Cleaning of the surfaces in the food industry using cold plasma

Reinigung von Oberflächen in der Lebensmittelindustrie mittels kaltem Plasma

V. Jovicic [1,2*], I. Jung [1], A. Zbogar-Rasic [1], M. J. Khan [1,2], A. Delgado [1,2]

[1] Institute of Fluid Mechanics (LSTM) at Friedrich-Alexander University (FAU), Erlangen, Germany
[2] Erlangen Graduate School in Advanced Optical Technologies (SAOT), Germany

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Abstract

According to the German Food Hygiene Regulation (LMHV), each food producer must comply with special rules and take precautions in order to preserve the welfare of consumers. This makes the cleaning, disinfection and sterilization of production installations and product distribution systems, one of the most challenging quality improvements potential [1].

Furthermore, according to the Bavarian State Office for the Environment [2], Annual amount of cleaning and washing chemicals discharged into the German wastewater system alone is reported to be around 194000 tons of surfactants, 32000 tons of phosphates, followed by several tons of perfumes and optical brighteners. These substances affect our ecosystem immensely. In order to partially replace them or to reduce their negative environmental impact, new plasma-enhanced processes for industrial cleaning are investigated [3].

This study quantifies the effects of air non-equilibrium (cold) plasma on the removal of oil, milk and egg stains from the test surfaces and determination of the sterilization effects related to this process. Contaminants were applied on a glass surface in a form of a thin film. Effects were quantified for different speeds of the plasma jet, distances from the plasma nozzle and different test times.

Visualization of the effects was done by two different imaging techniques: coloring of the substrates and by using optical setup based on the imaging of refracted light coming from a background ring-formed LED light source. Sterilization effects of atmospheric plasma treatment were tested with a complex nutrient medium for cultivation of the bacteria, which was inoculated with the E. coli bacteria incubated for 24 hours in a heating cabinet at 30°C. Analysis of the results were done using ImageJ and Halo software.

Introduction

Plasma is the ionized gas, a collection of ions, electrons and high energy species, capable to initiates chemical reactions. This was first discovered by a physicist Irving Langmuir, in year 1928 [4], [5]. On the basis of the ionization degree of the gas ranging from 100 % (fully ionized) to very low values (partially ionized), the term plasma is classified as thermal and non-thermal plasma. In thermal plasma, all the species present in the gas, e.g. the electrons and ions, have the same temperature ranging from 4000 K to 20000 K. On the other hand, the non-thermal (cold plasma) is produced using less power and the electron temperature is much higher than that of the bulk molecules present inside the gas [5-7]. The presence of these high energy species, capable of generating the chemical reaction is one way to introduce the advanced oxidation process (AOP), inside the process. Especially, the cold plasma is of much interest in food industry due to its lower temperature.

The German Food Hygiene Regulation (LMHV), regulates the production, storage, processing and preparation of all food products. According to LMHV and with the help of the Hazard Analysis of Critical Control Points (HACCP) concept, every company that produces or processes food goods has to define and document the food safety steps in order to preserve the welfare of consumers [8], [9]. Each year, food corporations worldwide suffer high costs in cases of product retrieval from the market and lost income in connection to the infected or contaminated food. This makes the cleaning, disinfection and sterilization of production installations and product distribution systems to one of the most challenging quality improvements potential in this field [1].

According to the German federal environmental office (Umwelt Bundesamt), cleaning and laundry agents being used in daily life in industrial and private level are contributing substantially to the chemical load of wastewater and therefore omnipresent. 1.3 million tons of cleaning agents and laundry detergents are sold to private end users. And only from private users, some 630000 tons of chemicals stemming from cleaning agents and detergents enter wastewater system annually, followed by several tons of perfumes and optical brighteners [10]. Many components of the wet cleaning chemicals are hardly or only partially degradable (phosphonates, EDTA, optical brighteners and several types of additives). These substances affect our ecosystem immensely. It is estimated that about 72% of wastewater pollution originates from the industry, closely followed by the agricultural sector and the households [11]. Plasma-enhanced processes for industrial cleaning are investigated to reduce their environmental impact.

