IMPACT OF SiO_2 NANOPARTICLES ON THE PROPERTIES OF SYNTACTIC FOAM

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Abstract: This study deals with the experimental investigation on syntactic foam, a composite material consisting of hollow glass microspheres embedded in an epoxy resin matrix. SiO_2 nanoparticles are added to the syntactic foam in order to improve the material's manufacturing process by decreasing the mixing viscosity of the uncured liquid material. First, the mixing viscosity is determined with a rotary viscosimeter. A minimum viscosity is observed at 2.5 wt.-% filling degree of the nanoparticles. Measurements of the breakdown field strength under ac and dc stress as well as measurements of loss factor, permittivity and resistivity are used to characterize the electrical properties. Specimens with sphere-sphere geometry are electrically stressed in a step test until breakdown occurs. The breakdown field strength of the nanofilled syntactic foam shows no significant difference to this one of syntactic foam without nanofiller. The permittivity, loss factor and resistivity are investigated with a protective ring configuration. The permittivity and the loss factor are slightly increased by the use of nanofillers, whereas the resistivity is not influenced by them. As main result of this study it can be ascertained that the manufacturing process of syntactic foam can be improved by the addition of SiO_2 nanofillers without an essential negative influences on the electrical properties.

1 INTRODUCTION

In general, syntactic foam is a composite material consisting of hollow microspheres embedded in a polymer matrix. In this study, epoxy resin is used as matrix material filled with glass microspheres of a wall thickness of 1 µm and mean diameter of about 100 µm. The name syntactic foam results from the microscopic structure of the material which is generated by the casting of hollow microspheres into the polymer matrix and strongly resembles a foam structure. Figure 1 shows a Scanning Electron Microscopy (SEM) picture of the material. Due to its foam structure, the density of this insulation material is much lower compared to conventional polymers or insulation oil and can therefore be used as a lightweight insulation material for high voltage applications.

Previous investigations show, that syntactic foam features good electrical properties for ac and dc applications [1-4]. As a result of inserting microspheres in pure epoxy resin during manufacturing process, the viscosity of the liquid material increases. This results in a maximum filling degree, which is currently reached at 55 vol.-%. At higher filling degrees the quality of the material reduces due to the high viscosity. Trapped air cannot escape out of the material during degassing process. To improve the quality of the material at filling degrees above 55 vol.-%, the viscosity can be reduced by adding nanoscaled particles - so called nanofillers - to the composite.

Ongoing investigations at the Institute for High Voltage Technology at RWTH Aachen University deal with the impact of SiO_2 nanofillers on the properties of syntactic foam. The focus of the present study lays on the experimental investigation on the optimum filling degree of the nanofiller and the resulting electrical properties of syntactic foam when nanofillers are added to the material.

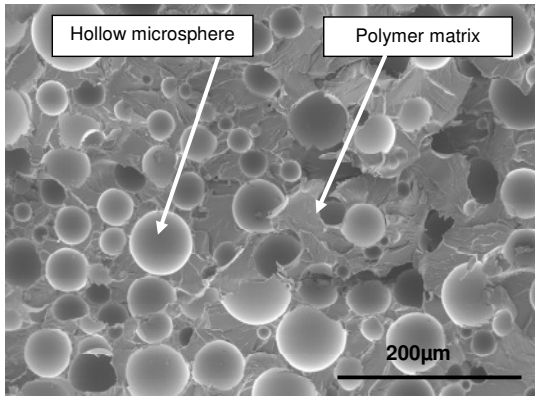

Figure 1: SEM-picture of the material structure of syntactic foam

2 THEORETICAL BASICS

2.1 Definition of Nanofiller

In the last twenty years several studies have been carried out on the behaviour of nanocomposites. In contrary to conventional composites, where the

filler has dimensions on the order of micrometers or more, nanocomposites contains “nanofiller” for which at least one dimension of the dispersed particles is lower than 100 nm [5-6]. It is well-known, that the addition of just a few weight percentages of nanofiller can have significant impact on electrical, mechanical, chemical and physical properties of polymers [6-8].

3 MATERIAL CHARACTERIZATION

The samples investigated in the present study are made of nanofilled and conventional syntactic foam. The base material for both types is a hot curing casting epoxy resin system. The base material is filled with 40 vol.-% hollow microspheres of borosilicate glass. The mean diameter of the microspheres is 45 µm. The nanofilled syntactic foam contains in addition to the microspheres 2.5 wt.-% of spheric SiO_2 nanofillers which have a diameter of 20 nm.

