Materials Today Communications 36 (2023) 106656

Contents lists available at ScienceDirect

Materials Today Communications

journal homepage: www.elsevier.com/locate/mtcomm

Dielectric properties of unidirectional and biaxial flax/epoxy composites at frequencies up to 1 GHz

Philipp Baron [a,*], Philipp Lenz [a], Klaus Peter Koch [b], Armin Wittmann [a], Georg Fischer [c]

[a] Laboratory for Applied Production Engineering, Trier University of Applied Sciences, Schneidershof, 54293 Trier, Germany
[b] Department of Engineering, Trier University of Applied Sciences, Schneidershof, 54293 Trier, Germany
[c] Institute for Electronics Engineering, FAU Friedrich-Alexander-Universität Erlangen-Nürnberg, Cauerstraße 9, 91058 Erlangen, Germany

ARTICLE INFO

Keywords:
Dielectric analysis
Relative permittivity
Flax/epoxy composites
Unidirectional fiber orientation
Biaxial fiber orientation
Microstructure flax/epoxy

ABSTRACT

The relative permittivity of flax/epoxy composites in unidirectional and biaxial orientations was mapped in the frequency range of 1 kHz to 200 kHz, and for the first time in the range of 1 MHz to 1 GHz. In addition, permittivity was investigated for the first time in the temperature range between − 20 °C and 50 °C. These composites, produced using the vacuum infusion process, are increasingly used for sustainable and lightweight structural components in the automotive industry. The relative permittivity was determined using a self-developed plate capacitor with an LCR bridge and an impedance analyzer. An examination of the microstructure of the flax/epoxy composites shows that the fibers are disordered in the composite, resulting in local variations in fiber volume fraction. Furthermore, it was shown that the matrix also infiltrates into the fiber itself, resulting in an increase of the matrix fraction. It was found that unidirectional fabrics had a higher relative permittivity than biaxial fabrics, due to a higher fiber volume fraction and lower proportion of epoxy. The results suggest that it is the fiber volume fraction, rather than the manufacturing process and fiber orientation, that primarily determines the relative permittivity. It was also found that the permittivity continues to decrease below room temperature and thus behaves in a manner typical of the material in this temperature range as well.

1. Introduction

The interest in natural fiber composites has continuously increased in recent years [1]. This is due to their outstanding mechanical properties, low cost, high lightweight potential and, above all, high environmental compatibility [1,2]. The renewable plant fibers even have a negative CO_2 balance due to their photosynthesis [2], which makes them one of the most relevant materials for science and industry in the future. These advantages make the renewable material particularly interesting for vehicle construction [3].

At present, natural fiber composites are already being used in automotive engineering for interior door panels, storage compartments, seat trim and even sunroof frames, among other things. In the latter, for example, weight savings of up to 50 % can be achieved compared to the classic metal design [1]. Fiber composites such as carbon or glass fiber composites are consequently being pushed further out of the market by progressive developments in the field of plant-based composites and their weight advantage. [4].

Due to the advantages mentioned above, not only the interior but also load-bearing structural components of the vehicle are to be developed from natural fiber composites in the future. For the use of natural fiber composites in safety-relevant components in the vehicle, however, mechanical component monitoring during use is essential. The reason for this is the fluctuating mechanical properties of the fiber due to non-uniform plant growth [1,5]. A new type of sensor element, consisting of electrically conductive materials which are inserted into the composite, is intended to monitor the wear condition of the composite with the aid of high-frequency alternating currents. The electrical mode of operation is based on wear measurement, which was already demonstrated for cables and wires in moving applications [6]. Here, transmission loss and phase response are measured in-situ over the service life and conclusions are drawn about the remaining service life based on a change in the signal. To be able to electrically design the HF-sensor element, for example, about its characteristic impedance, it is necessary to know the dielectric properties of the fiber composite material. Many publications already exist describing the complex relative permittivity of natural fiber composites. However, these neither cover the manufacturing process used, nor the relevant frequency range.

* Corresponding author.
E-mail address: P.Baron@mb.hochschule-trier.de (P. Baron).

https://doi.org/10.1016/j.mtcomm.2023.106656
Received 16 February 2023; Received in revised form 19 June 2023; Accepted 11 July 2023
Available online 14 July 2023

For the first time, a flax/epoxy composite material is to be used, which is produced using the vacuum infusion process. It has high mechanical specific strength and stiffness [7,8] and performs excellently compared to other natural fiber composites [2]. For flax/epoxy prepregs with a fiber volume fraction of 50 % and unidirectional fiber orientation with a frequency between 100 mHz and 1 MHz, it has already been shown that the temperature dependence of permittivity and loss angle decrease significantly above a frequency of 1 MHz [9,10]. This material behavior can be attributed to the polarization of the water molecule dipoles of the flat fiber [9–11]. Basically, higher the temperature, lower the orientation polarization. This relationship is described by the Debye equation, which relates the permittivity to the molecular quantities [12, 13]. At higher frequencies, the orientation polarization decreases, since due to their inertia the dipole molecules can no longer follow the external field and finally only displacement polarization takes place. At this point, the Debye equation changes into the Clausius-Mossotti equation, which neglects the orientation polarization [14].

In addition, flax/epoxy composites manufactured by hot-pressing method with fiber volume fractions of 30 %, 45 % and 60 % were investigated in the frequency range between 0.1 and 10 MHz [15]. A single measurement of the flax/epoxy composite at 9.375 GHz was also performed for fiber volume fraction of 60 %. This results in an ε'_r of 4.93 and $\tan\delta$ of 0.018. [15]. For flax reinforced polypropylene composites manufactured by compression molding, an ε'_r of 1 and a $\tan\delta$ of 0.06 could be measured at 1 MHz [16]. In the discussion section of this paper, a table summarizing the results of the complex relative permittivity of flax/epoxy composites from the literature In addition, results exist on the dielectric behavior of other natural fiber composites such as loofah [17], rice straw [17], sisal [17], jute [18], coir [19], banana [20], bamboo [21] and sunn hemp [22]. These are only comparable with the flax/epoxy composite to a limited extent and are therefore not considered further.

