

Full Length Research Paper

Effect of hybrid gas-liquid electrical discharge on liquid foods (milk)

G.M.El-Aragi and Y. M. Abedel Rahman

Plasma Physics and Nuclear Fusion Department, Nuclear Research Center, AEA, PO 13759 Cairo, Egypt.

Accepted 5 March 2008

Pulsed high voltage discharges have been recently developed as sterilization method to replace the traditional thermal processes for the sterilization of liquid foods such as milk. The gas-liquid hybrid discharge (HD) reactor consists of plexiglass cylinder containing the liquid of high voltage electrode above liquid's surface and grounded electrode submerged in the liquid. The HD could produce both arc discharge in gas and liquid phases. The high energy plasma arc produces a pressure shock wave, which kills the targeted micro organisms by causing physical damage to their cellular matrix either by the sudden recoil of the cell or by the micro-eddies created on the internal cell structure.

Bacterio assay, has shown that the number survivor micro organisms after treatment by 13.5 msec pulse discharge $N = 0.388 \times 10^9$ CFU/ml versus the initial number of viable microorganisms in the control sample before treatment (N_0) = 1.456×10^9 CFU/ml which translates to log reduction of about 0.57. The applied electric field strength (of about 2.5 MV/m) has more pronounced effect on inactivation the micro organisms than the length of the treatment time. The decimal reduction time or time required for a 1-log cycle reduction in the microbial population is about 24 msec. Bacterol analysis, has shown that the *Proteus mirabilis* and *Enterobacteri cloacae* (enterobacteriaceae) that were appearing in the control sample before treatment, were disappeared from treated sample.

Key words: Pulse discharge, sterilization, hybrid discharge, thermal processes, shock wave.

INTRODUCTION

The application of a pulsed high voltage discharge in an air gas phase over de-ionized water changes the liquid in several ways. Electrical discharges in air produce ozone and small amounts of nitrogen oxides. In liquids, the ozone is converted into hydrogen peroxide while the nitrogen oxides are converted into nitric acid. The corona discharges strike the water surface and dissociate water molecules into hydroxyl radicals and hydrogen atoms. Hydroxyl radicals may recombine to form hydrogen peroxide. Hydrogen atoms react with dissolved oxygen to formulate hydroperoxyl radicals, which recombine to yield hydrogen peroxide and oxygen. Recombination of hydrogen atoms produces hydrogen molecules. Also, the metal of the anode tip may be sputtered due to the high electric field strength and formation of anode hot spots and hence elementary metal or metal oxides may be found in the liquid after discharge is completed. In general, the pulsed high voltage discharges in liquid

generate plasma arcs that initiate a variety of physical and chemical effects as high electric field, intense ultraviolet radiation, overpressure shock waves and, especially, formation of various reactive chemical species acting on biological cells and chemical compounds. These effects have various important roles in different application regions in the liquid and the magnitude of their contributions strongly depends upon the energy of the discharge (Lukes, 2001)

In pulse power discharges of several MW (Sun et al., 1998; Willberg et al., 1996; Lang et al., 1998) a large part of the energy is consumed in the formation of a high temperature (several tens of thousands of degrees Kelvin) plasma channel, which emits light in a wide range of wavelengths (Sun et al., 1998). This radiation can cause photolysis effect, leading to dissociation of liquid molecules and formation of hydroxyl radicals. In addition, the expansion of the plasma channel against the surrounding liquid generates an intense shock wave with pressure on the order of hundreds of Mpa (Shah et al., 1999). The resulting shockwave can indirectly induce pyrolytic and free radical reactions via electrohydraulic

*Corresponding author. E-mail: elaragi@gmail.com.

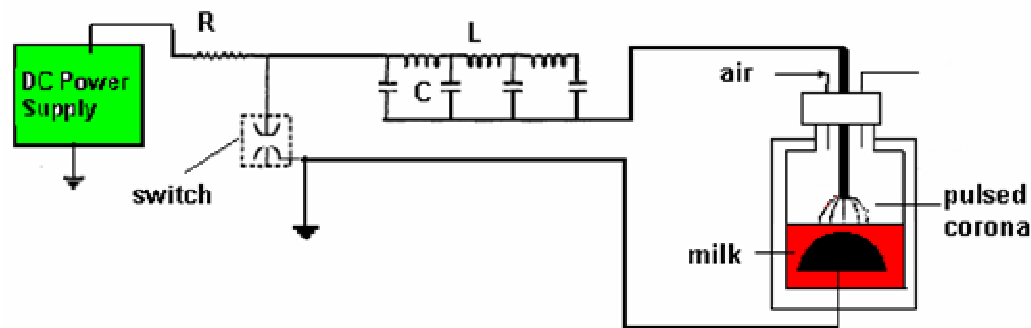


Figure 1. Experimental arrangement and circuit diagram of the hybrid gas-liquid electrical discharge system.