The base material and the fillers are mixed at a temperature of 60 °C. The correct order of the mixing process is shown in Figure 2. The mixture is degassed and then casted into a metallic mold. For the curing process with duration of 12 h the temperature is increased to 80 °C.

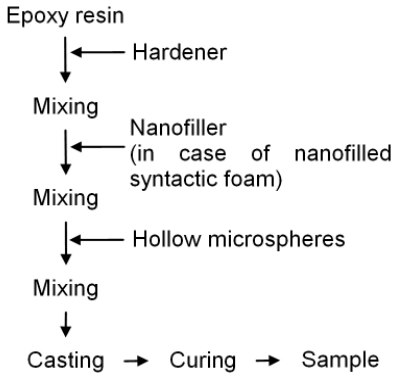

Figure 2: Preparation scheme for conventional and nanofilled syntactic foam

4 EXPERIMENTS

4.1 Mixing Viscosity

The mixing viscosity of the uncured, liquid materials is determined with a rotary viscosimeter at a temperature of 60 °C, which is the temperature at mixing process. The material is situated between a rotating cone and a stationary plate as shown in Figure 3.

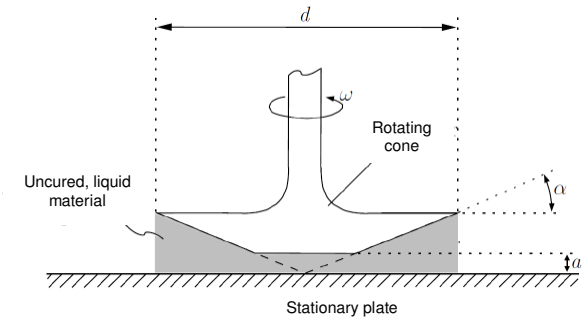

Figure 3: Rotary viscosimeter setup

4.2 Breakdown Field Strength

The electrical ac and dc breakdown field strength is determined according to IEC 60243-1 [9] and IEC 60243-2 [10], respectively. During the test the voltage is increased stepwise until breakdown occurs. The voltage starts at 20 kV, which is 40 % of the expected breakdown voltage, and is increased every 20 s by 1 kV. Figure 4 and Figure 5 show the measurement setup for ac and dc test, respectively.

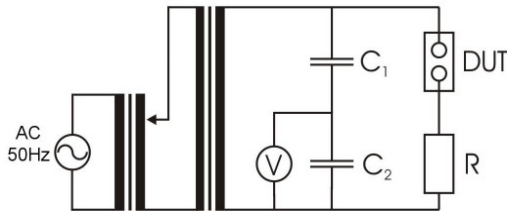

Figure 4: AC measurement setup

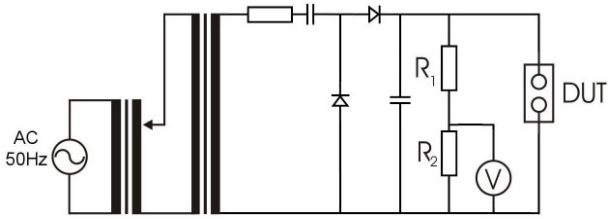

Figure 5: DC measurement setup

The specimens have a sphere-sphere electrode configuration with a distance between the two electrodes of 2 mm. The electrodes are embedded in the material. The diameter of the electrodes is 12 mm. By this configuration, a uniform electrical field with utilization factor of 0.865 is created. Five specimens per material are tested under ac as well as under dc stress.

4.3 Loss Factor and Permittivity

The loss factor and the permittivity of the material are determined according to DIN VDE 0303-4 [11].

The test setup is shown in Figure 6 and consists of a measurement circuit including the device under test and a reference circuit including a reference capacitor. As reference capacitor, a compressed gas capacitor of C_{ref} = 1 nF filled with 10 bar of N_2 and a loss factor lower than 10^{-6} is used. As test voltage, a 50 Hz sinusoidal voltage with 1 kV amplitude is applied. All measurements are carried out at a temperature of 20 °C.