The aim of this work is to measure the frequency dependent complex relative permittivity $\underline{\varepsilon_r} = \varepsilon'_r - j\varepsilon''_r$ of unidirectional (UD) and biaxial (BA) flax/epoxy composites fabricated by vacuum infusion method with high frequency alternating currents. Here, the dielectric properties will be presented for the first time in the frequency range up to 1 GHz. The fiber orientation plays a crucial role since it has an effect on the mechanical properties of the material [2]. In addition, the complex relative permittivity in the temperature range between − 20 °C and 50 °C is presented for the first time in the frequency area between 1 kHz and 200 kHz. This temperature range is particularly relevant for the subsequent application of the sensor element. Previous work shows measured values starting at 40 °C [9,10]. In frequency range up to 1 GHz, temperature plays a subordinate role, since there is no longer any influence on relative permittivity above a frequency of 1 MHz [9,10,23].

2. Methods

2.1. Materials

The samples are made of flax/epoxy using the vacuum infusion process [24]. Flax fibers of the type ampliTex Art. No. 5025 [25] with 280 g/m^2 (gsm) from the manufacturer bcomp are used. These scrims have the advantage that they can be combined to samples of any combination of fiber orientation due to the unidirectional fiber layer. Thus, unidirectional as well as biaxial specimen can be prepared from the identical starting material to allow high comparability of the results. Epoxy Resin L and Hardener GL1 are used to produce the composite.

Both the unidirectional and the biaxial scrims are made of four layers with the orientation UD = (0°, 0°, 0°, 0°) and BA = (0°, 90°, 90°, 0°). For this purpose, the fiber mats are aligned on a glass plate and then sealed airtight with a film. Due to this manufacturing process, usually only one face of the sample is smooth in the vacuum infusion process. For the present work, in deviation from the standard procedure, an additional Plexiglas pane is positioned between the flax fiber and the foil, so that a smooth sample surface can be ensured on both sides. This is essential for a dielectric measurement with surface contact, so that as little air as possible is trapped between the sample and the electrodes of dielectric probe, that could falsify the measurement. The epoxy resin is then drawn through the scrim with the aid of vacuum. The reason for choosing the dimensions of the specimen is to match the dimensions of the parallel-plate capacitor, which is presented in the following chapter. In addition, a sheet of Epoxy Resin L, which is used in the natural fiber composite, was cast to examine its dielectric properties separately. The specimen has dimensions of 450 × 450 mm with a thickness of 2.9 mm. In the supplemental files is a picture of the specimens used for the measurement.

For the determination of the fiber volume fraction, the specimen are embedded with cold embedding medium (CEM100 Blue) [26] and then ground with a 1500 grit silicon carbide paper. Subsequently, fiber and matrix are manually colored on the microsections shown using image processing software. Thus, the relationship between fiber and matrix can be determined. Automated determination is only possible to a limited extent, since the edge areas of the fiber are not accurately detected due to the small differences in contrast, resulting in errors. The fiber volume fraction of the unidirectional sample is approx. 45 %. At about 39 %, the biaxial specimen has a lower fiber volume fraction, which can be attributed to the greater packing density of the unidirectional fiber layer caused by the manufacturing process [27]. In unidirectional orientation, the fiber planes can slip into each other, which is not possible when the orientation is 90° to each other. The vacuum, which leads to planar loading on the flax fiber during specimen fabrication, enhances this effect. This better space utilization leads to a higher fiber volume fraction, since the volume between the fibers is minimal. A detailed consideration of the microstructure of the flax/epoxy samples is given in chapter 3.1. Microstructure analysis is performed using a JSM-6610 [28] scanning electron microscope (SEM) and industrial micro-computed tomography (μCT). The X-ray device uses a 225 kV microfocus X-ray tube and offers a real resolution of 900 nm/voxel.

2.2. Measurement

The measurement of the relative complex permittivity takes place with the aid of two measuring setups and instruments. This is due to the limited measuring ranges of the measuring instruments utilized. By combining the devices, a frequency range from 1 kHz to 1 GHz can be represented (Fig. 1).

The first measurement setup for frequencies from 1 kHz to 200 kHz consists of a parallel-plate capacitor with guard electrode according to DIN EN IEC 62631–2–1:2018 [29] and the Rohde & Schwarz HM8118 LCR Bridge/Meter [30]. Publications already exist with measured values of the complex relative permittivity of flax/epoxy composites in the selected frequency range from 1–200 kHz. To ensure comparable results

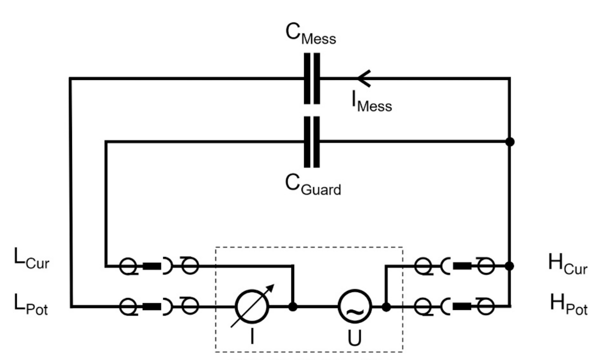

Fig. 1. Schematic measurement setup of parallel-plate capacitor and HM8118 LCR Bridge/Meter.