cavitation (Suslick et al., 1999). As the plasma channel cools, thermal energy is transferred to the surrounding liquid, resulting in the formation of a steam bubbles, in which the temperatures and pressures are high enough for the formation of transient supercritical liquid. Therefore, the oxidative degradation of organic compounds by such type of discharge is attributed mainly to the ultraviolet photolysis, electrohydraulic cavitation and supercritical liquid oxidation (Willberg et al., 1996).

Milk provides sufficient nutrients needed to support the growth of a host of microorganisms. Bacteria are capable of utilizing the proteins, fats, carbohydrates and vitamins in milk for their growth and metabolism. Different bacterial species may differ in regard to the enzymes they have or need to break down milk components.

New cold sterilizing techniques include sterilizing with pulsed electric field (PEF), ultrasonic and microwaves, high voltage, infrared and strong impulse of light in the visible wavelength. However, ultraviolet radiation is only effective in surface, and microwave can produce very high temperature not feasible for cold sterilization techniques. Pulsed electric field (PEF) can easily to be managed, and is now widely applied as a methods that we can be expected to be feasible at the industrial level (Ayhan et al., 2001).

The aim of this work is to examine the use of HD treatment system and the capability of pulsed plasma arc discharge directly in the liquid (milk), in killing microorganisms.

Experimental setup

The experimental apparatus is shown in Figure 1. The high-voltage electrode (discharging electrode) is 8 mm diameter copper rod and is connected to the pulse forming network "PFN" and to the power supply via a rotating spark gap switch. The ground electrode is submerged in the liquid, it is made of stainless steel and has a semi-spherical shape with a diameter of 4 cm. The distance between the tip of the discharging electrode and the liquid surface was set to 1.0 cm while that between the water surface and the submerged grounded electrode was set to 0.8 cm. The reactor nF capacitor charged through a 50 k ohm resistor by a negative dc high-voltage power supply and rotating spark gap switch. The

vessel has an inner diameter of 12 cm. The circuit consists of a 20 capacitor discharges when the voltage on the capacitor reaches the spark-over voltage of the spark gap switch, producing a narrow positive high-voltage pulse. The pulse generator provides a voltage pulse of 25 kV with about 8 ns rise time. The voltage is measured by a high voltage probe and the discharge current is monitored by a Rogowskii coil. The voltage and current waveforms are recorded using a digital storage oscilloscope (Lecroy Model 9300C)

The electrical circuit for production of high voltage pulses is based on discharging the capacitor into the liquid sample inside of the reaction chamber. The capacitor is negatively charged via a resistor and the negative side of the capacitor is grounded via the triggered rotating spark gap switch; hence, a positive pulse appears on the anode. The main advantage of a spark gap connected to the ground is that the trigger circuit can be kept at low voltage.

A pulse-forming network (PFN) accumulates electrical energy over a comparatively long time, and then releases the stored energy in the form of pulse. In practice a pulse-forming network PFN is charged by means of a high voltage power source, and then rapidly discharged into a load via a high voltage switch such as a spark gap or thyatron. A pulse-forming network (PFN) consists of a series of a high voltage energy storage capacitors and inductors. The combination of high voltage source, PFN, high voltage switch is sometimes called a power modulator or pulsar. A common form of a pulse-forming network (PFN) used in our experiment is the Guillemin type E-network, in which the capacitance and inductors are the same in each mesh and there is mutual inductance between adjacent coils. The rise time is determined by the rise time of the first mesh in the network, closest to the load. The value of a capacitor (C) and the inductance (L) of one a pulse-forming network (PFN) section are 5 nF and 4.5micro μ H respectively.

The experiment is conducted by injecting the sample (raw milk) into the treatment chamber, which is pre-cleaned and sterilized. The high voltage pulse generator parameters are adjusted to provide a 25 kV output voltage at 25 Hz discharge frequency for 300 s. After the treatment the sample is taken out of the chamber and placed in a sterile bottle. A control sample of raw milk is also placed in a sterile bottle.

RESULTS AND DISCUSSION

A Lecroy 200 MS/s 4-channel digital storage oscilloscope model (9300c) was used to recorded voltage and current waveforms, via a high voltage probe and a pulse current transformer, respectively; and to calculate the pulse energy and discharge power.

