



Microwave transparent crosslinked polystyrene nanocomposites with enhanced high voltage resistance via 3D printing bulk polymerization method

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ABSTRACT

Demanded by the application in ultrahigh pulsed power generator as vacuum insulator, the MgO-filled nanocomposites with enhanced high voltage resistance were developed on the basis of microwave transparent crosslinked polystyrene (CPS). Considering the severe thermal runaway problem, a proprietary fabrication method combing the in-situ preparation with 3D printing bulk polymerization technique was also innovated and bulk products with different filler loading were successfully fabricated. The microscope images suggested that the distribution of MgO in the products was uniform, whereat the excellent mechanical properties of CPS were well maintained. Furthermore, owing to the appropriate dielectric characteristics of MgO and tiny filler loading, the relative permittivity of the composites increased very slightly while the loss tangent below 20 GHz was less than 8×10^{-4} , indicating the excellent transparency at the microwave band. Most importantly, the electrical breakdown strength was sharply improved from about 450 to over 600 kV/mm by only 0.1% MgO, as the barrier effect of electric double layer at the tremendous interface had effectively suppressed excessive charge injection and retarded electrical failure. Regarding the high voltage resistance obviously improved and the other properties well maintained, the CPS/MgO nanocomposites designed and developed in this work are applicable for the targeting application.

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1. Introduction

With the development in communication or microelectronic sectors towards higher and higher frequency to over GHz, microwave (100 MHz–100 GHz) transparent materials, especially the microwave transparent polymers characterized with very low dielectric loss tangent are increasingly demanded [1,2]. The commonly used microwave transparent polymers include polyethylene (PE), polytetrafluoroethylene (PTFE), polystyrene (PS) and so on. In the high power electrical industry, the low loss polymers at very high frequency are also demanded to play an indispensable role as the key components, such as the vacuum insulator stack in ultrahigh pulsed power generator that further gives rise to a diverse

set of properties and promotes present research project [3,4]. Considering the specific working conditions, excellent mechanical properties over a temperature range up to over 100 °C are fundamentally required. Herein, crosslinked polystyrene (CPS) prepared through copolymerization of styrene (St) and divinylbenzene (DVB) has been pertinently developed to make up the incompetence of common low loss polymers [5]. Furthermore, as a primary enabling technology for vast cutting-edge technologies in both civilian and military fields, the high pulsed power generators provide ultrahigh energy compressed in extremely short time and space. According to the circuit design and simulation, the voltage applied to the insulator could be over several megavolts (MV) [4,6]. An excessively reliable resistance to the high voltage is another fundamental requirement for the polymer insulator, which otherwise once breakdown at even a bare probability, may result in malfunction of the electrical facility and serious accidents. Therefore, much effort should be directed to further improve the high voltage resistance of CPS which ideally should be comparable to PE, the best polymer insulator with electrical breakdown strength over 550 kV/mm [7].

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Symbolized with the publication of “nanometric dielectrics” by John Lewis [8], hybrid with inorganic nanofillers was regarded as one of the most promising methods to improve the high voltage resistance of polymers. The vast amount of studies have come to an agreement that inclusions of nanometric scale is superior over the micrometric ones [9]. Considering the non-negligible influence of filler size and dispersion, a multi-core model was also proposed to highlight the significance of the tremendous interface [10]. However, the fact is that addition of different nanofillers always leads to diverse outcomes as revealed in the systematic investigation conducted by Yang [11]. Therefore, selection of proper nanofiller is one of the keys to the voltage resistance enhancement, because the dielectric characteristics of inorganic nanoparticles play a determining role. Pertinent research conducted by our group previously has found that the nanoparticles with a combination of high polarizability and excellent resistivity at ultra-high electrical field are preferred for enhancing the breakdown strength [12]. Furthermore, in order to minimize the impairment of the low dielectric loss feature that is fundamentally required by the targeting application, the loss factor of nanoparticles at microwave frequency should be lower or comparable with that of crosslinked polystyrene. In consideration of the above criteria, nano-MgO was selected as the filler in this research and proper fabrication process of the nanocomposites should be established correspondingly.

Compared with that of common methods (melt and solvent blending, etc.), the in-situ polymerization is particularly an attractive way, which can effectively incorporate nanofillers into varieties of polymers, especially the thermosetting polymers with uniform dispersion. In spite of this, preparation of CPS nanocomposites has been never reported so far, since the St-DVB bulk polymerization is highly exothermic in nature, which even conducted in lab scale, the enthalpy cannot be dissipated in time, resulting in rapidly accelerated reaction accompanied with uncontrollable temperature runaway. Pertinently in our previous study [5], an innovative technology referred to as 3D printing bulk polymerization has been developed. It is supposed to divide the large scale bulk polymerization into vast number of finite ones at different but indiscrete stages, so that a balance between heat generation and dissipation of the reacting system could be well maintained under proper technological parameters. Considering the respective advantages of in-situ polymerization and 3D printing bulk polymerization, a combination of the two methods will be a win-win breakthrough for the development of CPS/MgO nanocomposites.