with those already known, the composite used in this work is therefore also analyzed in the frequency range between 1 kHz and 200 kHz. The diameter of the smaller capacitor plate, which is enclosed by the guard electrode, is 418 mm. The guard electrode has a gap of 0.5 mm to the capacitor plate and an outer diameter of 440 mm. It serves to minimize the stray fields in the edge region of the capacitor and thus improves the accuracy of the measurement result. The opposite capacitor plate without guard also has the outer diameter of 440 mm. The capacitor and the corresponding geometries were designed specifically for the tests with flax/epoxy composite material. The design was made according to the measuring range of the LCR meter and aluminum was used as the material. A picture of the measurement setup is shown in the supplemental files. Before each series of measurements, a calibration is performed. This is done by aligning, the short-circuit and open-circuit states for all measuring points (frequencies). It reduces system-related errors, such as the parasitic effects of the test leads. The electrical connection of the LCR bridge/meter is shown schematically in Fig. 1. As can be seen, the four-wire measurement technique is used to prevent parasitic resistance. To verify the self-built test rig, PTFE with defined dielectric properties was measured. The values from the data sheet could be reproduced. For the sake of clarity, the results of this control group can be viewed in the digital appendix.

The relative permittivity $\varepsilon_{r(NfC)}$ of the natural fiber composite is calculated by the following equation:

$$\varepsilon_{r(NfC)} = \frac{C_{NfC}}{C_{Air}} \cdot \varepsilon_{r(Air)} \cdot \frac{d_{NfC}}{d_{Air}} \quad (1)$$

As a reference the capacitance of the ambient air is measured, since its relative permittivity is known to be $\varepsilon_{r(Air)} = 1.00059$ [31]. To ensure a constant distance between the capacitor plates is guaranteed, spacers made of PLA are used in around the guard electrode. These therefore have no influence on the measurement of the capacitance. Subsequently, the unidirectional and biaxial flax/epoxy composites and the pure epoxy sample are then inserted between the capacitor plates. To ensure that the plates lie flat against the samples to be measured, they are additionally pressed together. Due to the size of the plate capacitor in relation to the ratio between fiber size and matrix of the flax/epoxy composite, local differences in the composite structure can be ignored during the measurement. The reason for this is that there is a sufficient averaging over the cross-section of the plate capacitor. As this measurement setup operates in the frequency range between 1 kHz and 200 kHz, a temperature dependence of the complex relative permittivity is to be expected. For this reason, the measurements are carried out in a Binder MK 112 [32] climatic chamber in the temperature range between − 20 °C and 50 °C. Since the climatic cabinet has a side opening for the test leads to pass through, the door can remain closed for the entire duration of the test.

The second measurement setup consists of the Keysight E4991A Impedance/Material Analyzer [33] and the 16453A Dielectric Material Test Fixture [34] with a measuring head diameter of 15 mm. This fixture allows measurements from 1 MHz to 1 GHz. Calibration is performed with a 7 mm cal kit at the substrate level. In addition to the open and closed states (open/short standard), a calibration standard made of polypropylene with known dielectric properties is used. Since the instrument performs the calculation according to the formula shown above independently after successful calibration, the relative permittivity of the natural fiber material can be output directly.

For both measurement setups, the unitless loss angle $\tan\delta$ is given. The conversion to the imaginary part of the relative permittivity is done according to the following equation:

$$\tan\delta = \varepsilon'' / \varepsilon' \quad (2)$$

Unlike the previously presented setup, the measuring head of this measurement setup is in the same order of magnitude as the fiber to matrix ratio. For this reason, it is necessary to move the specimen slightly during each measurement to account for local differences in the composite structure. If the specimen would not be moved, it is possible that e.g., a spot with increased fiber content would be measured and thus a falsification of the results occurs.

3. Results and discussion

In the first part of this chapter, the microstructure of the flax/epoxy samples is presented. On the one hand, this should serve to better characterize the samples so that it becomes apparent how the ratio between flax and epoxy is. This is important because the complex relative permittivity as a function of fiber volume fraction and thus the flax/epoxy ratio will be addressed. On the other hand, a better comparison to previous research results should be possible to support statements in the discussion section.

3.1. Microstructure analysis of the flax/epoxy specimen

Fig. 2 shows the microstructure of the unidirectional (Figs. 2, a1-a3) and the biaxial (Figs. 2, b1-b3) flax/epoxy specimen. The images were taken by micro-computed tomography, thus allowing non-destructive component observation. Three sections are shown of both specimen variants, which are taken in longitudinal section through the specimen at intervals of 5 mm.

The flax fibers have a relative movement to each other in the longitudinal direction. This movement is exemplified by the yellow circles (UD: I-III and BA: IV-VI), which mark one flax fiber each for both samples. The samples thus exhibit strong local differences in the composite structure as the fibers move among themselves. A statement about the complex relative permittivity can thus only be made with limited accuracy for local sections on the order of the plant fiber since the ratio between fiber and matrix is not constant. As already mentioned in the previous chapter, this inconsistencies of the composite material necessitates an averaging of the recorded measured values of the complex relative permittivity if the size of the measuring probe is not a multiple of the dimension of a plant flax fiber.

A closer look at the fiber reveals that the flax itself consists of many smaller plant fibers that are interwoven with each other. Since the resolution of micro-computed tomography, which was used for Fig. 2, is not sufficient for the individual observation of a single fiber, the individual observation is carried out with a scanning electron microscope.

Fig. 3(a) shows a cross-sectional micrograph of the flax/epoxy composite. The plant fiber itself consists of many smaller fibers. This becomes even clearer when looking at Fig. 3(b), which is a magnification of Fig. 3(a). It can also be seen that the vacuum infusion process allows the epoxy to reach into the interstices of the individual flax fiber components, thus minimally reducing the fiber volume fraction. With the help of the micro sections, it can also be ruled out that there is air in the composite, which has a significant influence on the complex relative permittivity and mechanical strength. Figs. 3c and 3(d) show two flax fibers without matrix, which again clearly show that a flax fiber is interwoven from many smaller fibers.