The measured peak value of the discharge current was

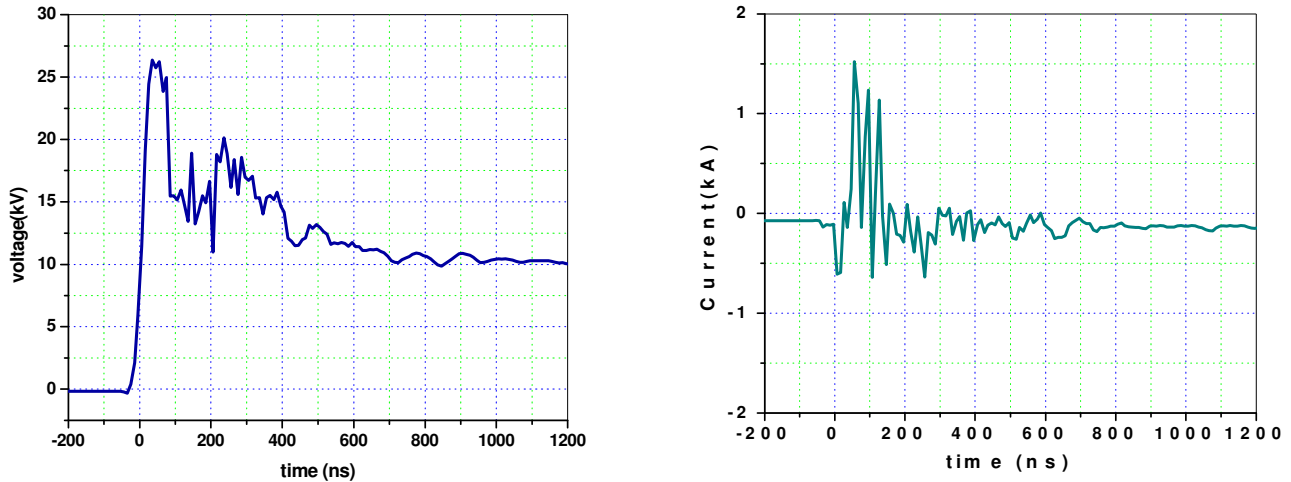


Figure 2. Typical discharge current and the voltage waveforms.

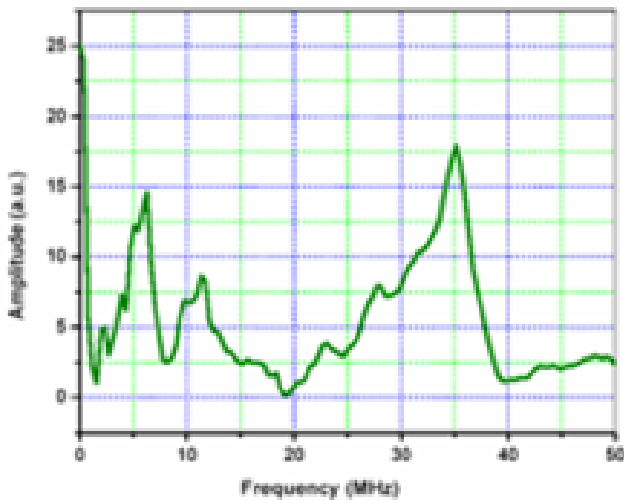


Figure 3. Fourier transformation of the discharge current pulse.

approximately 750 A during the pulse. Figure 2 shows the current and voltage waveforms measured as a function of time at an input energy of 9 J (maximum applied voltage 30 kV) at atmospheric conditions (air pressure of 1bar).

Fourier transform of the current pulse, as shown in Figure 3, shows the different frequency components in the pulse. It can be seen that most of the energy in the discharge current is between 100 kHz and 50 MHz. The spectrum have several peaks at different frequencies with the fundamental frequency component at 100 kHz and the harmonic frequency components at 6.25, 11.7 and 35.2 MHz. It is clear that the energy is spread over a wide frequency range, which is characteristic of short pulse discharges.

In an electrohydraulic discharge, cavitated bubbles are generated by the pulsed arc discharge and induce shock-

like pressure, which rises in the cavitation zone near the discharge electrodes. These cavitated bubbles are a major source of ultraviolet emission and radical species such as ozone, OH, H₂O₂, etc. Such chemical and physical reactions occur simultaneously in the cavitation zone and are expected to affect the treatment of the liquid (milk). Thus electrohydraulic discharge processes provide multiple mechanisms to achieve microbial inactivation.

