DFG Senate Commission
on Food Safety

SKLM

Opinion on the use of plasma processes for treatment of foods

Issued in German on May 25th 2012
English Version: December 14th 2012

Deutsche Forschungsgemeinschaft
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DFG

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The Commission would like to thank the members of the working group „Food technology and safety“: Prof. Dr. Dietrich Knorr (WG Chairman), Dr. Jörg Ehlbeck, Prof. Dr. Gerhard Eisenbrand, Prof. Dr. Karl-Heinz Engel, PD Dr. Christian Hertel, Dr. Thomas Holzhauser, Prof. Dr. Sabine Kulling, Dr. Oliver Schlüter for the preparation of the opinion and the SKLM staff represented by Dr. Michael Habermeyer and Dr. Angelika Roth for their support.

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The working group "Food technology and safety" of the DFG Senate Commission on Food Safety (SKLM) advises on new technologies concerning food processing. Treatment with plasma is a newly developed process which is currently used only on a pilot-scale in Europe. The novel plasma treatment technology is experimentally applied to consumer goods. There are also potential applications in the food sector, e.g. to inactivate microorganisms on food surfaces. There is still insufficient information on concomitant physical and chemical processes and changes induced in the food. On May 25th 2012, the SKLM issued a first statement on plasma treatment of foods in German. The English version was agreed on December 14th 2012.

Opinion on the use of plasma processes for treatment of foods

1 Introduction

Plasma processes are being used industrially, e.g. in medical engineering, material manufacturing, and illumination technique. In principle, plasma treatment may be used to decrease microbial contaminants at low temperatures, primarily on surfaces. The first laboratory-scale test series on the use of plasmas in the food sector is mainly studying possibilities of inactivating undesirable microorganisms on heat-sensitive foods because conventional thermal decontamination methods are more or less unsuitable for products such as fresh fruit and vegetables, meat, and eggs. Plasma treatment is also regarded as a potential alternative to other chemical (e.g. chlorine treatment) or physical methods (e.g. high-pressure, pulsed electric fields, ionizing irradiation). Advantages of plasma processes are: high efficiency at low temperatures (generally < 70 °C); precise generation of plasmas suitable for the intended use; just in time production of the acting agent; low impact on the internal product matrix; application free of water or solvents; no residues; resource-efficient. Owing to the cleaning and/or etching effects of plasma, which is already being exploited industrially, studies have so far concentrated on the possibility of controlled ablation of harmful substances, e.g. the removal of bacterial endotoxins from the surface of medical instruments. Possible adverse effects of plasma treatment on foods have rarely been investigated so far.

The aim of this SKLM opinion is to give an overview of the present state of knowledge and research needs for a safety assessment of plasma treatment of foods. Other applications of plasma to generate ultraviolet (UV) light or ozone are not taken into consideration in this opinion. Likewise, the application of plasma for specific surface modification is also not explicitly considered.

2 Definitions and terms

Terms and terminologies in plasma research and engineering are not always used consistently and depend on the specific objectives and the wide range of possible applications. The terms used in this opinion on plasma treatment of foods are therefore defined as follows:

Plasma is a gas containing free electrons, ions and neutral particles. The plasma state can be characterised, for example, by its thermodynamic properties using thermodynamic equations of state. A distinction is made between thermal and non-thermal plasmas. Thermal plasmas can be generated, e.g. by inductive coupling of high-frequency fields in the MHz range (ICP: inductively coupled plasma), by microwave coupling in the GHz range (plasma torch, e.g. PLexc) or by D.C. coupling (arc discharge). Non-thermal plasmas are used in low-pressure arc discharges, e.g. fluorescent lamps, in dielectric barrier discharges (DBD) e.g. ozone tubes, and in plasma jets. Just as diverse as the discharge devices are the possibilities of electronic control so that, together with pressure, gas flow and gas type a wide range of adjustable parameters is provided [1].

A **thermal plasma** is characterised by the existence of a thermodynamic equilibrium between the electrons, ions and neutral particles. The temperatures of a thermal plasma at atmospheric pressure generally are above 6000 K. This corresponds to a mean kinetic energy of less than 1 eV. Such a plasma can be indirectly applied to food, i.e. at a distance from the plasma source ensuring that the temperature remains within the desired range.

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. Contrary to thermal plasma non-thermal plasma can also be applied directly to thermally sensitive surfaces.

In food processing, the direct application of so-called "cold plasma" (see Tab. 1), as well as semi-direct or indirect treatment with thermal plasma is of interest as these can be used to treat the food at low temperatures (<70 °C).