3.2. Dielectric Measurement

Fig. 4(a-f) shows the real and imaginary parts of the relative permittivity versus frequency. The individual measurement points are averaged from three measurements. Fig. 4 shows the complex relative permittivity of the flax/epoxy composites and the epoxy matrix investigated in this work in the frequency range between 1 kHz and 200 kHz. The set of curves in each of the six figures displays the measured values for the temperatures in the range between − 20 °C and 50 °C. Each measurement point is averaged from three individual measurements. From the scatter bars, a small standard deviation is observed for all measurements.

Fig. 4(a) shows the real part of the permittivity of the unidirectional

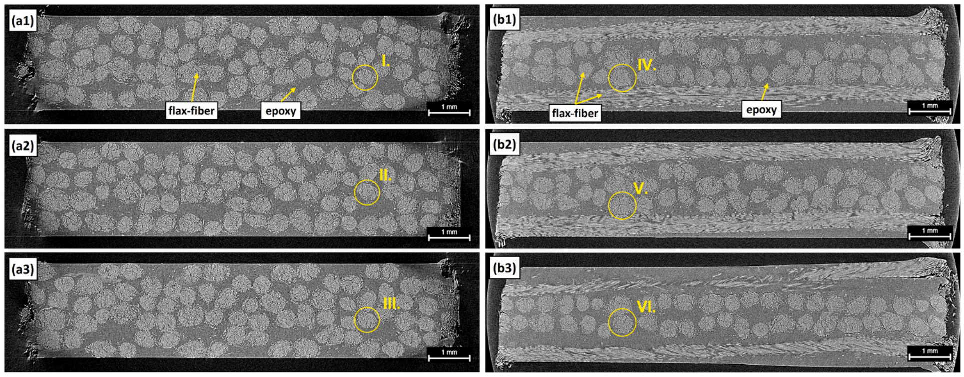

Fig. 2. Micro-computed tomography (μCT) for the unidirectional (a) and biaxial (b) microstructure of the flax/epoxy composites. (a1 - a3, unidirectional) and (b1-b3, biaxial) show cross-sectional images of the flax/epoxy composite at 5 mm. (I. - III., unidirectional) and (IV. - VI., biaxial) show one fiber each and illustrate the variance due to manufacturing.

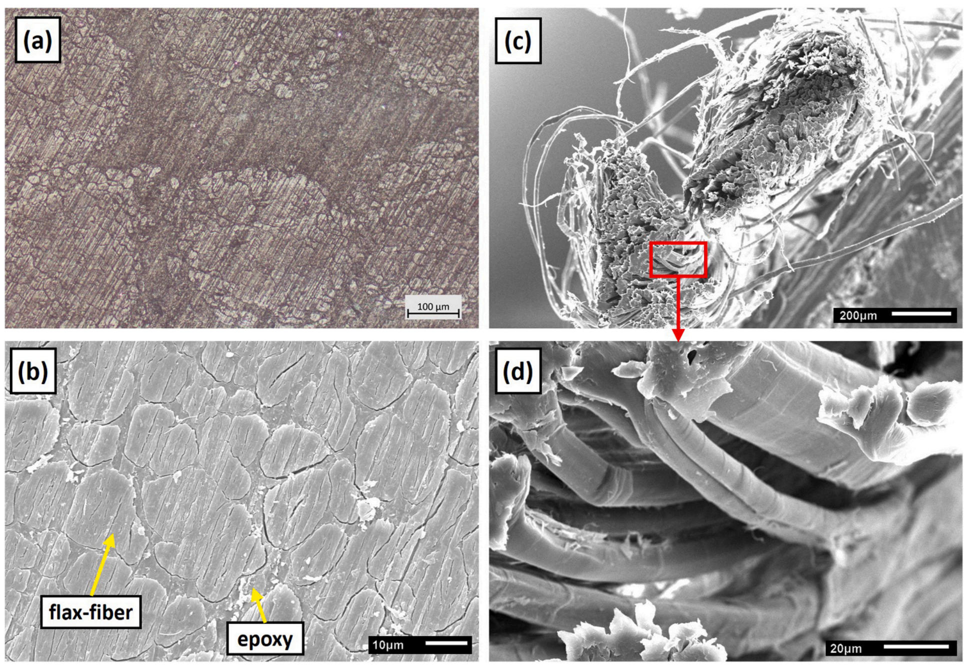

Fig. 3. Individual observation of the flax fiber in the composite. a) Microscope image of a micrograph of the flax/epoxy composite. b) SEM image of the flax/epoxy composite. c) SEM image of the pure flax fiber. b) SEM image of the pure flax fiber with magnification to the fiber constituents.

