainly focused on various plasma application for the reduction of fungi and fungal producing toxins in foods. Jet plasma treatment on walnuts eliminated *Aspergillus flavus* when it is exposed with plasma products for 10min (Amini and Ghoranneviss, 2016) [50]. chemical compound Similarly, CP treatments on almonds, Hazelnuts, Black pepper and Red pepper were studied and stated that it is effective against mycotoxin production and surface contaminating microbes salmonella and Bacillus species (Deng *et al*.,2007; Hertwig *et al*.,2015) [51, 52].

6. Advantages

- Microbial inactivation efficiency can be achieved at low temperature.
- Suitable for treating sensitive raw and fresh food products.
- Requires less power input for operation.
- Doesn't alter or damage the key food nutrients.
- Reduces the risk caused by thermal and chemicals processing of food materials.
- Reduces water usage and solvent system for processing.
- CP don't alter the sensory and nutritional properties of food materials.
- Plasma is environmentally safe once the reactive species are withdrawn from the power supply
- Equipment cost is low when least cost noble gases are used for processing (Coutinho *et al*., 2018; Keener and Misra, 2016; Bartos *et al*., 2017; Dey *et al*., 2016; Hertwig *et al*., 2018) [36, 53-56].

7. Limitations

- Treatment of bulky and irregularly shaped food is difficult.
- Restricted volume and size of the food for treatment.
- Several ROS species has limited penetration into food products.
- It may affects the sensory and nutritional attributes of the food to some extent during processing.
- It may accelerate lipid oxidation and causes negative impact (Coutinho *et al*., 2018; Mandal *et al*., 2018; Niemira, 2012; Pankaj and Keener, 2017) [36, 57-59].

8. Future aspect

In the current days, applications of cold plasma technology is reported to be used for various food products decontamination. However CP treatment is not commercially used in the food industry, since the current research is majorly concerned on the plasma properties and processing on various food products. The economic trait of cold plasma and the mechanism of inactivation is still not clear since it produces 75 species and there are more than thousand chemical reaction happening at nano, micro, milli and seconds time scale (Pushpam *et al*.,,2018) [60]. The regulatory aspects of CP treatment should be covered to ensure the health of food supplies. The effect of cold plasma on nutritional and sensory attributes has to be addressed to commercialise this technology. Despite the research on the microbial decontamination if the above consequences are studied and cleared, CP could be a breakthrough technology for future food preservation (Jermann *et al*., 2015) [61].

9. Conclusion

Safe food requirement being the current demand by consumer, necessitates food scientists and researchers of the food industries to focus on enhancing the food quality and shelf stability through various novel technologies. Cold plasma technology is gaining interest among researchers and scientists because of its distinctiveness among other thermal and non-thermal treatments includes operation under low temperature for short time period and food quality integration. In that facet, cold plasma can be versatile technology with great potential to benefit the areas of food industry.

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