Table 1: Overview of different types of cold plasma

Type	Description	Examples
Direct	Plasma is in direct contact with the substrate. Interaction based on irradiation (VUV, UV), charged molecules, radicals and reactive particles	• Plasma jet • DBD
Semi-direct	Distance between plasma and substrate is much larger than the mean free particle path. No interactions with charged particles. Antimicrobial effect due to irradiation, long-lived radicals as well as metastable and inhibitory substances	• SDBD with gap • Sterrad process with plasma-activated hydrogen peroxide
Indirect	Irradiation with UV and VUV light. Plasma is enclosed in a UV/VUV-transparent reactor. No interaction with plasma particles	• UV lamps
	Plasma is used to treat gas or liquids	• Ozone generator e.g. for drinking water treatment • PLexc-processed air (PPA)

For applications in the food sector, preference should be given to processes carried out at **atmospheric pressure** (e.g. plasma jet, dielectric barrier discharges) because they allow

continuous process control and do not accelerate undesirable phase transitions, compared to applications at **reduced pressure** (p < 1013 mbar) or **low pressure** (p < 10 mbar).

3 Processing principles and technical aspects

A non-thermal plasma is generated at atmospheric pressure by passing a process-gas (molecular or inert gas, e.g. air, nitrogen, argon, helium) through an electric field. Electrons arising from ionisation processes can be accelerated in this field so that they trigger impact-ionisation processes. If more free electrons are generated than are lost in the course of the process, a discharge can be built up. The degree of ionisation in technically used plasmas is usually very low, typically a few parts-per-thousand or less. The electrical conductivity generated via these free charge carriers is used to couple electric power. Free electrons colliding with gas atoms or molecules can transfer their energy, thus generating highly reactive species that can interact with the food surface. The electron energy is sufficient to dissociate covalent bonds in organic molecules. The dissociation energy required for single bonds is about 1.5 – 6.2 eV, about 4.4 – 7.4 eV for double bonds and 8.5 – 11.2 eV for triple bonds [2]. The dissociation energies of gases that can also be used as process gases are e.g. 5.7 eV (O_2) and 9.8 eV (N_2).

Light emitted in the plasma in the short-to-long ultraviolet range (100 – 380 nm) can induce photochemical reactions. Radicals, such as reactive oxygen species (ROS) formed in the presence of oxygen or reactive nitrogen species (RNS) formed in the presence of nitrogen, can lead to oxidation, cleavage or polymerization reactions. The UV radiation generally lies in the spectral range of natural sunlight interacting with the plant matrix. The density and dose of reactive species can be influenced via the method of generating the discharge. The electrons have an energy distribution function that depends on the excitation, gas type and pressure. This function is usually characterised just by its mean energy. However, for safety assessment, the high-energy fraction of the distribution function is also relevant. Due to the small number of high-energy electrons and the often small scattering cross-sections of reactions considered in this energy range, it is difficult to estimate the high-energy fraction of the distribution function.

4 Microbiological aspects

In medical engineering, both low-pressure and non-thermal atmospheric plasmas are used for sterilisation and decontamination of surfaces of heat-sensitive objects. Studies on lowering the microbial count using an atmospheric plasma have mainly been carried out on carrier materials such as glass, paper (filter paper) and plastics, such as polypropylene and polyethylene terephthalate. The results show a high potential to inhibit or inactivate microbes [3]. Likewise, an atmospheric plasma can also be used to lower the microbial count on

metallic surfaces, as has been demonstrated e.g. by first studies on the decontamination of tools used for processing meat [4].

During treatment with non-thermal plasmas, species such as hydroxyl radicals, hydrogen peroxide, ozone, singlet oxygen, superperoxide, nitrogen oxide as well as UV radiation act on the microorganisms [5-8]. These affect various macromolecules, such as DNA, proteins and lipopolysaccharides [9-14]. UV-induced DNA damage, photodesorption and radical etching have been described as mechanisms underlying the inactivation of microbes [15]. In the case of low-pressure plasmas, UV irradiation is regarded as the main factor for successful sterilisation [16, 17], whereas etching is regarded as the key inactivation mode for atmospheric plasmas [18-22]. (Lethal) damage occurs as a result of oxidation of cell components, accumulation of charged particles on the surface of the cells, lowering of the pH value with loss of pH regulation, breakdown of the membrane potential and energy generation [5, 7, 23-25]. If air is used as the process gas, the main radicals are $OH^{\bullet}$ and $NO^{\bullet}$, which can undergo ensuing reactions in aqueous media, thus significantly lowering the pH value (down to 3.5) [26-28]. When using atmospheric plasma within packaged foods, the microbial inactivation is attributed to ozone, respectively nitrogen oxides formed in the plasma [29-32]. Encapsulation of bacteria cells, a characteristic of many pathogenic bacteria, also affects the inactivation results. Such inactivation differences were observed for non-encapsulated (*E. coli* K12) and encapsulated (*E. coli* NCTG 11601) *Escherichia coli* cells [33]. Inactivation efficiency is also affected by the bacterial density on the surface being treated and the physiological state of the bacterial cells [22, 34, 35]. Flow cytometry analysis with *L. innocua* and *E. coli* allowed to differentiate between loss of bacterial cultivation potential and cell death after plasma treatment [36]. Bacterial biofilms are reported to be particularly resistant to treatment with an atmospheric plasma [37-39].

