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Effects of operating parameters on plasma-induced PET surface treatment

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Abstract

Plasma treatment is widely used to modify the surface properties of polymers, since it is a versatile, fast and environmentally benign technology. Different types of plasma sources can be utilized for surface modification, however, dielectric barrier discharges (DBDs) have received much attention due to their great flexibility. Therefore, in this paper, a DBD operating in air at medium pressure is used to modify the surface of a polyethylene terephthalate (PET) film. The influence of the main operating parameters (discharge power, gas flow, pressure and frequency) on the surface properties is studied in detail using contact angle measurements. Results show that the efficiency of the surface treatment increases with increasing discharge power, increasing pressure and decreasing gas flow. Results also show that the frequency of the discharge has no influence on the treatment efficiency.

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Keywords: Dielectric barrier discharge; Polyethylene terephthalate; Contact angle measurements; Operating parameters

1. Introduction

Plasma treatment is one of the most versatile techniques in surface modification. It has been widely used to improve the adhesion of coatings to metals and polymers [1], to increase the wettability and the printability of polymers [2], to enhance the biocompatibility of implants [3], ... Plasmas, often considered as the fourth state of matter, are composed of an ionized gas containing a mixture of ions, electrons, radicals and photons. When these energetic active species collide with a surface, some of the chemical bonds will break making the surface more reactive. Depending on the type of gas in which the plasma is generated, certain new functional groups can be grafted on the surface [4]. Therefore, plasma surface modification can induce a specific surface chemistry without altering the bulk properties of the material [5].

Dielectric barrier discharges (DBDs) or "silent" discharges are widely studied for the treatment of polymer films. A DBD is obtained between two electrodes, at least one of which is covered with a dielectric, when an AC high voltage is applied between the electrodes. The most interesting property of DBDs is that in most gases the breakdown starts at many points, followed by the development of independent current filaments (named microdischarges). These microdischarges are of nanosecond duration and are uniformly distributed over the dielectric surface [6].

In this paper, a DBD operating in air at medium pressure is used to alter the surface properties of a polyethylene terephthalate (PET) film. Despite the fact that DBDs are widely studied, there is currently little published work on the effects of varying operating parameters on the resulting surface treatment. Therefore, in this paper, the influence of the main operating parameters, i.e. treatment time, power, pressure, frequency and gas flow, on the surface properties is studied in detail using contact angle measurements.

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2. Experimental set-up

2.1. DBD set-up

The used dielectric barrier discharge system is schematically presented in Fig. 1. The DBD is created between two circular copper electrodes, 40 mm in diameter. Both electrodes are covered by a ceramic (Al_2O_3) plate with a thickness of 0.7 mm and an area of 50 mm × 50 mm. The air gap between the two ceramic plates is 2 mm. The upper electrode is connected to an AC high voltage source with a variable frequency (1–20 kHz), while the lower electrode is connected to earth through a resistor R or a capacitor C.

Before starting the experiments, a polyethylene terephthalate (PET) film is placed on the lower ceramic plate. After introduction of the sample into the reactor, the discharge chamber is pumped down to several kPa, while an air flow (Air Liquide – Alphagaz 1) flows between the electrodes. The gas flow rate is controlled by a mass-flow controller (MSK Instruments). The gas outlet is connected to a rotary vane pump and a needle valve is used in such a way that the pressure is kept constant during plasma treatment. The voltage applied to the electrodes is measured using a high voltage probe (Tektronix P6015A), whereas the discharge current is obtained by measuring the voltage over a 100 Ω resistor connected in series to ground. The current and the voltage waveforms are recorded using an oscilloscope (Tektronix TDS210 – 60 MHz). The current waveform shows numerous short peaks superimposed on the capacitive current: these peaks are an indication of the microdischarge activity and each peak corresponds to a series of microdischarges [6].

The resistor can be replaced by a capacitor of 10 nF; the voltage measured across this capacitor is then proportional to the charge stored on the electrodes. This latter measurement, together with the applied voltage, is widely used to obtain the voltage-versus-charge plot or so-called Lissajous figure [7]. The electrical energy consumed per voltage cycle U_{el} can be estimated by measuring the area of the Lissajous figure. The electric discharge power P_{el} can be obtained by multiplying U_{el} with the frequency of the feeding voltage [7].

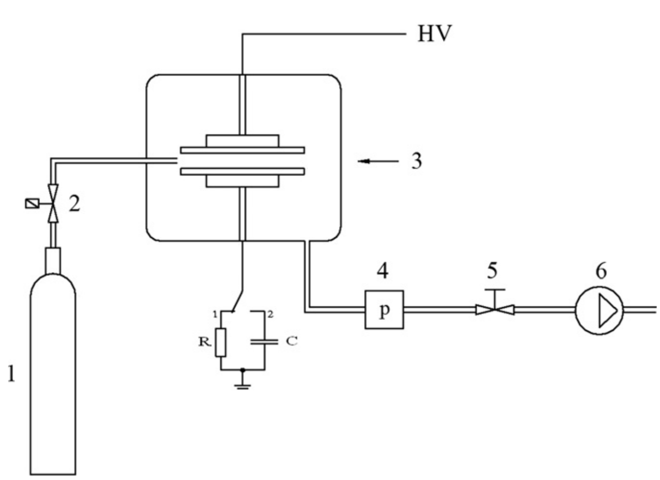

Fig. 1. Experimental set-up of the DBD discharge (1. gas cylinder, 2. mass-flow controller, 3. plasma chamber, 4. pressure gauge, 5. needle valve, 6. pump).

2.2. Contact angle measurements

After plasma treatment, contact angles are obtained using a commercial Krüss Easy Drop optical system (Krüss GmbH, Germany). This system is equipped with a software operated high-precision liquid dispenser to precisely control the drop size of the used liquid. The drop image is then stored, via a monochrome interline CCD video camera, using PC-based acquisition and data processing. Using the computer software provided with the instrument, measurement of the static contact angles is fully automated. In this work, 2 μl distilled water drops are placed on the PET films after plasma treatment. The values of the static contact angles, shown in this paper, are obtained using Laplace–Young curve fitting based on the imaged sessile water drop profile and are the average of eight measured values. The standard deviation on the average contact angles is smaller than 2%.

3. Results

3.1. Influence of treatment time and power

Fig. 2 depicts the evolution of the contact angle measured on the plasma treated PET films as a function of treatment time for different electrical discharge powers. The frequency of the DBD is kept constant at 10 kHz and the operating pressure is 5.0 kPa. The gas flow between the electrodes is set to 200 sccm (standard cubic centimetres per minute). The water contact angle of the untreated PET film is 74.8° and decreases to 33° for all discharge powers with increasing treatment time. However, when the PET film is treated during more than 1 s, the contact angle does not change anymore as a function of treatment time. This shows that there is a saturation of the plasma effect on the PET film. The large diminution of the contact angle after plasma treatment demonstrates the strongly increased wettability induced by the air DBD treatment even after very short treatment times. This behaviour can be attributed to the incorporation of oxygen-containing functionalities, resulting in a strong surface oxidation and therefore, an increased hydrophilicity of the polymer surface [8]. The molecular oxygen in the discharge is activated, ionized and dissociated resulting in the formation of extremely reactive oxygen species, which react immediately with the polymer surface [9]. As shown in a previous article, air plasma treatment of a PET film introduces C=O and O–C=O polar groups on the polymer surface [8].

Increasing the discharge power leads to a faster decrease in contact angle on the plasma treated PET films, as can be seen in Fig. 2. This can be explained by the fact that