IEEE/CSC SUPERCONDUCTIVITY NEWS FORUM (global edition) January 2015.
ASC 2014 manuscript 2MPo1-04 published online in *IEEE Trans. Appl. Supercond.* *(IEEE XPLORE)* DOI: 10.1109/TASC.2014.2376190 on November 26, 2014.

Investigation on the Dielectric Material Parameters and the Electric Conductivity of Syntactic Foam at the Liquid Nitrogen Temperature Range

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***Abstract*—Liquid nitrogen (LN_2) based insulation systems for superconducting components are state of the art. As the dielectric strength of LN_2 based insulation systems can be significantly reduced if bubbles occur an alternative insulation system could be a solid insulation system using LN_2 only for cooling but not as electrical insulation material. This paper discusses syntactic foam as a solid substitution of LN_2 based insulation systems. Syntactic foam consists of a polymer matrix with embedded hollow microspheres (HMS) which have diameters of several 10 µm. Compared to the pure matrix material the HMS filled matrix features a lower density and a significantly reduced thermal contraction when being cooled to cryogenic temperatures. Several syntactic foams are investigated regarding their dielectric parameters (relative permittivity, loss factor) and electric conductivity at liquid nitrogen temperature (LNT). The results show that the investigated parameters of syntactic foam are almost constant in the temperature range of LNT. Furthermore, the loss factor and the relative permittivity at LNT are lower than at room temperature. These effects can be explained by the existence of a secondary glass transition of polymers. At cryogenic temperatures a decrease of the electric conductivity of syntactic foam is detected presumably due to the rise in required energy to lift valence electrons to the conduction band of the polymer at lower temperatures.**

***Index Terms*—Cryogenic temperature, dielectric parameters, electrical conductivity, liquid nitrogen, syntactic foam.**

I. Introduction

In urban areas with an increasing demand of electric power the installation of superconducting components into the power distribution network is an alternative to conventional components due to their higher current carrying capacities [1–3]. The electrical insulation system of superconducting power components is commonly based on liquid nitrogen (LN_2) so that LN_2 simultaneously has a cooling and an insulating function. As LN_2 is a cryogenic liquid it has some disadvantages concerning its dielectric properties e.g. the dielectric strength can be significantly reduced if bubbles occur due to heat generation within the conductor [4]. Furthermore, the routine tests of superconducting power components which are performed immediately after production lose their significance after LN_2 has to be removed for delivery of the component. With a refill of the cable on-site also impurities can get into the component which influence the dielectric and electric properties of the LN_2 insulation [5].

An alternative to LN_2-based insulation systems are solid insulations so that LN_2 has only a cooling function and the disadvantages of LN_2 as an electrical insulation are remediated. Since pure polymeric insulations feature high thermal contractions due to cooling them to liquid nitrogen temperature (LNT) it is necessary to use fillers which are able to reduce the thermal contraction. Otherwise, the insulation will delaminate from metallic or superconducting electrodes as they show much lower thermal contractions than pure polymers [6].

This paper deals with syntactic foam as a solid insulation system. Syntactic foam is a composite material which consists of a polymeric matrix and embedded hollow microspheres (HMS) with diameters of several 10 µm which reduces the thermal contraction of the pure matrix significantly [7]. A scanning electron microscopy picture of syntactic foam is shown in Fig. 1. Since investigations on the dielectric strengths under AC, DC and impulse voltage demonstrate that syntactic foams show good dielectric properties at LNT [8–10] syntactic foam is supposed to be an alternative solid insulation system to LN_2-based insulation systems of superconducting power components. In this paper syntactic foam is examined concerning its dielectric parameters (relative permittivity, loss factor) and its electrical conductivity at LNT as these parameters have a close impact to the heat generation within an electrical insulation. In superconducting components this generated heat needs to be cooled by the cryogenic cooling systems which generate high cooling losses due their low efficiencies.