The goal of this study is to investigate the static and dynamic surface cleaning and sterilization in food industry processes using non-thermal (cold) plasma. Olive oil, cow milk and egg were used as a contaminants applied on a glass surface in a form of a thin film.

Materials & Methods

Vacuum plasma chamber (static)

A specially designed glass chamber and an electrical unit is used for generating plasma, as shown in the Fig. *1*. Vacuum of 10^{-3} to 10^{-4} Pa is generated in glass cylinder volume of 0.29 m^3. Al and Cu electrodes are used in the plasma chamber with a 90 and 45 mm diameter and a height of 2 mm. An electric field of 1.5 kV/m is applied between the electrodes, through high frequency (HF)-high voltage (HV) generator. Cleaning of the food products containing olive oil, egg and milk were tested under the vacuum chamber plasma. Substrate was applied as a thin film over the

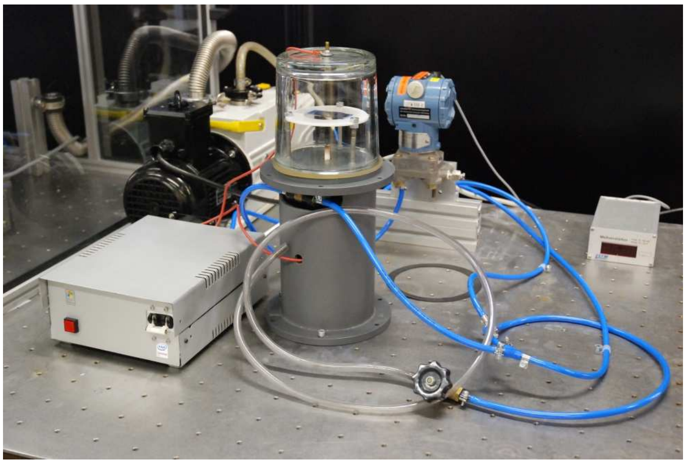

Fig. 1. *Vacuum plasma setup*

quartz glass. Initially, the experiment were performed using two disc Al electrodes with the size according to the chamber, for the treatment time of 10 minutes. The combination of rod and disc electrodes were also investigated with the increased exposure time up to 60 minutes.

Atmospheric plasma jet (dynamic system):

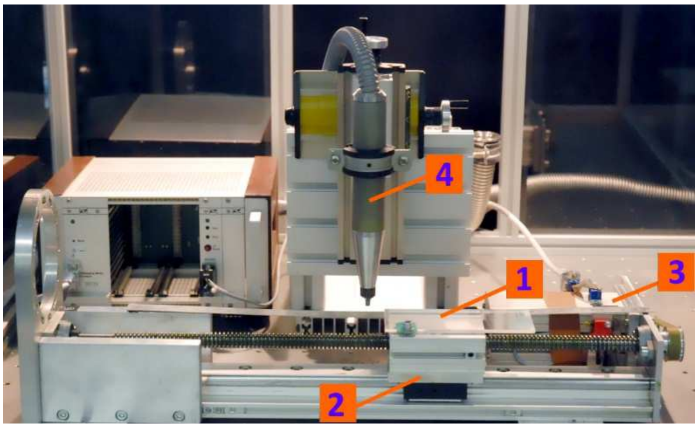

Fig. 2. *Atmospheric plasma setup*

At the substrate – gas interface, non-thermal electric discharge initiates chemical and physical processes. The most abundant active species in the atmospheric plasma are OH·, H·, O·, and NO· radicals. These species react with other plasma species, as well as with the targeted pollutants.

A 140mm·55mm·3mm quartz glass plate (Nr.1 in Fig. 2) caring oil film on its upper surface was used. During the tests, glass plate was fixed on a moving base (90mm·75mm) carrier (Nr. 2 in Fig. 2) moved by desired speed under the plasma nozzle (Nr. 4 in Fig. 2). Distance of the plasma nozzle outlet from the sample surface was adjustable with the precision of 0.1mm. Speed of the sample movement along the 370mm long axis was controlled by LabView© sequence, over PC controlled ISEL® step motor (Nr. 3 in Fig. 2). Diener® PlasmaBeam generator, operating at 10 kV and 20 kHz was used. Two types of oil were tested: olive oil Mazola® produced by Peter Kölln KGaA, Germany and sun flower oil, produced by Alnatura GmbH, Germany. Glass sample plate caring the oil film was than treated with the plasma jet at different speeds of glass movement and different heights of the plasma nozzle from the glass plate.