flax/epoxy sample. At room temperature (20 °C), the permittivity is $\varepsilon'_r = 5 - 5.1$. The real part of the permittivity increases as a function of temperature. This spreading is due to the improved conductivity of the composite when the temperature increases [9,35] and has been shown several times in the context of natural fiber composites [9,10,12,35–37]. Furthermore, at the frequency of 1 kHz, a stronger effect of temperature on permittivity can be detected. This effect is even stronger in the imaginary part ε''_r, which will be discussed below. This is due to the dispersion and associated polarization effects of the water molecules in the plant fiber, which have already been explained at the beginning. For high frequencies and temperatures, the relative permittivity start decreasing. It is striking that the measured values scatter strongly at a frequency of 1 kHz, compared to the higher frequency measured values. Small loss angles mean resistances in the Megaohm range. Thus, the currents to be measured at the applied excitation of 1 V are in the range of microamperes, which are difficult to resolve. The interpretation of the imaginary part of the relative permittivity at 1 kHz is therefore only possible to a limited extent. A consideration of measured values below 1 kHz is therefore also not to be evaluated as meaningful. These measurement inaccuracies are found for all measurements shown in Fig. 4. The imaginary part of the relative permittivity of the unidirectional flax/epoxy sample is shown in Fig. 4(b). Again, at 1 kHz there is a clear spread as a function of temperature. This already drops significantly at 10 kHz. For the measured value at 50 °C, ε''_r drops from 1 to 0.24. The reason for this behavior is polarization effects of the water molecules [12]. Fig. 4(c-d) shows the real and complex permittivity of the biaxial flax/epoxy specimen. The characteristics of the measured values for both flax/epoxy samples are the same, which is conclusive since both samples are made of identical materials. The results differ in the magnitude of the measured permittivity from those of the unidirectional specimens. The unidirectional flax/epoxy composite has a higher permittivity over the entire frequency range than the biaxial flax/epoxy scrim. At the temperature of 20 °C and 200 kHz, the unidirectional flax/epoxy sample has a permittivity ε'_r of 5.1 and the biaxial sample has a permittivity ε'_r of 4.52. This is due to the higher fiber volume fraction of the unidirectional specimen. In Fig. 4(e-f), pure epoxy resin has a significantly lower permittivity than the composite of flax fiber and matrix. At a temperature of 20 °C and frequency of

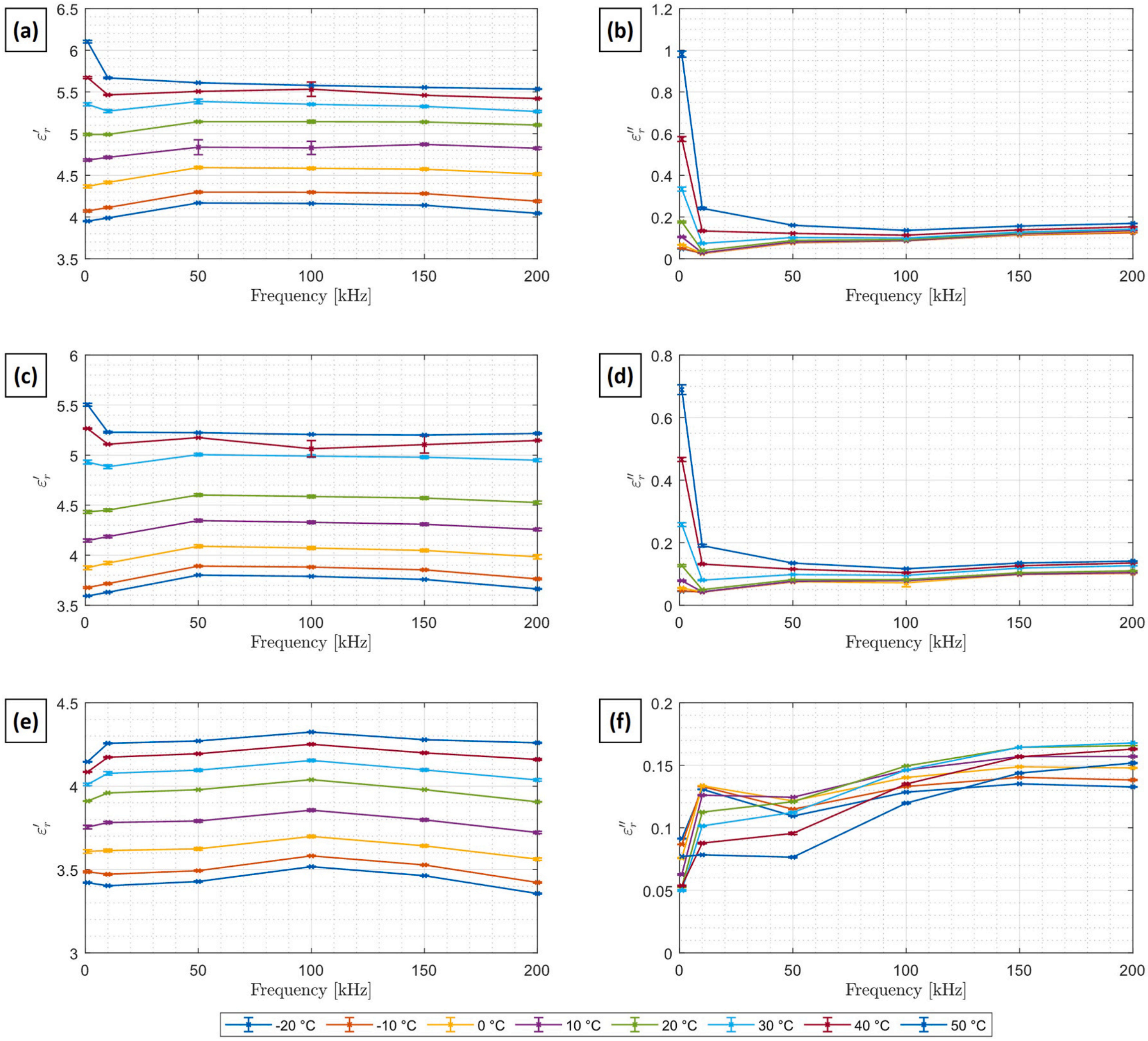

Fig. 4. Complex relative permittivity in the frequency range between 1 kHz and 200 kHz as a function of temperature for (a-b) unidirectional flax/epoxy composites, (c-d) biaxiale flax/epoxy composites, and (e-f) pure epoxy.