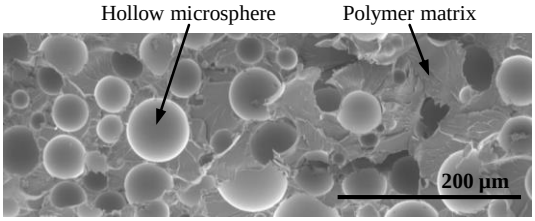

Fig. 1. Scanning electron microscopy picture of syntactic foam.

Manuscript received August 11, 2014. This work was supported by the Deutsche Forschungsgemeinschaft under Grant SCHN 728/10-1.

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II. Investigation Method

A. Material Characterization

In this study an epoxy resin (ER) and an unsaturated polyester resin (UPR) serve as matrix materials of syntactic foam. Furthermore, hollow microspheres with glass walls and hollow microspheres with ceramic walls are used as fillers. The glass microspheres are silanized (silanized glass HMS - SGHMS) to reach a stronger bonding between HMS and the matrix. The SGHMS have a mean diameter of 45 µm, a wall thickness of 1 µm and are filled with sulfur dioxide (SO_2). The ceramic microspheres (CHMS) have a mean diameter of 75 µm, a wall thickness of 7.5 µm and are filled with carbon dioxide (CO_2). An overview of the HMS parameters is given in Table I. The filling degrees of HMS within the syntactic foam are 30 and 50 percentage by volume (vol.%). The measurements are also performed on samples of pure epoxy and unsaturated polyester resin (0 vol.%) to generate reference values.

TABLE I
Overview of the Microsphere's Parameters

Material	Mean diameter	Wall thickness	Filling gas	Glossary
Silanized glass	45 µm	1 µm	SO_2	SGHMS
Ceramic	75 µm	7.5 µm	CO_2	CHMS

B. Sample Preparation

Both matrix materials, ER and UPR, are hot cured polymers. After mixing the polymer with the HMS the compound is degassed and cast into a metallic mold. Then the compound is cured in a furnace at 100° C for 12 h and post cured at 120 °C for 2 h.

The test samples are plate shaped samples with a base area of 100 mm x 100 mm and a thickness of 3 mm.

C. Generation of LNT-Range

The operating temperature of superconducting power components is commonly in the range of 65 – 77 K [11]. Hence, the investigations on the dielectric parameters and the electrical conductivity are performed within this temperature range. To generate these temperatures a pressure-sealed cryostat is used as test vessel. This cryostat consists of a double-walled steel container which can be hermetically sealed by a cover. The double–wall includes a high vacuum to thermally insulate the inner part of the cryostat. As LN_2 has a temperature of 77 K at 0.1 MPa the temperature can be reduced according to the vapor pressure curve of LN_2 by reducing the atmospheric pressure inside the cryostat. At a pressure of 17.4 kPa the temperature of LN_2 reaches 65 K [12]. The pressure inside the cryostat can be reduced by a vacuum pump and is regulated by a computer controlled valve. Both the pressure and the test temperature can be measured by sensors. The test arrangement which is used for the determination of the dielectric parameters and the electric conductivity is completely immerged into LN_2. A schematic of the cryostat and the temperature controlling system is shown in Fig. 2. To cool the samples down to LNT they pass three steps of pre-cooling by using the gaseous phase of LN_2 before they are immerged into LN_2. The three steps are 6 cm and 3 cm above the LN_2 surface and exactly on the surface. On each step the samples remain for 15 min. Before being tested the samples are cooled directly in LN_2 for further 15 min.

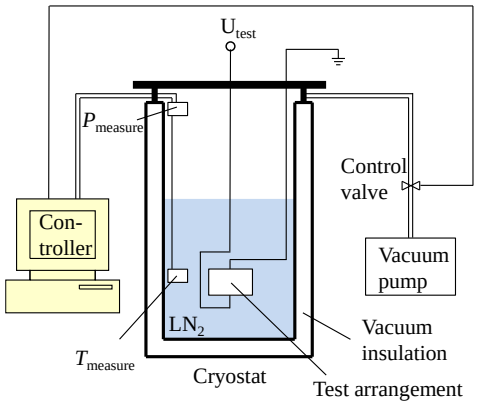

Fig. 2. Schematic of the cryostat used as test vessel.

