



## Enhancement of oil spreadability of biscuit surface by nonthermal barrier discharge plasma



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### ABSTRACT

The application of nonthermal dielectric barrier discharge (DBD) plasma for altering the hydrophilicity or hydrophobicity of polymer surfaces is well known. In this work, we demonstrate the potential of DBD plasma in enhancing the surface hydrophobicity of freshly baked biscuits, evident from the increased spread area of vegetable oil. The electrical and optical characteristics of the DBD plasma source have also been described. The spread area of individual oil drops has been measured using a digital image analysis technique. An exponential order of increase in the spread area has been observed at 80 kV with respect to treatment time, following DBD treatment. The findings reported in this work have applications in the industrial preparation of biscuit and cracker variants, where post-baking oil spray is desired. The effect of plasma treatments on the chemical constituents of biscuit requires further investigation.

**Industrial relevance:** There is a substantive evidence for the need of an alternate process which allows retaining functionality of sprayed oil in biscuits, with less oil or fat and increased spread. A novel process based on barrier discharge atmospheric pressure nonthermal plasma for enhancement of oil spreadability on dry, snack food products is developed. For a given volume of oil, up to 50% more spreading of oil can be achieved within few seconds of the process as compared to currently used methods. The plasma source can be easily integrated into a continuous biscuit and cracker production or finishing line.

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### 1. Introduction

Biscuits occupy a notable position among all cereal based bakery items and possess wide diversity in terms of their formulation, processing method and varieties. Biscuits, cookies and crackers are produced from wheat flour, sugar, fat and leavening agent, along with other minor ingredients. The ingredients are mixed together to obtain the dough, sheeted to optimal thickness, cut to desired shape, followed by baking. The process may have additional sub-steps depending on the type of biscuit and the choice of the process. It is a common practice in many biscuit production processes to apply the freshly baked biscuits with a spray of vegetable oil. The oil is applied either by spraying or by means of a device similar to an enrober in which the crackers pass through a flowing curtain of oil. In an old labour-intensive technique, the crackers were dumped while hot into wire baskets and dipped

into a tank of melted fat (Panda, 2003). Oil spraying is often accomplished using pressurised nozzles, spinning discs or electrostatic charge (Manley, 2001). The oil present on the surface improves the palatability and imparts an aesthetic appearance.

Oils used as surface coatings for savoury crackers are best if they have limited absorption into the cookie and remain as a glossy film. A thicker oil film on the biscuit surface leads to diffused reflectance and is undesirable. Furthermore, excess fat or oil on surface has a tendency to set and cause adhesion with touching biscuits when packed in stacks. This adhesion may result in damage to the thin surface of the biscuits as they are separated before eating (Manley, 2001). From a nutritional point of view, high-fat intake has been associated with various health disorders such as obesity, cancer, high blood cholesterol and coronary heart disease (Akoh, 1998; Sudha, Srivastava, Vetrmani, & Leelavathi, 2007). Therefore, there is a substantive evidence for the need of an alternate process, which allows retaining functionality of sprayed oil in biscuits, nevertheless with less oil or fat and increased spread.

The spreading characteristics of a liquid on a surface is dictated primarily by the surface energy, which is expressed as the hydrophilicity or hydrophobicity of the surface. This is a well-known phenomenon,

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often encountered in polymer and textile science, and surface engineering. Surface treatments are usually employed to improve the surface properties of polymeric films (Pankaj et al., 2014). The application of non-thermal plasmas to modify surface properties of polymers has already experienced a rapid growth (De Geyter, Morent, Leys, Gengembre, & Payen, 2007). The gas discharge plasma can be defined as a partially ionized gas containing neutral particles as well as an equivalent number of negative electrons and positive ions (Bárdos & Baránková, 2010). Plasma treatment only changes the uppermost atomic layers of a material surface without modifying the bulk properties (Poll, Schladitz, & Schreiter, 2001). Industrial plasma processing of food packaging films is already in practice (Ozdemir, Yurteri, & Sadikoglu, 1999). Non-thermal non-equilibrium atmospheric pressure plasmas are being actively researched due to their enormous potential for technological applications (Misra, Kadam, & Pankaj, 2011). Noteworthy, nonthermal plasma treatments have also gained importance in food science research as a means for microbial decontamination of foods and food processing surface, enzyme inactivation, and rapid pesticide dissipation (Misra, Pankaj, et al., 2014; Misra, Patil, et al., 2014; Misra, Tiwari, Raghavarao, & Cullen, 2011; Misra, Ziuzina, Cullen, & Keener, 2013).

The aforementioned concerns associated with biscuit production and analogies drawn from plasma and surface science form the motivation of present study. The specific aims of this study were as follows: (1) to characterise the electrical and optical features of the DBD plasma source used for treatment of biscuits via (current–voltage measurements and optical emission spectroscopy (OES), respectively), (2) to develop an image analysis method to quantify the spreading area of the oil, and (3) to determine if nonthermal barrier discharge plasma treatment increases the spread area of oil on biscuits.