200 kHz, the permittivity ε'_r of the epoxy is 3.9 and that of the unidirectional flax/epoxy sample is 5.1. For the matrix material, a temperature dependence over the frequency range can also be seen. This is due to an increase in the mobility of the polymer molecules, which leads to an increase in the real part of the permittivity [38,39]. The opposite behavior at 1 kHz compared to the flax/epoxy sample can only be explained by the lack of organic content in the epoxy. It should also be mentioned at this point that measurement inaccuracies can occur at 1 kHz. For the imaginary part of the permittivity of the epoxy, an increase over temperature can be seen, especially in the range between 1 kHz and 10 kHz, which has already been shown several times [39–42].

The lower proportion of epoxy resin in the composite thus ensures that the influence of the pure flax fiber on the electrical measurement is greater and the overall permittivity is higher in the unidirectional scrim. This differentiation between flax fiber and epoxy is independent of the scrim orientation and is already known [9]. The relative permittivity of the plant fiber has also been found to be greater than that of the epoxy in other natural fiber composites [17,18,20].

To compensate for the disadvantages of measuring in the low frequency range and to produce results closer to the intended range of use at a few GHz, the identical samples were analyzed in the second step using the Keysight E4991A Impedance/Material Analyzer and the 16453A Dielectric Material Test Fixture. Readings were taken in the frequency range between 1 MHz and 1 GHz. The results of the real and imaginary parts of the relative permittivity are shown in Fig. 5. A total of 801 measurement points were recorded over the frequency range, each of which is averaged from ten individual measurements. Attention was paid to moving the specimens back and forth under the measuring head for each new measurement to consider potential influences of the fiber structure. The measured values at 1 MHz vary greatly. The reason for this is that the measurement method used does not work accurately in this frequency range. Here, too, this behavior is due to the combination of a small measured value and high inaccuracy caused by the relatively small measuring frequency. In Fig. 5, for the sake of clarity, a scatter bar is shown only at every 20th value, since otherwise the measurement curve itself would no longer be recognizable.

Fig. 5(a) shows that the real permittivity of epoxy, unidirectional and biaxial composites decreases with frequency. This frequency dependence is characteristic and, as mentioned earlier, can be explained by the different polarization effects [43]. Also in this measurement, a clear difference between the unidirectional and biaxial flax/epoxy fabrics can be seen, which can be attributed to the different fiber volume fraction. Thus, it is proven that the fiber orientation and the related fabrication-related fiber volume fraction has an influence on the real permittivity. A comparison of the results in Fig. 4 and Fig. 5 shows that the results are conclusive with each other. At a frequency of 200 kHz, a permittivity ε'_r of 5.1 was shown for the unidirectional flax/epoxy sample at 20 °C. The results in Fig. 5 link to a permittivity ε'_r of 5.1. The

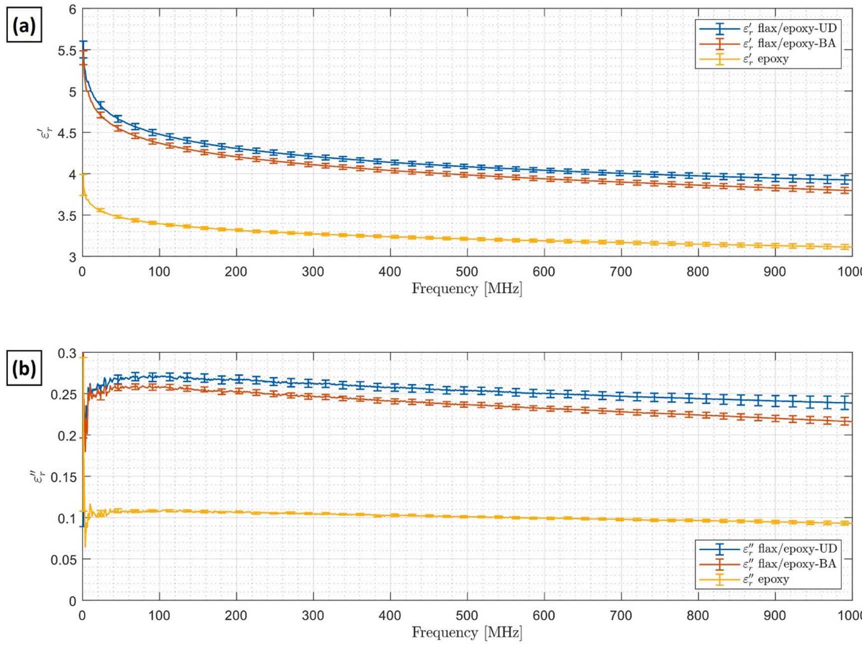

Fig. 5. Real part (a) and imaginary part (b) of the permittivity of unidirectional and biaxial flax/epoxy composites and epoxy in the frequency range between 1 MHz and 1 GHz.

readings in Fig. 5 tie in at a frequency of 20 MHz with a permittivity ε'_r of 4.8 of identical magnitude. It can be clearly seen that there is a large scatter in the measured values for both the real and imaginary parts of the permittivity at a frequency of 1 MHz. The results in this frequency range must therefore be viewed critically, since the measuring instrument does not get them resolved cleanly.

The imaginary part of the permittivity is shown Fig. 5(b). The frequency dependence and the difference between unidirectional and biaxial flax/epoxy fabrics can also be seen here. Only the increasing trend of epoxy is no longer evident in the frequency range between 1 MHz and 1 GHz, which has already been shown in the past [39,44]. The reason for this is the β relaxation process, which comes into play at high frequencies and low temperatures [45]. The magnitude of the imaginary parts shown agrees with those in Fig. 4.