D. Test Arrangement

The plate shaped samples of syntactic foam which are investigated concerning their dielectric parameters and their electrical conductivities are placed in an axially symmetric test arrangement according to [13] consisting of the electrodes E_1, E_2 and E_3 (Fig. 3). E_1 is the electrode which will be connected to the test voltage, E_2 is the measuring electrode and E_3 serves as a guarding electrode. The guarding electrode features a homogeneous electrical field inside the sample and dissipates leakage currents on the surface around the sample to prevent parasitic errors.

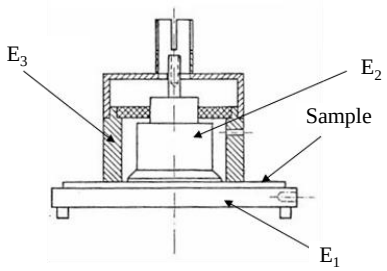

Fig. 3. Schematic of the test arrangement to determine dielectric parameters and the electrical conductivity of plate shaped samples.

The effective area A of the measuring electrode is calculated with the radius r of E_2 and the gap g between E_2 and E_3 by:

$$A = \pi \frac{(2r + g)^2}{4} \quad (1)$$

For the used arrangement the effective area is A = 20 cm².

E. Determination of the Dielectric Parameters

To measure the dielectric parameters (ε_r, tan δ) of syntactic foam a measurement system combined with the test arrangement shown in Fig. 3 is used. The measurement system performs an impedance measurement of the capacitance of the test arrangement with included test sample and of a reference

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capacitor in parallel within a suitable frequency range and determines the phase shift between the currents of both current paths. The test voltage is U_{test} = 1 kV. The reference capacitor is a compressed-gas capacitor which is filled with sulfur hexafluoride (SF_6) and has a capacitance of C_{ref} = 1 nF. The loss factor tan δ and the capacitance of the test arrangement C_{test} are calculated. With the aid of C_{test} and the vacuum capacitance of the test arrangement

$$C_0 = \varepsilon_0 \frac{A}{h} \tag{2}$$

where h is the thickness of the sample and A the effective area of the arrangement the relative permittivity is given by:

$$\varepsilon_r = \frac{C_{test}}{C_0} \tag{3}$$

To determine the influence of the temperature on the dielectric parameters measurements are carried out at ambient temperature (AT) and within the temperature range of LNT. The measurements within LNT are started at a test temperature of 77 K while reducing the temperature to 65 K. For both AT and LNT three samples of each type of syntactic foam are investigated.

F. Determination of the Electric Conductivity

To determine the electric conductivity of syntactic foam the test arrangement with included sample is connected to a DC voltage of $U_{=}$ = 500 V. The current through the sample is measured with the aid of a shunt R_s = 224 Ω in series. Fig. 4 shows the equivalent circuit of the test circuit.

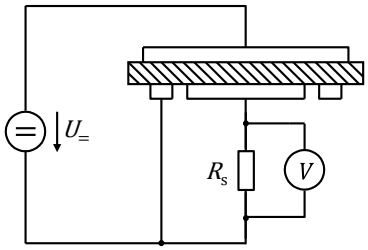

Fig. 4. Equivalent circuit to determine the electric conductivity.

The electric conductivity is calculated by

$$\lambda = \frac{h}{R_{test} \cdot A} \tag{4}$$

where h is the thickness of the sample, R_{test} the measured resistance of the sample and A the effective area known from equation (1).

The measurement is performed for three samples of each type of syntactic foam and each temperature.