Of particular relevance for the food industry are endospore-forming bacteria, e.g. clostridia, as well as spores of *Bacillus* species, frequently used in plasma research. *Bacillus subtilis* spores have durable coat layers making them comparatively resistant to conventional sterilisation methods [40]. Experiments with UV filters confirmed that UV radiation from low-pressure plasmas plays a key role in the inactivation of *Bacillus* spores [16, 41, 42], whereas in the case of atmospheric plasmas, reactive particles in the plasma are responsible for spore inactivation [21, 43]. Evidence suggests that *Bacillus subtilis* spores are insufficiently inactivated by an atmospheric plasma with a low UV fraction. In this case, the gas composition [34, 44] and plasma treatment parameters [45] become key factors. Thus treatment of *Bacillus subtilis* spores with an atmospheric plasma of helium and oxygen for up to 60 minutes lowered the count by 2 log units, whereas treatment of vegetative cells of *E.coli* and *Staphylococcus aureus* under the same conditions lowered the count by 3 to 5 log units in a few minutes [34]. Another study with helium and nitrogen showed that the count of *Bacillus subtilis* spores was reduced by 1 to 2 log units within 180 seconds. A treatment time of up to 20 minutes is necessary to achieve an inactivation of 5 to 6 log units [46].

5 Effects on food

Food being treated with plasma is exposed to the reactive components in the same way as the contaminating microorganisms. Therefore, the aim is to achieve the highest possible reduction of the microbe count with the lowest possible effect on food quality. Investigations regarding changes of food-related substances have been carried out with isolated compounds. Substance losses were observed depending on the plasma system and exposure time [47-49]. The impact on the chemical composition of plant systems has only been studied with lamb's lettuce (*Valerianella locusta*) [50, 51]. After plasma treatment an increased flavonoid content was reported [51]. The reason behind this observed increase has not yet been elucidated. Scanning electron micrographs of plant surfaces treated with low-temperature plasmas revealed changes due to erosion phenomena in the upper epidermis. Plasma treatment of fresh spinach leaves and subsequent cold storage (24 h) caused discolourations [29, 30]. Possible sensory changes have been rarely investigated to date [52, 53].

The inactivation kinetics of microorganisms due to plasma treatment are also greatly influenced by the surface structure [22, 54-59] and thus strongly varies depending on the food surface [56, 59, 60]. Therefore, investigations using model systems cannot be simply transferred to the conditions prevailing on complex food surfaces. Studies regarding food decontamination using a non-thermal atmospheric plasma are compiled in Table 2.

Table 2: Applications of non-thermal atmospheric plasma to food matrices and selected experimental parameters

Type of food	Treated sample	Reduction of microbial count [log unit]	Microorganism tested	Plasma source / process gas	Reference
Fruit Vegetables	Spinach	up to 5.8	*E. coli O157:H7*	DBD, air, O_2	[30]
	Strawberry Cherry tomato	up to 4		DBD, air	[32]
	Apple	2.9 – 3.7	*E. coli O157:H7,* *Salmonella stanley*	Gliding arc dried, filtered air	[61]
	Apple Cantaloupe melon skin Iceberg lettuce	1 – 3.5	*E. coli O157:H7,* *Salmonella spp.,* *L. monocytogenes*	DBD	[62]
	Melon skin Mango skin	1 – 3	*E. coli,* *G. liquefaciens,* *P. agglomerans,* *S. cerevisiae*	Plasma jet, He+O_2	[58]
	Melon flesh Mango flesh	1 – 2.5	*E. coli,* *G. liquefaciens,* *L. monocytogenes,* *S. cerevisiae*	Plasma jet, He+O_2	[63]
	Sweet pepper	0.8 - 2	*Pantoea agglomerans*	DBD, He+O_2	[39]