Visualization of the effects was done using two different imaging techniques. In the first case, oil sample was painted with blue oil colour produced by Daler Rowney-Georgian Oil, England in the mass ratio 4:1. In the second case, an optical setup based on the imaging of refracted light coming from a background ring-formed LED light source was used. The advantage of this method was the absence of additives in the tested oil film. In both cases images were taken using digital Nikon DX 80 camera with Sigma 180mm/f=2.8 EX-DG-OS Macro-Lens. Analysis of the test outcomes using ImageJ and Halo software gave similar results.

Sterilization effects of atmospheric plasma treatment were tested with a complex nutrient medium for cultivation of the bacteria, which consisted of 5 g/l yeast extract, 10 g/l of tryptone, 10 g/l sodium chloride and 1 g/l glucose, dissolved in 800 ml of distilled water, autoclaved at 121°C. The prepared medium was inoculated with the E. coli bacteria. In order to reach maximum growth, the bacteria was incubated for 24 hours in a heating cabinet at 30°C. After the cultivation, the application of the bacterial suspension was made on a slide with dimensions 76mm·26mm·1.5mm, previously cleaned with 70% ethanol and exposed to the Bunsen burner flame. Contaminated substrate was then applied on the whole surface of the sample plate and exposed to the plasma jet.

Results and Discussion

Low pressure plasma in the glass chamber was used to test the cleaning process of olive oil (A), milk (B) and egg (C) layers on the glass plate. The disc electrodes and the rod electrodes were used to observe the influence on the said substrates. Fig. *2*-5 show the results obtained using vacuum plasma.

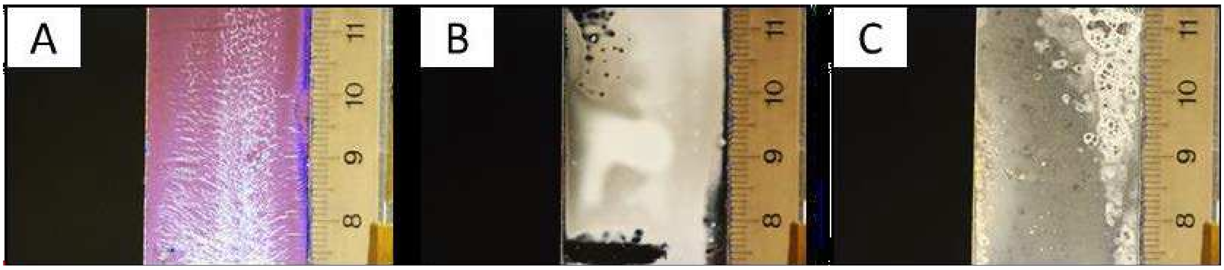

Fig. 2. *Results after 10 min. of treatment with Al-Al electrodes (A-olive oil, B-milk and C-eggs)*

Fig. 3. *Results after 10 min. of treatment with Al-Fe electrodes (A-olive oil, B-milk and C-eggs)*

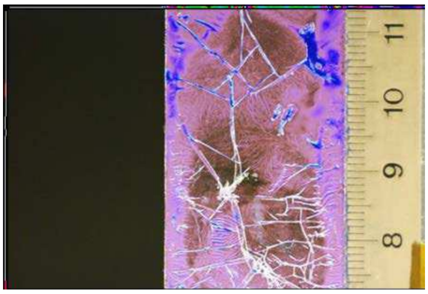

Fig. 4. *Results after 60 minutes of exposure of olive oil sample*

Different surface modifications can be observed in Fig. 3. and 4. for all three tested samples (A-olive oil, B-milk and C-eggs).