III. Results

A. Influence of the Temperature within the LNT-Range

Fig. 5 exemplarily shows the trends of the relative permittivity and the electric conductivity of pure epoxy over the temperature range of 65 – 77 K. It can be observed that a reduction of temperature from 77 K to 65 K does not have a measurable influence on the investigated parameters as their values are almost constant. The same independency from temperature has been found for the dielectric loss factor and the different types of syntactic foam. Thus, the following results of the measurements within the range of 65 - 77 K are only marked as LNT.

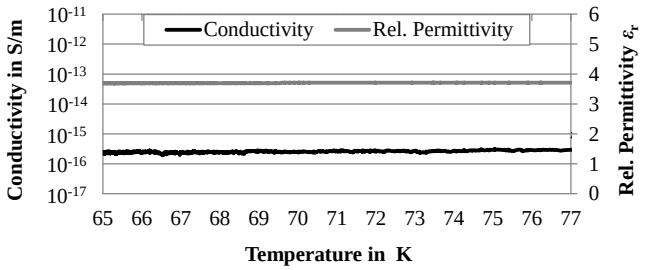

Fig. 5. Relative permittivity and electric conductivity of pure ER within 65 - 77 K.

B. Dielectric Parameters

The relative permittivity of syntactic foam based on ER and UPR over the filling degree is shown in Fig. 6 and Fig. 7, respectively. For each series of measurement the 95 % confidence interval is given. For each type of syntactic foam and AT as well as LNT it can be observed that the permittivity decreases with increasing filling degree.

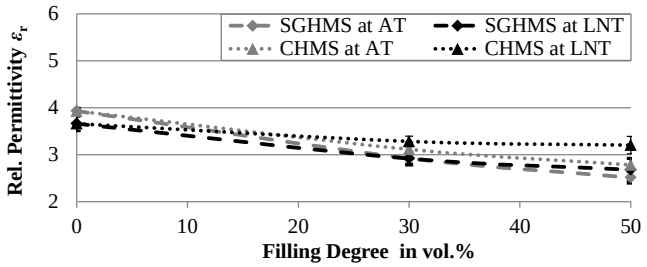

Fig. 6. Relative permittivity of syntactic foams based on ER.

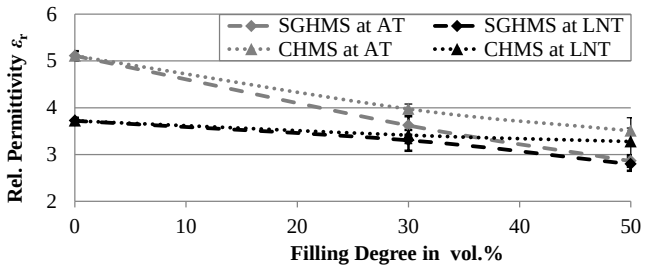

Fig. 7. Relative permittivity of syntactic foams based on UPR.

Furthermore, at LNT the permittivities of the pure matrix materials are lower than at AT. For UPR this effect is bigger than for ER. Comparing the filler types it can be seen that SGHMS feature a lower permittivity than CHMS.

The loss factors of the different types of syntactic foam are illustrated in Fig. 8 (ER) and Fig. 9 (UPR). For all samples the loss factor at LNT is lower than at LNT. Compared to ER the gap between the values of the loss factors at AT and LNT is much higher for UPR based syntactic foams than for ER based foams. Except for the loss factor at AT of ER based syntactic

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foams with 50 vol.% of HMS all loss factors nearly remain on the same value for each syntactic foam and filling degree.

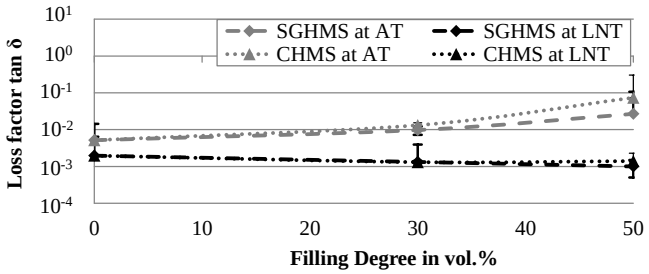

Fig. 8. Loss factor of syntactic foams based on ER.