The standard approach to describing changes in microbial population as a function of time uses the survivor curve equation:

$$\log [N / N_0] = -t / D$$

where:

N = microbial population at any time, t

N₀ = initial microbial population

D = decimal reduction time, or time required for a 1-log cycle reduction in the microbial population.

When microbial populations are exposed to pulsed electric fields (PEF), the applied electric field intensity should be above specific threshold determined by the critical electric field intensity for the target microorganism. A model similar to those for temperature and pressure can be used to describe the influence of electric field intensity on the rate of microbial population reduction. The proposed model would be (Peleg, 1995; Peleg and Cole, 1998; Anderson et al., 1996):

$$\log [D / D_R] = -(E - E_R) / Z$$

where:

D_R = decimal reduction time at a reference electric field intensity (E_R).

The electric field coefficient in this model is defined as: z(E) = the increase in electric field intensity (E) required

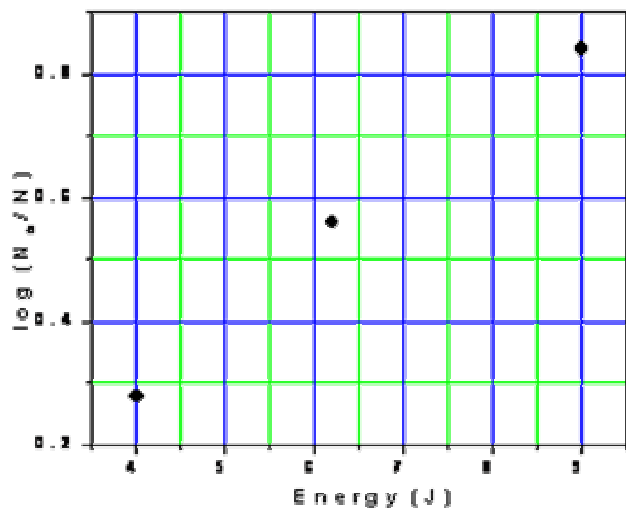


Figure 4. Rate of inactivation in log-reduction as a function of energy release (J)

to reduce the decimal reduction time (D) by 1-log cycle at a specific temperature and pressure.

A similar model has been proposed and used by Hülshager and others (1981) (Hülshager, 1981) and was applied by Jeyamkondan and others (1999) (Jeyamkondan et al., 1999), given by

$$\log [N / N_0] = \log [t / t_c] \{- (E - E_c) / K\}$$

where:

t = treatment time, which is the product of number of pulses and pulse width.

t_c = critical treatment time or treatment time below which no inactivation of microorganisms occurs

E_c = critical electric field strength or electric field strength below which no inactivation of target microorganism occurs.

K = specific rate constant (a coefficient with magnitude based on the slope of the survivor curve obtained at several levels of electric field intensity.) Larger magnitudes of the coefficient K would suggest a higher resistance to changes in electric field intensity.

The left hand side is the standard log-reduction and one log reduction refers to 90% reduction in the initial population of microorganisms. The electric field strength has more pronounced effect on inactivation of the microorganisms. The effect of electric discharges on the microorganisms (bacteria) in the raw milk is shown in Figure 4. Changes in the populations of bacteria (N) in the treated milk relative to the initial populations (N₀) as a function of discharge energy (J) released during the treatment. Raw milk with electrical conductivity < 10⁻¹² ohm⁻¹ cm⁻¹ was used, pulse repetition frequency less than 50 Hz and the initial concentration of bacteria was ≈ 1.5 × 10⁹ colony-forming units ml⁻¹. Pulsed high voltage discharge generates high electric field intensities

, which is considered superior to traditional heat treatment of foods because it avoids, or greatly reduces, the detrimental changes of the sensory and physical properties of foods (Quass, 1997). The large field intensities are achieved through discharging a capacitor into the reaction container. The energy at the reactor is generally converted into high E-field intensities, ultraviolet radiation, shock wave, and radical species such as O₃, H₂O₂, O● and OH● by intense spark discharge. The liquid will also experience temperature rise due to thermal effects from the arc, the joule heating is eminent and large fraction of energy will also be transferred to heat. The temperature of the liquid was measured using thermometer about 42°C

The strong arc discharge needs strong E-field and this field is proportional to the shock wave intensity. Therefore, these results mean that the shock wave is an effective parameter of the destruction of microorganisms in food. Discharge characteristic depends largely on the voltage capacitance and milk conductivity. During the discharge, it is noted that the phenomenon of sonoluminescence that is, emission of light by bubbles in a liquid (milk) excited by sound waves. Cavities are generated in a fluid (that is, bubbles) when the tensile phase (negative pressure) of the acoustic wave is sufficiently strong to rip the fluid apart.