It can be concluded that selected vacuum plasma system is inefficient for the cleaning of above shown contaminated surfaces. Longer exposure times also do not result in an efficient removal of contaminants (Fig. 5)

In comparison to the static (vacuum) plasma system, the dynamic plasma system based on the atmospheric air plasma jet was tested using the same test samples as shown in the Fig. 3. and 4. For these experiments, plasma nozzle with diameter of 2 mm was used and the volume flow of plasma gas (air) was 7-10 ln/min. Speed of the plasma nozzle movement over the test surfaces was variated (from 1 up to 10 mm/s). In addition, influence of plasma nozzle outlet distance from the test surface was variated (up to 15 mm) and its effects were tested.

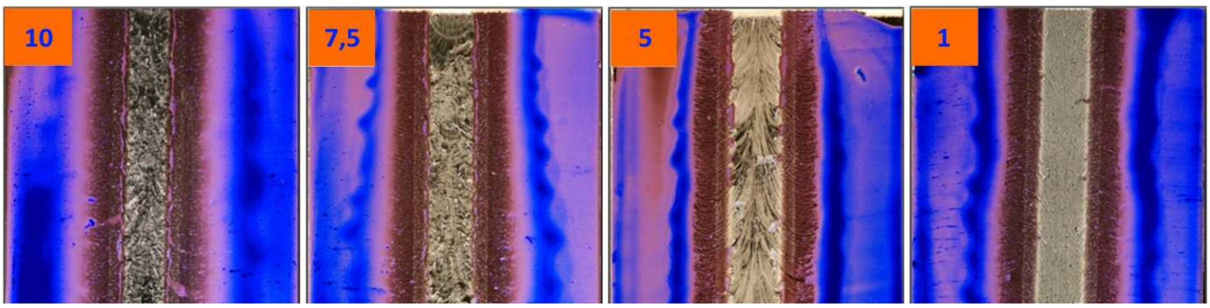

Fig. 6. *Test results of oil removal by different speeds of sample (1-10mm/s). Central (b&w) stripes are plasma jet traces, where oil was removed. Pink color region, between central stripes and outer (blue) layers, is the regions with partial oil removal and outer layer (blue color) is oily surface which was not in contact with plasma beam.*

It has been shown that increased speed and especially the increased height between nozzle and sample, results in decay of quality of surface cleaning (oil removal). Best results were obtained for low sample speeds of 1-2 mm/s and for the distance of the plasma nozzle from the oil film, in the range below 10 mm (Fig. 6.).

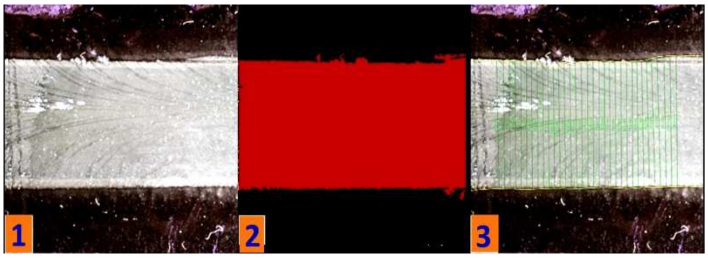

Fig. 7. *Sample treated with 5 mm distance between plasma nozzle and test plate and application velocity of 1 mm/s.*

In the Fig. 7. (1), a photo of a sample after the test is shown. White shade in this photo represent cleared surface and black shade is not removed oil film. In the Fig. 7. (2), the result of the ImageJ software is shown for the same sample. Based on the given threshold value, the software marks clean surface in a red color and the black area not cleaned surface. Fig. 7. (3) shows similar results of the Halo software used to measure how wide the cleaned stripes are.

For a plasma nozzle with outlet diameter of 2 mm positioned 10 mm above the test sample, average width of the cleaned surface (plasma trace) was app. 15.64 mm wide. In the case of the 5 mm distance between plasma nozzle and surface, average plasma trace was app. 15.30 mm.

The experiments were also performed to observe the sterilization of the surfaces. The results of the cold air plasma for the surfaces contaminated with E. coly bacteria are shown in the Fig. 8.