It was shown that the fiber orientation of a flax/epoxy sample produced by the vacuum infusion process has an influence on the complex permittivity due to the fiber volume fraction. The higher the fiber volume fraction, the greater the complex permittivity due to the increasing influence of the flax fiber compared to the matrix. This behavior as a function of fiber orientation can be supported by the studies on flax/epoxy composites [15] and bamboo fiber and nanoclay-reinforced epoxy [30], as it was shown that the dielectric properties increase with fiber volume fraction. The frequency dependence of the two measured components follows the characteristic course of the permittivity or susceptibility, which is additively composed of the various mechanisms of electrical polarization (displacement polarization, orientation polarization, ion polarization, etc.), which in turn depend on frequency. Therefore, as expected, all $\underline{\varepsilon_r}$ values decrease (continuously) with increasing frequency. Local maxima in the imaginary part cannot be identified so far. This is to be considered in future investigations over an extended frequency range.

A comparison with measured values from the literature in this frequency range is only possible to a limited extent, since corresponding values are only available up to 1 MHz. Table 1 shows the quantitative results of the complex relative permittivity of unidirectional flax/epoxy composites at a frequency of 200 kHz from the literature. This is intended to place the results presented in this paper in the context of previous research. A direct comparison with the results in the frequency range between 1 MHz and 1 GHz is not possible, since measured values from the literature are limited to 1 MHz.

Table 1
Overview relative permittivity of unidirectional flax/epoxy composites at 200 kHz.

Fiber volume fraction [%]	ε'	ε''	Manufacturing process	Temp. [°C]	Reference
50	2	0.07	prepregs	40	[9]
50	10	0.6	prepregs	40	[10]
30	4	0.08	hot-pressing	20	[15]
45	4.2	0.126	hot-pressing	20	[15]
60	4.4	0.176	hot-pressing	20	[15]

The measured values of both the real and the imaginary part of the relative permittivity fluctuate. The results of the real part from [10] are particularly striking. Fig. 4 shows a real permittivity ε'_r of 5.1 and a complex permittivity ε''_r of 0.18 for the unidirectional flax/epoxy sample at a temperature of 20 °C and a frequency of 200 kHz. Since the fiber volume fraction is 45 %, the results presented here can be compared with those from [15]. It can be clearly seen that the values are in the same order of magnitude and differ only slightly. Thus, it can also be concluded that the manufacturing method has at most a minor influence on the dielectric properties. If there are increased air inclusions in a manufacturing method due to the manufacturing process, the permittivity will be lower compared to the results shown here. Based on the results from [15], it can also be seen that the complex permittivity increases with increasing fiber volume fraction. This confirms the measured differences between the unidirectional and biaxial samples due to their different fiber volume fraction. Only the already mentioned results from [10] do not fit into the order of magnitude of the key figures presented here. A comparison with measured values in the temperature range below 20 °C is not possible, since no values exist for this in the literature.

4. Conclusion

The aim of this publication was to show for the first time the complex relative permittivity of unidirectional and biaxial flax/epoxy composites in the temperature range between − 20 °C and 50 °C for the frequency range between 1 kHz and 200 kHz. In addition, the complex permittivity in the frequency range between 1 MHz and 1 GHz was to be mapped for the first time for the plant fiber composite. In this work, the samples prepared by vacuum infusion method were investigated using a self-developed parallel-plate capacitor and an impedance/material analyzer.

For a more detailed description of the two flax/epoxy samples, a micro-computed tomography and an SEM image were also obtained. These show that local differences occur in the relationship between fiber and matrix and that these must be considered when using small measuring heads to determine permittivity. Another result of the microstructure analysis was that the matrix penetrates the individual flax fibers and thus slightly reduces the fiber volume fraction. Based on the images shown, it can be ruled out that air inclusions occurred during the manufacture of the samples, which would falsify the result.

It was shown that the unidirectionally fabricated flax/epoxy composite has a larger complex relative permittivity over the entire frequency range than the biaxial scrim due to its higher fiber volume fraction. The ratio between fiber and matrix material due to manufacturing is responsible for this. The resin used has a lower relative permittivity than the flax fiber. Thus, the fiber volume fraction plays a crucial role in the magnitude of the complex relative permittivity. In addition, the complex relative permittivity of a plant fiber composite was investigated for the first time in the temperature range between − 20 °C and 20 °C, which is relevant for the new sensing element. This shows that the behavior is analogous to the temperature ranges already investigated from room temperature. This means that with decreasing temperature, the relative permittivity in the frequency range between 1 kHz and 200 kHz also decreases further above zero. At the frequency of 200 kHz, a permittivity ε'_r of 5.1 was found at 20 °C, whereas at − 20 °C the permittivity ε'_r already dropped to 4.05. This is due to the decreasing conductivity of the composite over the temperature.

The presented results of the complex relative permittivity also show a characteristic behavior for natural fiber composites in the frequency range up to 1 GHz. At the frequency of 1 GHz and room temperature, a permittivity of ε'_r of 3.9 and ε''_r of 0.24 could be measured for the unidirectional sample and a permittivity of ε'_r of 3.8 and ε''_r of 0.215 for the biaxial sample. This means that the order of magnitude of the measured values is comparable with previously published results, which, however, were considered at most up to 1 MHz. Since only other manufacturing processes were used in these, it can also be stated that the manufacturing process has only an indirect influence on the dielectric properties. As already mentioned, the fiber volume fraction is more decisive. With the help of the knowledge gained, it will be possible in the future to design and use impedance-controlled RF-lines structures as sensor elements for flax/epoxy composites using high-frequency alternating currents. In addition, the use of the natural fiber material is made possible in many areas of application in electrical engineering.

In future work, attention should be paid to a finer breakdown of the orientation of the clutches with respect to each other, considering the fiber volume fraction. Here, it is necessary to identify non-linear correlations of the mentioned factors on the complex relative permittivity. Of course, the frequency range must also be increased in the future and the frequency dependence of the individual components must be investigated.