The tensile phase of the shock wave is large enough to generate violent cavitation events. Shock waves of sufficient intensity (function of the discharge circuit, pulse duration, liquid conductivity, electrode size, gap distance and reactor configuration) will break the attractive forces in the existing molecules and create gas bubbles (Hart and Henglein, 1985; Shah et al., 1999).

These gas bubbles grow until they reach a critical size. Upon reaching the critical size, the gas bubbles implode or collapse. The implosion of microscopic gas bubbles of a liquid ruptures the cell membrane and collisions with other organisms and particular matter can also cause further mortality within the target area (Pétrier and Francony, 1997; Suslick et al., 1999).

Conclusion

The HD treatment was effective in the destruction of different types of bacteria. Enterococcus spp (Gram-positive) short chains or pairs of cells. "Fecal" streptococci (but are not coliform); common in fecal matter, but also in the dairy farm environment. Used as indicator organisms in some foods. Acid producers. Some strains have some heat resistance. Also Proteus mirabilis spp (Gram-negative) destroyed through generation of ultra-violet radiation, shock wave, ozone and free radicals.

ACKNOWLEDGEMENTS

I wish to express my thanks to Prof. Dr H. M. Soliman Head of Division of Material and Nuclear Manufacturing

for all her support and encouragement throughout this work and my thanks to all colleagues in the plasma department specially to Mr. T. Abdel Razeq for his efforts during this work.

REFERENCES

- Anderson WA, McClure PJ, Baird-Parker AC, Cole MB (1996). The application of a log-logistic model to describe the thermal inactivation of *Clostridium botulinum* at 213B at temperatures below 121-1 °C. *J. Appl. Bacteriol.* 80: 283-290.
- Hart EJ, Henglein A (1985). Free radical and free reactions in the sonolysis of aqueous iodide and formate solutions. *J. Phys. Chem.* 89 (20): 4342-4347
- Hülshager H, Pottel J, Niemann EG (1981). Killing of bacteria with electric pulses of high field strength. *Radiat. Environ. Biophys.* 20: 53-65.
- Jeyamkondan S, Jayas DS, Holley RA (1999). Pulsed electric field processing of foods. *Rev. J. Food Protect.* 62(9) : 1088-1096.
- Lang PS, Ching WK, Willberg DM, Hoffmann MR (1998). Oxidative degradation of 2,4,6-trinitrotoluene by ozone in an electrohydraulic discharge reactor. *Environ. Sci. Technol.* 32 (20): 3142-3148.
- Peleg M (1995). A model of microbial survival after exposure to pulse electric fields. *J. Sci. Food Agric.* 67(1): 93-99
- Peleg M, Cole MB (1998). Reinterpretation of Microbial Survival Curves. *Crit. Rev. Food Sci.* 38(5): 353-380
- Pétrier L (2001). Water treatment by pulsed streamer corona discharge, Ph.D. Thesis, Prague. pp. 9-14
- Pétrier CH, Francony (1997). Ultrasonic waste-water treatment: incidence of ultrasonic frequency on the rate of phenol and carbon tetrachloride degradation, *Ultrason. Sonochem.* 4 (4): 295-300
- Quass DW (1997). Pulsed electric field processing in the food industry. A status report on PEF. Palo Alto CA. Electric Power Research Institute. CR-109742.
- Shah YT, Pandit AB, Moholkar VS (1999). *Cavitation Reaction Engineering*. New York: Kluwer Academic Plenum Publishers.
- Sun B, Sato M, Harano A, Clements JS (1998). Non-uniform pulse discharge-induced radical production in distilled water. *J. Electrostatics* 43 (2): 115-126.
- Suslick KS, Didenko Y, Fang MM, Hyeon T, Kolbeck KJ, McNamara III W, Mdeleleni MM, Wong M (1999). Acoustic Cavitation and its Chemical Consequences. *Philosophical Transactions of the Royal Society of London.* 357: 335-353.
- Willberg DM, Lang PS, Hochemer RH, Kratel A, Hoffmann MR (1996). Degradation of 4-chlorophenol, 3,4-dichloroaniline and 2,4,6-trinitrotoluene in an electrohydraulic discharge reactor. *Environ. Sci. Technol.* 30(8): 2526-2534.
- Zehra Ayhan, Hye Won Yeam, Howard Zhang Q (2001) . Flavor, Color, and Vitamin C retention of Pulsed Electric Field processed orange juice in different packaging materials. *J. Agric. Food Chem.* (49): 669-674.