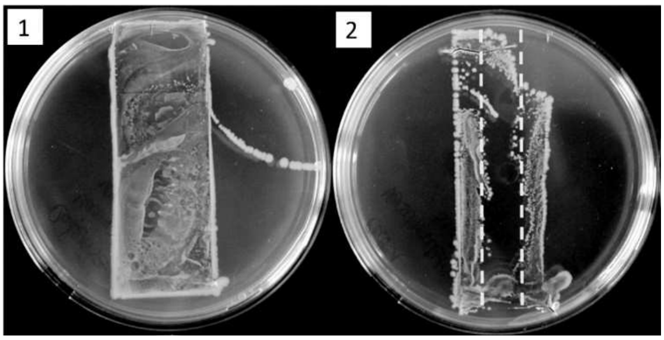

Fig. 8. *Test results of cold air plasma sterilization effects on the treated surface.*

Photo 1 (left) shows an imprint of the non-treated glass plate sample on an agar plate (Petri dish that contains a growth medium of E. coly).

Photo 2 (right) shows similar imprint of E. coli substrate after the cold air plasma jet treatment. Plasma traces are marked with dotted line.

As it can be noticed in Fig. 8. - (2), that exposure of cold air plasma beam, even for a very short time of 5 – 10 seconds results in practically total annihilation of all microorganisms present on the surface of the material or in applied substrate.

Conclusions

This paper provides the experimental study of the application of non–thermal plasma application in cleaning and sterilization of the surfaces, especially in the food industry. Two types of plasma mechanisms were investigated i.e. static and dynamic. The static plasma is produced in a vacuum chamber and dynamic, atmospheric plasma is produced in a form of a jet. Surfaces contaminated with olive oil, cow milk and eggs were investigated under both processes. The study shows that the static plasma is not beneficial in full cleaning of surfaces. However, certain modifications on the surface can be observed (e.g. bubbles on the surface of olive oil film) and these surface modifications are stronger as the exposure time increases.

The dynamic plasma consists of a cold air plasma jet by which the contaminated surface can be cleaned and sterilized very effectively. Study proves that the speed of the jet and the height of impact are the important factors influencing the cleaning and sterilization process. The best results can be obtained at a jet speed of 1 mm/s and 5 mm height of plasma jet.

Acknowledgements

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References

[1] **G. P. D. Wildbrett,** *Reinigung und Desinfektion in der Lebensmittelindustrie*. 2006.

[2] **B. Schaefer- Sümmermann,** “Wasch- und Reinigungsmittel,” *Fachkd. Warenkd. Drog.*, pp. 389–438, 2011.

[3] **R. Lauterbach**, “umweltbelastungen beim atmosphärischen plasmaspritzen,” 1992. [Online]. Available: http://www.lwt.mb.tu-dortmund.de/cms/de/Forschung/Ausstattung/TS/atmosphaerische_plasmaspritzanlage/index.html.

[4] “Plasma – The fourth state of matter.” [Online]. Available: http://www.plasmatreat.co.uk/plasma-technology/what-is-plasma.html.

[5] **C. H. Kruge,** “Partially Ionized Gases to and to.”

[6] **A. Bogaerts, E. Neyts, R. Gijbels, and J. van der Mullen,** “Gas discharge plasmas and their applications,” *Spectrochim. Acta, Part B*, vol. 57, no. 4, pp. 609–658, 2002.

[7] **B. Jiang, J. Zheng, S. Qiu, M. Wu, Q. Zhang, Z. Yan, and Q. Xue,** “Review on electrical discharge plasma technology for wastewater remediation,” *Chem. Eng. J.*, vol. 236, pp. 348–368, 2014.

[8] “German Food Hygiene Regulations.” [Online]. Available: http://www.bode-science-center.com/center/glossary/german-food-hygiene-regulations.html.

[9] “HACCP quality assurance.” [Online]. Available: http://www.bode-science-center.com/center/glossary/haccp.html.

[10] “Umwelt Bundesamt.” [Online]. Available: https://www.umweltbundesamt.de/en/topics/chemicals/cleaning-agents-laundry-detergents.

[11] Industrieverband Körperpflege und Waschmittel e.V., “Bericht Nachhaltigkeit in der Wasch-, Pflege- und Reinigungsmittelbranche in Deutschland,” pp. 1–52, 2012.