## 2. Materials and methods

### 2.1. Materials

Wheat flour, bakery shortening, sugar and refined-bleached and deodorised (RBD) palm oil were all obtained from a local supermarket. Methyl Red-O dye was obtained from Sigma-Aldrich, Ireland.

### 2.2. Preparation of biscuits and oil spraying

The formulation used for biscuit making is presented in Table 1. Biscuits were prepared using the method of Wehrle, Gallagher, Neville, Keogh, and Arendt (1999), with modifications. Briefly, the ingredients were kneaded using a dough mixer (5KPM5 KitchenAid, St Joseph, Michigan, USA), followed by sheeting to a thickness of 2–3 millimetres and cutting out 7.5 cm diameter pieces. Baking was carried out for 10 min at 165 °C in baking oven (SCC101E Combi-Oven, Rational AG, Landsberg am Lech, Germany). The said baking conditions allowed the prevention of excessive cracking of the biscuit surfaces, which otherwise impose difficulties in feature extraction from images.

### 2.3. Plasma treatment

A schematic of the experimental set-up employed in the study is presented in Fig. 1. The DBD system comprises of two circular aluminium

plate electrodes (outer diameter = 158 mm) with a 10 mm thick Perspex dielectric in contact with the voltage electrode. The ground electrode was placed on an insulated Z-stage. A polypropylene (PP) box (material thickness = 1.2 mm) containing the biscuit sample is placed directly in the field between electrodes. The high-voltage step-up transformer (Phenix Technologies, Inc., USA) powered at 230 V, 50 Hz delivers a high-voltage output in the range 0–120 kV<sub>RMS</sub>. Two different voltages were applied across the electrodes for the experimentation viz. 80 and 90 kV (peak to peak) at 50 Hz. The rigid PP box had dimensions of 212 mm × 154 mm × 22 mm and also served as a dielectric material. All treatments were done with atmospheric air as the working gas. The atmospheric air condition at the time of packaging and treatment was 22% relative humidity (RH) and 20 °C, as measured using a humidity-temperature probe connected to a data logger (Testo 176 T2, Testo Ltd., UK). The biscuit samples were subjected to direct plasma field for 1, 3 and 5 min (in a randomised manner). Direct exposure refers to placement of biscuits within the area of coaxial electrodes.

### 2.4. Simulation of oil spray

An organic dye, Oil Red-O, was introduced into the oil at a level of 10 ppm, followed by centrifugation at 10,000 rpm for 10 min (Sanio MSE Mistral 3000ii, UK), to remove any suspended dye particles in the oil. The Oil Red-O is a lipophilic dye, which was added to improve contrast of the oil in images, whereby it allows accurate identification of the pixels. Oil droplets of 10 µL volume were drained on the surface of biscuits using a micropipette. Ten droplets were spotted on each biscuit (control as well as plasma treated). The biscuits were left undisturbed for 5 min to allow spreading and diffusion, following which image was acquired. All treatments were done in replicates and the experiments duplicated to give a total of 32 samples.

### 2.5. Electrical characteristics

The electrode bias voltage was monitored in the time domain using a high-voltage probe (North Star PVM-6) coupled to a 10:1 voltage divider to allow recording of the full voltage waveforms on an Agilent InfiniVision 2000 X-Series Oscilloscope (Agilent Technologies Inc., USA). A current transformer probe (Bergoz CT-E1.0S) was used to record the current waveforms. The waveforms were acquired in high-resolution mode (which performs in-situ boxcar averaging). The voltage was analysed both in the time and frequency domain, the later by means of fast Fourier transform (FFT) analysis.

### 2.6. Optical emission spectroscopy

Emission spectra of the discharge emissions within packages were acquired at 1.5 nm resolution with a computer controlled Stellarnet EPP 2000C-25 spectrometer, in which light from the plasma is coupled via an optical fibre. The diffraction grating in the spectrometer had a radius of curvature of 40 mm, 590 grooves per mm and an entrance slit width of 25 µm. The fibre had a numerical aperture of 0.22 with optimum performance in the ultraviolet and visible portion of the spectrum. The spectrometer operates in the wavelength range 190 nm to 850 nm. The spectra were evaluated qualitatively to identify the active species generated by the discharge.

### 2.7. Image analysis

Control and plasma-treated biscuits were photographed using an Olympus digital single-lens reflex (DSLR) camera (Olympus E5, Olympus, Melville, NY, USA), held on a tripod. The obtained digital images were then imported into R software (R 2.11.1, R Foundation for Statistical Computing, Vienna, Austria). Analysis of the digital images was conducted using existing routines within the EBImage package (Pau, Oles, Smith, Sklyar, & Huber, 2012) and in-house routines.

**Table 1**  
Formulation of ingredients used in the preparation of biscuits.

Ingredients	Formulation
Refined Wheat Flour	42.5%
Icing Sugar	21.3%
Ground Almonds	4.2%
Unsalted Butter	21.3%
Whole Egg	8.5%
Water	2.1%

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