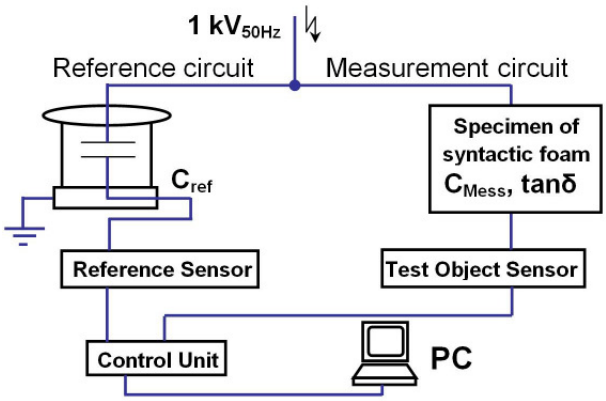

Figure 6: Test setup for the measurement of loss factor and permittivity

The plate specimens have dimensions of 10 cm x 10 cm and a thickness of 3 mm (schematically shown in Figure 7). For the tests, the specimens are placed between a high voltage plate electrode and a measurement electrode. For a better contact between specimen and electrodes the specimens are coated with graphite. An additional protective ring electrode around the measurement electrode is used to avoid any disturbances of the measurement caused by surface currents or electrical field distortions at the edge of the measurement electrode. Five specimens per material are tested.

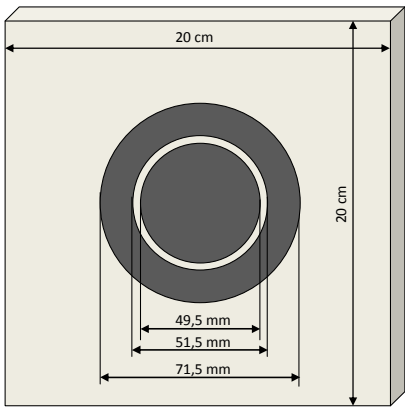

Figure 7: Specimen for loss factor and permittivity measurements coated with graphite

4.4 Volume Conductivity

A test setup according to IEC 60093 [12] is used for the determination of the volume conductivity of syntactic foam (Figure 8). The test voltage U_{HV} is 1 kV dc.

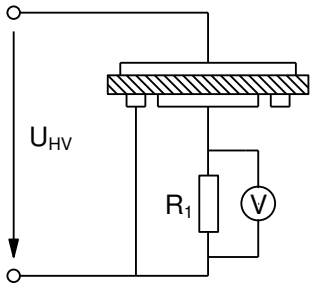

Figure 8: Test setup according to IEC 60093

The same specimen geometry as for the loss factor and permittivity measurements is used (Figure 7). The specimen is placed between the high voltage electrode and the measurement electrode. The current resulting from the application of a high voltage consists of volume and surface current. To determine the volume conductivity of the material it is necessary to measure the volume current separately. Therefore, a grounded protective ring electrode is additionally placed on the specimen around the measurement electrode. Prior to all measurements the electrodes are short-circuited for 4 h to discharge any surface charges, stored on the specimen. Before applying the test voltage, this short-circuit is removed.

The volume current is determined by the measured voltage drop $u_m(t)$ over a resistance R_1 = 220 MΩ, connected in series to the specimen. The volume conductivity of the specimen is determined by equation (1). Here, U_{HV} is the supply voltage, A = 20 cm² the cross-section of the measuring electrode and d = 3 mm the specimen's thickness.

$$\sigma_{specimen}(t) = \frac{u_m(t)}{U_{HV} - u_m(t)} \cdot \frac{d}{A \cdot R_1} \qquad (1)$$

The application of a dc voltage U_{HV} causes transient processes inside of the insulation material. Therefore, the voltage $u_m(t)$ across the resistance R_1 is recorded as a function of the load duration. The measurement is stopped when the voltage have nearly reached a constant value. It is known from previous investigations, that the temperature also influences the conductivity of syntactic foam [1]. Hence, the whole measurement is carried out at a constant temperature of 20 °C within a climatic chamber. Three specimens per material are tested.

5 RESULTS AND DISCUSSION

5.1 Mixing Viscosity

First, the impact of the hollow microspheres' (hms) filling degree on the mixing viscosity is presented in Figure 9. Filling degrees of 40 vol.-% and 55 vol.-% are compared to pure epoxy resin. It can be observed, that the viscosity strongly increases with an increasing filling degree.

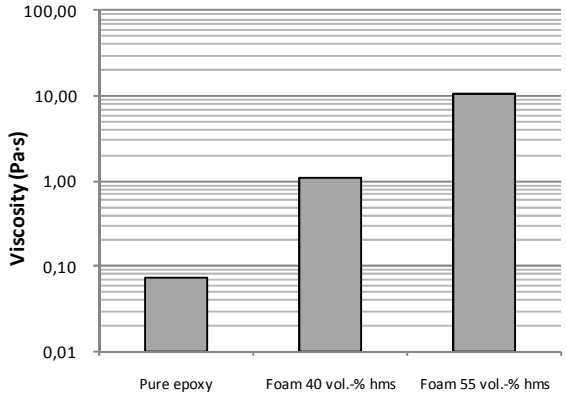

Figure 9: Viscosity of syntactic foam with different filling degrees of hollow microspheres

Furthermore, the impact of 2.5 wt.-% and 5 wt.-% nanofiller on syntactic foam with 40 vol.-% hollow microspheres is presented in Figure 10. The results of syntactic foam with 40 vol.-% hollow microspheres show a minimum viscosity of the liquid material at 2.5 wt.-% filling degree of SiO_2 nanofiller. The mixing viscosity of syntactic foam with 2.5 wt.-% nanofiller is reduced by approximately 7 % compared to conventional syntactic foam, whereas syntactic foam with 5 wt.-% features a higher viscosity.