Cereals Nuts	Almonds	1.8 - 5	*E. coli*	DBD, air (?)	[64]
	Hazelnuts Peanuts Pistachio nuts	-	*Aspergillus parasiticus*	Low-pressure plasma, air, SF_6	[53]
	Cereal grains Tomato seeds Legume seeds	-	*Aspergillus parasiticus, Penicillium MS1982*	Low-pressure plasma, air, SF_6	[52]
Fish Meat Eggs	Processed ham, sliced	0.2 – 1.7	*L. monocytogenes spp*	Needle/plate system, He	[59]
	Soft cheese, sliced	1 – 8			
	Chicken breast Ham	1.3 – 6.5	*L. monocytogenes*	Plasma jet, He, N, O_2	[65]
	Chicken breast Chicken leg	0.5 - 3	*Campylobacter jejuni* *Salmonella enterica*	DBD, air	[66]
	Cold-smoked salmon	1 – 5	*Lactobacillus sakei, Photobacterium phosphoreum*	DBD, Ar, CO_2	[67]
	Bacon	1 – 4.6	*E. coli,* *L. monocytogenes,* *Salmonella typhimurium*	Needle/plate system, He, He+O_2	[60]
	Egg (with shell)	up to 4.5	*Salmonella enteritidis,* *Salmonella typhimurium*	DBD, air (?) + H_2O	[68]
	Chicken meat Chicken skin	up to 3.5	*Listeria innocua*	System type? He, O_2	[56]
	"Ready-to-eat" Bresaola ham	0.4 – 1.6	*Listeria innocula*	DBD, O_2, Ar	[69]
Juices	Apple juice	up to 7	*E. coli O157:H7*	Needle/plate system, submersed	[70]
	Orange juice	5	*Staphylococcus aureus* *E. coli* *Candida albicans*	DBD, air	[71]

The efficiency of the method also depends on the specific properties of the product. Thus the specific energy input, heating of the product and temperature distribution are as important as material properties, composition, geometry and whether the material being treated is uniformly shaped, in pieces, powdered or a liquid. Pores, capillary openings, a high water content and the buffering capacity are influencing the inactivation efficiency of the plasma. The process temperature is a particularly suitable parameter for comparative assessment of plasma methods. Other parameters, e.g. electron energy distribution, plasma composition and the specific energy input may also be used; however, they have been difficult to determine so far. Selected relevant parameters are listed in Table 3.

Table 3: Technical characteristics and influencing parameters to describe plasma treatment.

Individual systems	Category	Example parameters
System	Plasma parameters	Type of plasma generation
		Geometry
		Voltage
		Current
		Pressure
		Gas mixture
	Applicator parameters	Chamber volume
		Treatment pressure
	Product parameters	Treatment area/volume
		Dosage
		Process temperature
Plasma	Radiation	Spectral power distribution
	Charged particles	Electron density
		Ion energy distribution
		Ion density
	Neutral particles	Type
		Density
		Lifetime
		Reactivity
		Temperature

Most studies carried out so far have used particulate food products. However, liquid foods, e.g. juices, can also be plasma-treated [70, 71]. Currently available data reveal that it is possible to achieve microbial count reductions in food of up to 6 log units, in some cases even up to 8 log units (see Table 2). However, general conclusions cannot be drawn from these individual observations. For example, *Listeria innocua*, a reference microbe for pathogenic *Listeria monocytogenes*, was reduced by 3 log units (4 min) on the surface of chicken meat, but only by 1 log unit on chicken skin, even after an exposure time of 8 min [56].

The use of plasma to decontaminate the surface of sensitive products, e.g. freshly cut foods is also being investigated [63, 69]. It is to be taken into consideration that bacterial cultures can also grow invasively into the food (e.g. through stoma of plant leaves) or migrate into food tissues so that plasma treatment may not reach them [58].

6 Allergenicity aspects

The technical processes involved in food treatment may affect the allergenicity of food constituents. In most cases of IgE-mediated allergy to food the allergenic activity of proteins is lowered or remains stable, whereas an increase is rarely observed [72-77]. This has not

yet been investigated for plasma treatment. If cells are not lethally damaged by the plasma treatment, it cannot be excluded that the plant's defence system may trigger formation of stress-induced secondary metabolites and induce pathogenesis-related proteins (PR) [78], some of which have a high allergenic potential. The plasma treatment process must thus be designed to avoid the formation of these substances as far as possible. However, there is currently no data available on the allergenicity of the resulting products. In view of the lack of substantiated scientific studies, it is not possible at present to make general statements on modulation of the allergenic potential by plasma treatment.

