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Investigation of the conditions for gas breakdown in a porous dielectric

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Plasma treatment of porous polymeric materials is a promising method for creating new materials that can be used in various applied problems, including medicine, in the development of new types of biocompatible and biodegradable polymeric materials. This work is devoted to the study of the processes of plasma treatment of porous polymeric materials depending on the size and type of pores in order to clarify the breakdown conditions and optimize the treatment process. A convenient semi-empirical model of the development of breakdown in a porous dielectric is proposed.

Keywords: plasma treatment of a dielectric, Paschen's law in a porous medium, breakdown voltage of a porous material.

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Plasma treatment of polymeric materials is a well-known process for modifying the properties of their surfaces (increasing the hydrophilic behavior and adhesion of dyes and adhesive materials). Treatment of not only the surface, but the entire volume of porous polymeric materials can have a much greater effect and can be used to create new materials that can be used in various applications, including the development of new types of biocompatible and biodegradable polymeric materials.

Plasma treatment of pores in polymeric materials involves plasma ignition inside these pores or, in other words, electrical breakdown of gas within a gap between electrodes, first described by Paschen [1]. The Paschen law is still being refining and modifying [2–5] for the conditions important from practical point of view, in particular for microgaps. The purpose of this work is to study the process of breakdown in a porous dielectric material, which can be considered as a breakdown of a group of successive microgaps when the Paschen's breakdown voltage is achieved in each of them.

The experiments were carried out in a discharge chamber with a gas inlet and outlet, consisting of lower and upper halves and a grounded metal bottom with a sealing ring of silicone rubber. A closed flask of glass with a flat bottom with a diameter of 20 mm filled with NaCl water solution with a conductivity of 20 mS/cm served as a high-voltage transparent electrode with a glass barrier. A sample to be treated was placed into the 2.5 mm thick gap between the high-voltage and grounded electrodes.

To study the breakdown process in the porous material placed into the discharge gap, the voltage and current oscillograms of the electrode system were recorded. Also, oscillograms at empty gap were recorded for comparison. The experiments were carried out with samples of porous polyurethane with completely interconnected pores and porous polystyrene with incompletely interconnected pores in the air at various pressures (p).

Fig. 1 shows an example of current and voltage oscillogram measured at an empty gap and at a sample of porous polyurethane with interconnected pores with an average size of 0.17 mm. Also, Fig. 1 shows calculated values of the capacitive current, i.e. the product of voltage derivative with respect to time and capacitance of the electrode system and lead wires (2.9 pF). The capacitive current allows us to calculate the discharge current (the difference between the measured current and the capacitive current) for more precise determination of the moment and voltage of the breakdown.

Also, the current and voltage oscillograms were used to calculate values of the flown charge and energy input as a function of time. The material with partly isolated pores demonstrated order of magnitude lower values of current, discharge and energy input, because actually only open part of the pores was involved in the breakdown where pressure was reduced down to the level of ambient pressure.

The obtained values of breakdown voltage were compared with the calculated values obtained for the assumption of identical pores with a size equal to the average pore size in the material as determined from microphotography. The calculation was based on the Paschen's formula adopted for barrier discharge in microgaps as described in [4,5]:

$$U = \frac{Bpd}{\ln(Apd) - \ln\left(\ln\left(1 + \frac{1}{\gamma}\right)\right)}$$
$$A = 14.73(\text{Torr} \cdot \text{cm})^{-1},$$
$$B = 450 \text{ V}/(\text{Torr} \cdot \text{cm})$$



Figure 1. Oscillograms of voltage U, measured current I and capacitive current I_c for the air gap (a) and gap filled with porous polyurethane with completely interrelated pores (b). Medium — air, pressure is 80 Torr, gap thickness is 0.25 cm.

with a variable value of γ that decreases with increase in *pd* in inverse proportion to its square:

$$\gamma = \frac{1.67 \cdot 10^{-3} (\text{Torr} \cdot \text{cm})^2}{(pd)^2}$$

The gap thickness (d) is taken equal to the size of pore and the total voltage is calculated as a product of the voltage at one pore and their quantity fitted into the gap. These curves of breakdown voltage in the discharge gap filled with porous dielectric material are shown in Fig. 2 for different sizes of pores.

The experimental data is based on the average pore size. Also, curves for the air gap without porous dielectric gap): medium — air, gap material are shown (cl. thickness is 0.25 cm. This simple approximation yields reasonably good match with experimental results despite the significant difference between the range under study pd and the range used for the initial derivation of the Paschen's formula [4,5], which is used in the calculation. For more detailed description of the breakdown process and better estimates of its parameters, a simple semiempirical model was developed that describes the breakdown in a porous dielectric material with Gaussian distribution of pore sizes with parameters selected close to those of the real distribution (obtained by means of microphotography). In the model, pores of different sizes form vertical columns or capacitors connected in series, with voltages calculated with consideration of their capacitance. If the Paschen's breakdown threshold [4,5] is exceeded at a pore, its voltage is zeroed and voltages at other pores are recalculated until the moment of breakdowns termination. As a result, the flown charge and energy input were calculated as function of the applied voltage. Results of the modelling for polystyrene (with an average pore diameter of 0.17 mm) and polyurethane (with an average pore diameter of 0.36 mm) are shown in Fig. 3.

The closure of part of pores in a real sample results in considerable drop of actual current and flown charge as compared with the calculated values, because the breakdown actually takes place only in the open part of pores, where the air pressure is lower. However, the calculated breakdown voltage corresponding to the value of significant flown charge (for example, 20% of maximum charge) is quite close to the experimentally measured value.



Figure 2. Comparison of the estimated dependence of breakdown voltage for the discharge gap filled with porous dielectric material with pore sizes of 0.12, 0.17, 0.36 mm and air discharge gap (*cl. gap*) on the product of pressure and gap thickness for various pore sizes with experimental data (exp).



Figure 3. Calculation of the energy input and flown charge as a function of voltage at the discharge gap filled with porous polystyrene (PS, average pore diameter is 0.17 mm) and porous polyurethane (PUR, average pore diameter is 0.36 mm). Medium — air, pressure is 80 Torr, gap thickness is 0.25 cm.

For the material with open pores, the model yields a good quantitative match of the flown charge $(6.6 \,\mu\text{C} - \text{calculation}, 7.5 \,\mu\text{C} - \text{experiment})$ and energy input $(8.0 \,\text{mJ} - \text{calculation}, 7.2 \,\text{mJ} - \text{experiment})$ at the moment when voltage has reached its maximum value of 9.2 kV.

Thus, the following conclusions can be made.

1. In this work experiments were carried out to determine the breakdown voltage in samples of polystyrene foam and porous polyurethane in pulse barrier discharge at different air pressures.

2. Electrical parameters of discharge in a porous material were studied depending on size and type of pores.

3. Estimate calculations of Paschen's curves for a gap filled with porous dielectric material for the assumption of identical pores were carried out. A good match with experimental results was obtained.

4. A convenient semiempirical model is proposed for breakdown development in a porous dielectric material with a pore size distribution close to the actual distribution. A good quantitative match with experiment is obtained for the breakdown voltage. For the material with interconnected pores, a good quantitative match with experiment is obtained for the flown charge and energy input values.

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Conflict of interest

The authors declare that they have no conflict of interest.

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