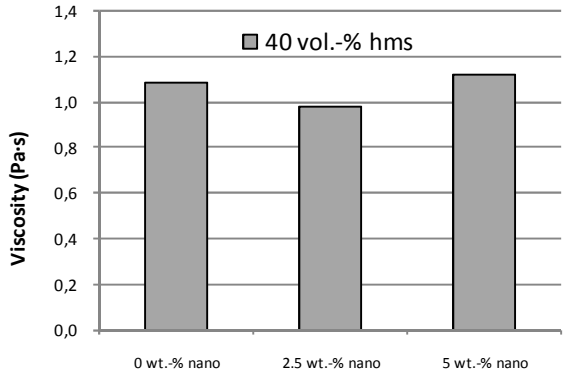

Figure 10: Viscosity of syntactic foam (40 vol.-% hms) with different filling degrees of nanoparticles

Additionally, the impact of 2.5 wt.-% and 5 wt.-% nanofiller on syntactic foam with 55 vol.-% hollow microspheres is presented in Figure 11. A filling degree of about 55 vol.-% represents the maximum filling degree of hollow microspheres, which currently can be manufactured. At higher filling degrees the quality of the material reduces due to the high viscosity.

The results of syntactic foam with 55 vol.-% hollow microspheres also show a minimum viscosity of the liquid material at 2.5 wt.-% filling degree of SiO_2 nanofiller (Figure 11). Here, the nanofiller show a more significant effect in the mixing viscosity. At 2.5 wt.-% nanofiller it is reduced by approximately 42 % compared to conventional syntactic foam. The viscosity of syntactic foam with 5 wt.-% is reduced by 31 % compared to conventional syntactic foam.

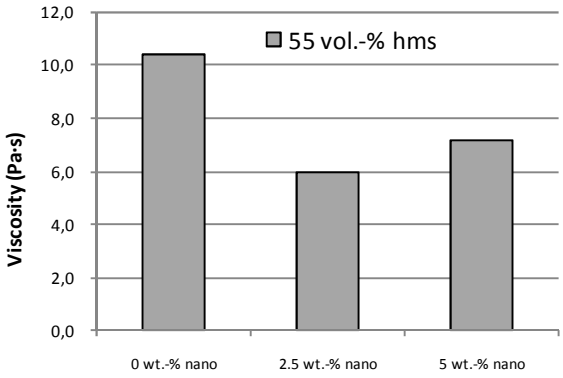

Figure 11: Viscosity of syntactic foam (55 vol.-% hms) with different filling degrees of nanoparticles

In general, the quality of a casting compound material reduces if its mixing viscosity at manufacturing process is very high. Trapped air cannot escape out of the material during degassing process. To improve the quality of the material the viscosity has to be reduced. Hence, an optimum filling degree of the SiO_2 nanofiller for the application as "viscosity reducer" in syntactic foam is reached at 2.5 wt.-%. Due to this result, all following electrical investigations are carried out on syntactic foam filled with 2.5 wt.-% nanofiller.

5.2 Breakdown Field Strength

The results of the breakdown measurements under electrical ac and dc field stress are shown in Figure 12. The results are given as mean values with 95 % confidence intervals. It can be observed that the nanofillers have no significant impact on the ac and dc breakdown field strength of syntactic foam under short-term stress.

The electrical breakdown process in syntactic foam for ac and dc field stress is initiated by gas discharges inside the hollow microspheres [13]. For ac stress gas discharges lead to an erosion of the microsphere walls. Thus, the epoxy resin matrix and its modification play a subordinate role for breakdown process of syntactic foam. For dc stress gas discharges lead to an increasing

electrical field in the epoxy resin matrix between two adjacent microspheres. Thus, an impact of the nanofillers on the breakdown could be potentially expected. Nevertheless, such an effect of the nanoparticles cannot be observed for syntactic foam used in this study.