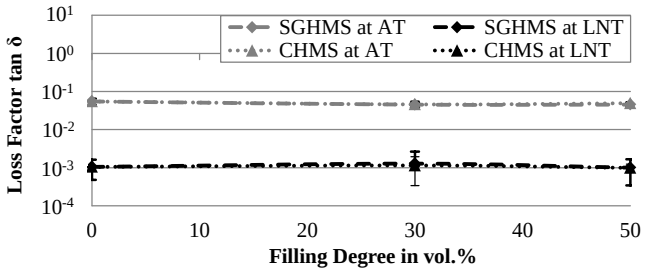

Fig. 9. Loss factor of syntactic foams based on UPR.

C. *Electric Conductivity*

The electric conductivity at different test temperatures of syntactic foams based on ER and UPR including the 95 % confidence interval are shown in Fig. 10 and Fig. 11, respectively.

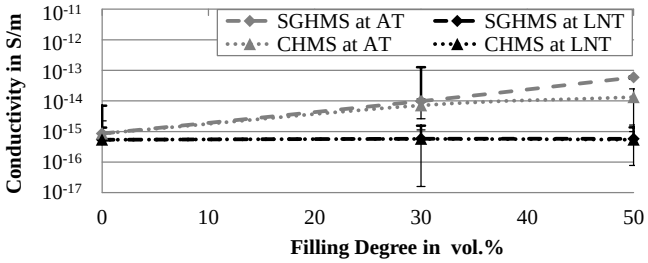

Fig. 10. Electric conductivity of syntactic foams based on ER.

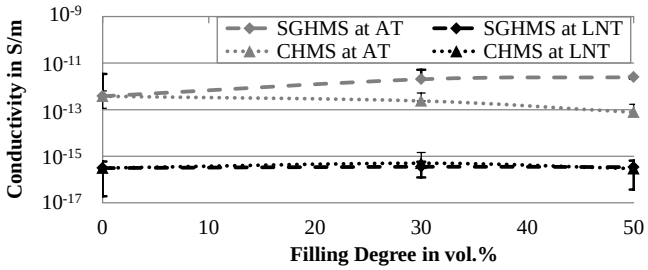

Fig. 11. Electric conductivity of syntactic foams based on UPR.

It can be observed that pure UPR at AT has a conductivity which is higher than at LNT by two orders of magnitude. The conductivity of pure ER at AT and LNT is similar to the conductivity of pure UPR at LNT. Furthermore, at LNT the filling degree of syntactic foam has no influence on the conductivity for both matrix materials. At AT the conductivity of ER based foams is increased with increasing filling degree, though CMHS feature a slighter increase than SGHMS.

For UPR based syntactic foams at AT SGMHS lead to a slight increase of the conductivity with increasing filling degree, whereas CHMS lead to a slight decrease.

IV. Discussion

Measurements of the relative permittivities of syntactic foam show that the permittivities decrease with increasing filling degree. The matrix materials have permittivities in the range of 3 – 5 so the permittivity of syntactic foam is reduced due to the gas inside the HMS which has a permittivity $\varepsilon_{r,gas} \approx 1$. As the permittivity of the gas does not rise for lower temperatures the same trend can be recognized for measurements at LNT. However, at LNT the permittivities of the matrix materials are reduced compared to the values at AT. This can be explained by a secondary glass transition temperature which is passed by cooling the materials to LNT. This glass transition can be identified at temperatures around 140 K [14]. By passing the secondary glass transition temperature molecule rotations freeze. That means that the mobility of molecules and molecule chains is decreased. Thus, the permittivity is reduced at LNT. Since electron polarization is the only remaining polarization process left [14], the relative permittivities of ER and UPR are almost similar at LNT although they are different at AT.

