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Surface modifications of polystyrene and their stability: A comparison of DBD plasma deposition and direct fluorination



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ABSTRACT

Polystyrene (PS) as an industrialized insulating material is widely used in the pulsed power system, due to the good mechanical and electrical properties. However, the surface flashover of dielectric material limits the development of pulse power technology seriously. It is believed that the surface charge dynamics are related to the surface flashover closely. Therefore, in this paper, several surface modifications including plasma deposition assisted by dielectric barrier discharge (DBD) and direct fluorination (with/without O_2) are utilized to enhance the surface electrical properties of PS. The surface charge dynamics characteristics before and after surface modifications are investigated. The experimental results show the decays of surface charges for DBD plasma deposition and oxyfluorination become faster significantly. The surface potential of PS after DBD plasma deposition and oxyfluorination decreases to around 0 V over 100 s. The FTIR spectra indicates that a large amount of polar hydrophilic groups are introduced on the surface of PS samples by DBD plasma deposition and oxyfluorination to the dissipation of surface charges. The decay rate of surface charge after fluorination layer. Due to the rapid reduction of accumulated charges, the flashover voltage of all modified samples are increased by more than 30%. Furthermore, the stability of surface modification layer is compared. It is found that the stability of fluorination is better than oxyfluorination and DBD plasma deposition.

1. Introduction

As a kind of industrialized insulating material, polystyrene polymer (PS) has high buck resistivity and good mechanical properties. Thus, it is widely used as dielectric window and spacer in field of the pulsed power system and high voltage technology [1-3]. However, surface flashover at the vacuum/dielectric interface has limited the development of high voltage and high capacity of the power system [4-6]. According to the theory of secondary electron emission avalanche (SEEA), the surface flashover process is related to the surface charge dynamics closely [7,8]. Charging of polymer may owing to some external situations, such as corona discharge, defects near the triple junction, secondary electron multipactor etc. During charging, in spite of the surface charges decay simultaneously due to surface conduction or other decay processes, the surface potential induced by surface charge lasts several hours [9]. Moreover, the accumulated charges distort the electric field distribution so as to reduce the dielectric strength, even cause flashover [10,11].

Therefore, some investigators are devoted to improve the dynamics characteristics of surface charge by surface modification [12-14]. The methods of surface modification include direct fluorination [15], coating treatment [16], nanoparticles doping [17,18], electron beam irradiation [19] etc. Direct fluorination can induce a fluorination layer by a series of chemical reactions in the mixture gas including N₂, F₂ or O2. Du et al. [20] reported that fluorination of certain time could significantly improve the decay rate of the surface charge as well as the surface flashover voltage of polypropylene samples. An et al. [21] showed that a rapid potential decay on the surface fluorinated epoxy samples was due to high surface conductivity, and also the experiments indicated a significant influence of ambient humidity on the potential decay. Currently, surface modification assisted by low temperature plasma is usually applied for modifying the surface properties of polymers and polymer composites, due to the reasons including low consumption, flexible operation, high quality of the surface finish and the ecological-friendly [22-25]. Especially, atmospheric-pressure pulsed discharges including plasma jet and dielectric barrier discharge

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Fig. 1. The X-ray diffraction patterns of the untreated PS.

(DBD) are promising methods for surface modification [26]. Ref. [27] demonstrated that the voltage-resisting performance of epoxy resin sample was significantly improved by $SiC_xH_yO_z$ thin films deposited with atmospheric-pressure plasma jet (APPJ). Wang et al. reported that after the plasma deposition by atmospheric pressure dielectric barrier discharge the initial surface charge of epoxy resin decreased by 12% and the surface charge dissipation was accelerated, which were attributed to the lower density of shallow charge traps introduced by SiO_x film deposition [28].

Considering the engineering application, the high stability and reliability of modified surface is necessary. However, the aging characteristics and mechanisms of surface modification have not clarified yet. Therefore, in this paper, surface modifications methods including plasma modification and direct fluorination are used to improve the surface electrical properties of PS, respectively. The relationship between electrical insulation performance and physicochemical properties of surface are analyzed. In addition, the stability characteristics of modified surface are investigated.

2. Experimental details

2.1. Modification materials

The modification object material is commercially available polystyrene (PS). PS is a polymer synthesized by radical polymerization of styrene monomer, whose formula is $(C_8H_8)_n$. It is a kind of amorphous polymer with atactic tacticity owing to the benzene randomly oriented in the molecule chains. As shown in Fig. 1, the XRD analysis of untreated PS indicates that X-ray powder diffraction peak curves bread



Fig. 3. Surface potential decay characteristics for PS samples by different modifications.

with no diffraction peak, which means that the PS is in an amorphous state. The networks of PS are formed by molecule chain winding and the main chains are composited by saturated hydrocarbon. The molecular dipole moment of PS is 0.5 D approximately. Namely, PS has a good electrical insulation performance, especially in high frequency and high voltage condition. The experimental samples are cut into pieces with 50 mm \times 50 mm and the thickness is 2 mm.

2.2. DBD plasma deposition

The DBD plasma deposition system and experimental process are shown in Fig. 2. The DBD reactor is paralleled circular aluminum plates with diameter of 40 mm. The upper electrode is covered by K9 glass with thickness of 2 mm. The lower electrode is bare electrode and the sample are placed on the lower electrode. The discharge gap is set to 2 mm. The excitation source is a microsecond pulse generator (MPC-40D, Institute of Electrical Engineering, CAS) with a pulse rise time of 0.5 µs, a pulse width of 8 µs and a pulse repetition frequency (PRF) range 1 to 3000 Hz [28]. In this experiment, the PRF and voltage are set to 1500 Hz and 11 kV, respectively. The working gas is high purity Ar (99.999%). The tetraethylorthosilicate [TEOS, Si-(OC₂H₅)₄], as a precursor, is placed in a bubbling bottle and heated. Ar with 0.5 slm is pumped into the bubbling bottle. The mixture of Ar/TEOS with 2 slm pure Ar is blown into the DBD reactor through the lateral blowing device. The discharge voltage and current are measured by high voltage probe (Tektronix P6015A) and current probe (Person coil 6595), respectively. The voltage and current waveforms are recorded by oscilloscope (Tektronix DPO2024, bandwidth 200 MHz, sampling rate 1 G/



Fig. 2. The DBD deposition system and experimental process.

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