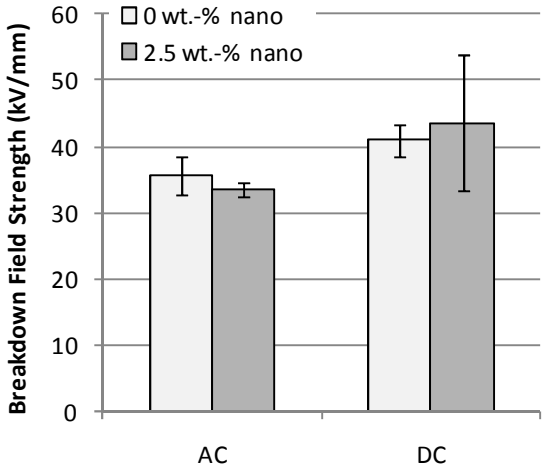

Figure 12: Ac and dc breakdown field strength

5.3 Loss Factor and Permittivity

The results of the measurements show a slight increase of loss factor as well as permittivity, if the material is filled with 2.5 wt.-% SiO_2 nanofiller (Table 1).

Table 1: Results of loss factor and permittivity measurements

Filling Degree	Permittivity	Loss Factor
0 wt.-% nanofiller	2,59	$6{,}7 \cdot 10^{-3}$
2.5 wt.-% nanofiller	2,66	$8{,}3 \cdot 10^{-3}$

The permittivity of SiO_2 particles is slightly higher than of epoxy resin. Therefore a slight increase of the permittivity of syntactic foam modified with nanofiller can be observed. Furthermore, the loss factor also increases as it is known for conventional microfilled polymers.

5.4 Volume Conductivity

The measurement results are given in Table 2.

Table 2: Results of volume conductivity measurements

Filling Degree	Volume Conductivity (S/m)
0 wt.-% nanofiller	$2{,}0 \cdot 10^{-16}$
2.5 wt.-% nanofiller	$2{,}0 \cdot 10^{-16}$

Despite of a slightly higher conductivity of the SiO_2 nanofiller than this of pure epoxy resin, no impact of 2.5 wt.-% SiO_2 nanofiller on the volume conductivity of syntactic foam can be observed. The results can be explained by percolation theory according to [14]. For low filling degrees of nanofiller, the particles do no connect each other to form a network structure, which can change the conductivity.

6 CONCLUSION AND OUTLOOK

6.1 Conclusion

This study focused on SiO_2 nanofiller and its impact on the electrical properties of epoxy resin based syntactic foam. Syntactic foam features a very low density and good electrical properties and can therefore be used as insulation material for lightweight and compact high voltage components. Currently, main challenge is the simplification of its manufacturing process. It has been shown that the mixing viscosity increases by adding microspheres to the pure epoxy. This could lead to trapped gas bubbles inside of casted material due to an insufficient degassing process. Hence, a lower mixing viscosity can help to improve the material quality at high filling degrees of hollow microspheres and to facilitate the manufacturing process.

To decrease the mixing viscosity of the uncured, liquid material different filling degrees of SiO_2 nanofiller were added to the material. Measurements of the mixing viscosity showed a minimum viscosity at a filling degree of 2.5 wt.-%. Here, the viscosity could be reduced by values up to 40 % compared to conventional syntactic foam without nanofiller.

The followed investigations on syntactic foam filled with 2.5 wt.-% nanofiller assured, that the nanofiller has no essential negative influence on the electrical properties of syntactic foam. Electrical breakdown tests, measurements of loss factor and permittivity as well as measurements of the volume conductivity were carried out. The electrical breakdown field strength of the nanofilled syntactic foam under ac and dc stress showed no significant difference to this one of syntactic foam without nanofiller. The permittivity and the loss factor were slightly increased by the nanofillers, whereas the resistivity is not influenced by them.

As main result of this study it can be ascertained that the manufacturing process of syntactic foam can be improved by the addition of SiO_2 nanofillers without an essential negative influences on the electrical properties of syntactic foam.

6.2 Outlook

In this study the focus laid on the impact of SiO_2 nanofiller on the electrical properties of syntactic foam. Investigations on the impact on the mechanical and thermal properties are also planned.

Further investigations are planned to determine the impact of SiO_2 nanofiller on electrical treeing inside syntactic foam. Especially under long-term stress, the time to breakdown of polymers increases by the use of nanofillers [15]. Therefore, the time to breakdown under long-term stress of nanofilled syntactic foam will be analysed.

Furthermore, the mixing viscosity of syntactic foam with filling degrees between 10 and 40 vol.-% of hollow microspheres will be investigated. Here, for each material, the optimum filling degree of nanofiller will be determined.

7 ACKNOWLEDGMENTS

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