However, there is quite enormous challenge that the pre-dispersed nanoparticles in monomers will inevitably precipitate or agglomerate since no stir is allowed during the polymerization process. Enabled by proper modification that gave stable colloidal suspension of nano-MgO in monomers, present work has strived to establish such a proprietary process, through which bulk products of microwave transparent CPS/MgO nanocomposites were prepared, followed by a systematic investigation about the effects of MgO on the microwave transparency and high voltage resistance in detail. To the best of the author's knowledge, this is the first study presenting such a processing technique and the development of CPS nanocomposites with enhanced high voltage resistance for targeting application in the ultrahigh pulsed power generator as vacuum insulator is groundbreaking.

2. Experiment

2.1. Materials

Styrene (St) was used as the starting monomer, while commercial divinylbenzene (DVB) consisted of 80% DVB (*p*- and *m*-

isomers) with the rest being mostly mono-vinyl isomer of ethyl styrene was used as crosslinking agent. They were all provided by Sigma Aldrich and purified through reduced-pressure distillation. In formulating the copolymerization, only DVB was regarded as crosslinker, while the others were assumed to react as styrene. The recrystallized benzoyl peroxide (BPO) was utilized as the free radical initiator.

The virgin magnesium oxide (MgO) of about 200 nm is commercially available and provided by Jingcai Chemical Ltd. (Shenzhen, China). In order to exclude the effects of ion contamination on the dielectric properties and electrical insulation of CPS, the as received MgO was washed with deionized water and dried in an oven at 250 °C for 12 h to get rid of the physically absorbed species and Mg(OH)₂.

The other analytical reagents or solvents, such as vinyltriethoxysilane (VTEO), ethanol, ammonia solution (28%) and toluene etc. were purchased from Sigma Aldrich and used without any further purification.

2.2. Preparation of CPS/MgO nanocomposites

The CPS/MgO nanocomposites were in-situ prepared through 3D printing bulk polymerization. In order to minimize the sedimentation tendency and realize better dispersion in this stir-free polymerization process, surface grafting of MgO with PS was applied. The detailed procedures were schematically shown in Fig.1.

2.2.1. Introduction of reactive vinyl groups on the MgO surface

To obtain the polymer-grafted inorganic nanoparticle with higher efficiency, it is preferred to initiate the grafting polymerization from the groups on the particles surface. Hence reactive vinyl group was firstly introduced onto the MgO nanoparticles through reaction of VTEO with the hydroxyl groups on the surface. 1.0 g MgO nanoparticles were dispersed in 200 ml ethanol through ultra-sonication for 30 min and the suspension was transferred to a 500 ml flask equipped with a reflux condenser. Then 5 g VTEO and 5 ml ammonia solution which have been previously dissolved in 50 ml ethanol were successively added into the suspension. The mixture was refluxed at the boiling temperature of ethanol over 24 h of stirring. After that, the suspension was centrifuged at 10000 rpm for 10 min. The precipitate was extracted with ethanol for 24 h to remove the excessive and physically absorbed VTEO on MgO. Finally, the treated MgO was dried in vacuum oven at 50 °C for 12 h and the obtained product was denoted as v-MgO. The content of the introduced double bonds was measured according to the method stated in Ref. [13].

2.2.2. Surface grafting of MgO with PS

The grafting polymerization of styrene onto MgO was conducted adopting the sonochemistry approach so as to avoid particles agglomeration during the reaction. All the experiments were carried out in a glass flask equipped with a water jacket to keep the mixture constantly at 70 °C and purged with argon gas during the whole process. Typically, 1.0 g v-MgO was homogeneously dispersed in toluene through ultra-sonication for 30 min. Then styrene with BPO pre-dissolved was added in one batch for polymerization. The concentration of styrene and BPO were 0.5 mol/L and 2.5 mmol/L, respectively. After different periods, the reacting mass was quenched by liquid nitrogen to stop the polymerization and submitted to centrifugation. Then the precipitate was washed with solvent, followed by extraction for 48 h to isolate the PS-grafted MgO from the absorbed polymers. Finally, the grafted nanoparticles were dried under vacuum at 50 °C for 12 h. The obtained product was denoted as g-MgO.

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