The measurements of the electric conductivity of syntactic foam show that the conductivity is influenced by the filling degree of HMS at AT. For syntactic foam based on ER the conductivity increases with increasing filling degree of HMS due to the higher conductivity of the walls of SGHMS and CHMS compared to ER. Since the increase of the conductivity of syntactic foam is higher when using SGHMS instead of CHMS the conductivity of the SGMHS themselves is supposed to be higher than the conductivity of the CHMS. For syntactic foams based on UPR at AT it can be found that the conductivity of the SGHMS is supposed to be higher than conductivity of the CHMS, too. The more conductive SGHMS feature a slight increase of the conductivity whereas the less conductive CHMS lead to a slight decrease of the conductivity. At LNT the conductivities of syntactic foams based on ER and UPR are unaffected by the filling degree. This means that the conductivity of the HMS has to be reduced as well as the conductivity of the matrix materials. Otherwise, the conductivity of syntactic foams would be increased by increasing the filling degree. It is assumed that the lower conductivity of syntactic foams at LNT is based on lower thermal energy of the matrix materials at lower temperatures. At AT electron movement within an insulation material can be explained by the imperfect lattice. Electrons which are injected out of the electrodes by high field stresses can be lifted from lattice imperfections to the conduction band by external energy. Since the thermal energy of the insulating material is lower at LNT more external activation energy is needed to lift electrons to the conduction band [15]. Furthermore, the work functions of the electrodes are increased at LNT so that a higher electrical field is necessary to inject electrons into the insulation material [16].

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The loss factor of syntactic foam at AT can be observed as nearly independent from the filling degree for syntactic foams based on UPR. A slight increase of the loss factor of ER based foams at high filling degrees shows that the hollow interiors of the HMS has an impact on the loss factor if a crucial amount of HMS are filled into the matrix. Because of the higher loss factor of pure UPR compared to ER it is assumed that this crucial filling degree of HMS for UPR based syntactic foams is higher, too. Thus, an increase of the loss factor cannot be identified up to a filling degree of 50 vol.%. At LNT the loss factor of syntactic foams of both matrix materials is reduced and constant with the filling degree. The loss factor is a function of the conductivity and the permittivity. Since the permittivity decreases with increasing filling degree for each syntactic foam a change in the loss factor with increasing filling degree is expected. It has not yet been found if the significantly reduced pressure within the HMS at LNT, which is immerged from isochoric phase transitions of the filling gas [9] has an influence on the loss factor of syntactic foam.

At last, the investigated parameters do not show a measurable dependency of the temperature, when it is changed within the range of 65 – 77 K. While the relative permittivity keeps constant there is no further secondary glass transition which syntactic foam passes during the cooling process. Therefore, the material structure does not significantly change within this temperature range so that no significant change of the other parameters investigated is observed.

V. Conclusion

In this paper syntactic foam based on ER or UPR and filled with silanized glass HMS or ceramic HMS is investigated regarding its dielectric parameters (relative permittivity and loss factor) and its electric conductivity. The main results are as follows:

- All investigated parameters are independent of temperature within the range of 65 - 77 K. The assumed reason is that syntactic foam does not pass another secondary glass transition below a temperature of 140 K and above a temperature of 65 K.
- Due to the relative permittivity of the filling gasses $\varepsilon_{r,gas} \approx 1$ the HMS reduce the permittivity of syntactic foam for ambient and liquid nitrogen temperature. With decreasing temperature the permittivity is decreased as well. This is assumed to be because of the secondary glass transition the matrix materials pass at a temperature of about 140 K by cooling which freeze molecular mobility.
- The electric conductivity of syntactic foam is reduced at LNT. The assumed reason is the thermal energy of the materials is lower, thus, a higher external energy by terms of an electrical field is necessary to lift electrons into the conduction band of the insulation material. Furthermore, due to higher work functions of the electrodes at LNT injections of electrons occur at higher electrical fields.

Acknowledgment

The authors would like to thank Mr. Patrick Poss for his active support during this investigation.

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