In addition to immediate type allergic reactions to proteins, particularly low molecular-weight constituents of food plants (e.g. essential oils from herbs and spices) may also elicit T-cell-mediated contact allergies (type IV allergies). Structural changes of such constituents induced by plasma treatment cannot be excluded and could theoretically change their potential to elicit type IV allergies. No data are available so far.

7 Safety aspects/evaluation criteria

Products or product groups treated with a plasma must be subjected to a case-by-case assessment. The plasma process must be described with respect to its technical parameters. This not only applies to the process itself (working gas, degree of ionisation, treatment geometry, exposure time, temperature, pH value, system layout, etc.). In addition, a profile as comprehensive as possible of the plasma-induced physical/chemical/biochemical/microbiological changes in the food is required. No investigations have been conducted so far on whether toxic compounds are formed as a result of plasma treatment. At present, the available information regarding the consequences of plasma treatment on various foods is insufficient for a safety assessment of the process. The impact on microbiological safety must also be taken into account in order to achieve an adequate health evaluation.

In the case that plasma treatment leads to significant changes and affects the nutritional value, constituent composition and/or content of undesirable substances in the food, the treated products must be considered within the scope of the Novel Food Regulation.

8 Summary and research needs

Plasma treatment opens new perspectives for lowering the microbial count on food surfaces. For a health assessment sufficiently substantiated microbiological data are still too scarce. This also applies to the impact of plasma treatment on potential compositional changes in the food, especially with regard to potentially harmful components. The assessment of plasma treatment is additionally impeded by the lack of standardisation and incomplete descriptions

of the process parameters. Furthermore, the application range, i.e. which foods are suitable for plasma treatment, has not yet been sufficiently elucidated.

Development of criteria to assess plasma treatment of foods requires not only a detailed, standardised characterisation of the process parameters and the method, but also elucidation and characterisation of potential changes to substances in the treated foods. This calls for a comprehensive profile of the plasma-induced physical/ chemical/ biochemical/ microbiological changes in the food, also taking into account the penetration depth. Furthermore, an assessment of the microbiological safety requires adequate studies on inactivation of food-relevant microorganisms on or in foods. To clarify the microbiological applicability range, food-relevant microbes as well as adequate reference microbes should be tested. These should include encapsulated bacteria (pathogenic enterobacteria), temperature-resistant spore-forming bacteria (*Geobacillus stearothermophilus*), acid-tolerant bacteria (*Lactobacillus acidophilus, Acetobacter* and *Gluconobacter* species) and irradiation-resistant bacteria (*Deinococcus radiodurans*).

The possible impact on the allergenicity of foods also requires investigation.

According to the present state of knowledge, plasma treated products have to be assessed case-by case.

Glossary

Etching: Etching involves the interaction between radicals (e.g. $OH^{\bullet}$ or $NO^{\bullet}$) and substrate molecules that results in detachment of the molecule from the substrate. Also named as "volatization".

DBD: Dielectric barrier discharge occurs between two electrodes, at least one of which is insulated (the dielectric barrier); first made popular by the ozone tube developed by Werner von Siemens; cold non-thermal plasma; widely used in industrial applications, e.g. to prepare plastic films for printing; operates at atmospheric pressure; excitation at 50 Hz to 20 kHz.

Plasma jets: Plasma jets are generally cold plasmas that are usually excited in the radio-frequency (rf) range (1-27 MHz) and are blown out of the capillary or tubular discharge unit by the gas stream (usually noble gases He or Ar).

Photodesorption: UV-induced decomposition of large molecules into smaller volatile components that are released from the surface.

Plasma torch: Plasma torches are generally operated at atmospheric pressure to produce a thermal plasma. They are usually excited by microwaves and are carried out of the discharge unit by a gas stream.

PLexc is a special type of self-igniting plasma torch developed by INP Greifswald.

PLexc-processed air (PPA): PLexc using air as the working gas.

SDBD: Surface dielectric barrier discharge. This is a DBD variant in which discharge takes place on the surface of the electrode unit. It does not require a counter-electrode.

SDBD with gap: Use of SDBD at a greater distance from the product being treated.

Sterrad process: Commercially available sterilisation process developed by Advanced Sterilization Products. It uses plasma-activated hydrogen peroxide to e.g. sterilise medical products.

Vacuum ultraviolet radiation (VUV): Ranges from 100 nm to 190 mm, depending on the definition; the ultraviolet range is from 190 nm to 380 nm.

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