CRediT authorship contribution statement

Philipp Baron: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data curation, Writing – original draft, Visualization. **Philipp Lenz:** Methodology, Software, Validation, Formal analysis, Writing – review & editing. **Klaus Peter Koch:** Validation, Writing – review & editing, Funding acquisition. **Armin Wittmann:** Resources, Writing – review & editing, Funding acquisition. **Georg Fischer:** Resources, Writing – review & editing, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The data used are available in the supplemental materials.

Acknowledgements

The project “Intelligent Composite Materials” is funded by the Carl Zeiss Foundation. The authors also thank Stefan Trapp, Marie Kandels, Jie Zou, Alexander Spielberger, Christoph Zimmer, and Lukas Herrmann for their support.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.mtcomm.2023.106656.

References

[1] C.I. Madueke, O.M. Mbah, R. Umunakwe, A review on the limitations of natural fibres and natural fibre composites with emphasis on tensile strength using coir as a case study, Polym. Bull. (2022) 1–18, https://doi.org/10.1007/s00289-022-04241-y.

[2] K.L. Pickering, M.A. Efendy, T.M. Le, A review of recent developments in natural fibre composites and their mechanical performance, Compos. Part A Appl. Sci. Manuf. 83 (2016) 98–112, https://doi.org/10.1016/j.compositesa.2015.08.038.

[3] K. Lau, P. Hung, M.-H. Zhu, D. Hui, Properties of natural fibre composites for structural engineering applications, Compos. Part B Eng. 136 (2018) 222–233, https://doi.org/10.1016/j.compositesb.2017.10.038.

[4] M. Alkbir, S.M. Sapuan, A.A. Nuraini, M.R. Ishak, Fibre properties and crashworthiness parameters of natural fibre-reinforced composite structure: a literature review, Compos. Struct. 148 (2016) 59–73, https://doi.org/10.1016/j.compstruct.2016.01.098.

[5] G. Seychal, E. Ramasso, P. Le Moal, G. Bourbon, X. Gabrion, V. Placet, Towards in-situ acoustic emission-based health monitoring in bio-based composites structures: does embedment of sensors affect the mechanical behaviour of flax/epoxy laminates, Compos. Part B Eng. 236 (2022), 109787, https://doi.org/10.1016/j.compositesb.2022.109787.

[6] P. Lenz, P. Baron, A. Wittmann, G. Fischer, Wear-induced attenuation on transmission lines and their causes. Transactions on Electrical and Electronic Materials, Springer, 2022, https://doi.org/10.1007/s42341-022-00423-7.

[7] J. Hu, S. Yin, T.X. Yu, J. Xu, Dynamic compressive behavior of woven flax-epoxy-laminated composites, Int. J. Impact Eng. 117 (2018) 63–74, https://doi.org/10.1016/j.ijimpeng.2018.03.004.

[8] M. Kersani, S.V. Lomov, A.W. van Vuure, A. Bouabdallah, I. Verpoest, Damage in flax/epoxy quasi-unidirectional woven laminates under quasi-static tension, J. Compos. Mater. 49 (2015) 403–413, https://doi.org/10.1177/0021998313519282.

[9] M. Karray, A. Triki, C. Poilâne, P. Picart, M. Gargouri, Dielectric relaxation phenomena in flax fibers composite, Fibers Polym. 17 (2016) 88–96, https://doi.org/10.1007/s12221-016-5552-6.

[10] A. Triki, M. Karray, C. Poilâne, P. Picart, M. Gargouri, Dielectric analysis of the interfacial polarization of alkali treated woven flax fibers reinforced epoxy composites, J. Electrostat. 76 (2015) 67–72, https://doi.org/10.1016/j.elstat.2015.05.017.

[11] A.N. Fraga, E. Frullloni, O. de La Osa, J.M. Kenny, A. Vázquez, Relationship between water absorption and dielectric behaviour of natural fibre composite materials, Polym. Test. 25 (2006) 181–187, https://doi.org/10.1016/j.polymertesting.2005.11.002.

[12] I. Ben Amor, H. Rekik, H. Kaddami, M. Raihane, M. Arous, A. Kallel, Effect of palm tree fiber orientation on electrical properties of palm tree fiber-reinforced polyester composites, J. Compos. Mater. 44 (2010) 1553–1568, https://doi.org/10.1177/0021998309353961.

[13] L. Restuccia, G.A. Kluitenberg, On generalizations of the Debye equation for dielectric relaxation, Phys. A Stat. Mech. Appl. 154 (1988) 157–182, https://doi.org/10.1016/0378-4371(88)90186-0.

[14] E. Talebian, M. Talebian, A general review on the derivation of Clausius–Mossotti relation, Optik 124 (2013) 2324–2326, https://doi.org/10.1016/j.ijleo.2012.06.090.

[15] S. Zhang, Y. Li, Z. Zheng, Effect of physiochemical structure on energy absorption properties of plant fibers reinforced composites: dielectric, thermal insulation, and sound absorption properties, Compos. Commun. 10 (2018) 163–167, https://doi.org/10.1016/j.coco.2018.09.006.

[16] H. Prashanth PVS, E. Jayamani, K.H. Soon, Investigation of electric properties of flax reinforced polypropylene/strontium titanate composite for multilayer dielectric applications, IOP Conf. Ser. Mater. Sci. Eng. 1136 (2021) 12008, https://doi.org/10.1088/1757-899X/1136/1/012008.
[17] E. Jayamani, S. Hamdan, M.R. Rahman, M.K.B. Bakri, Dielectric properties of lignocellulosic fibers reinforced polymer composites: effect of fiber loading and alkaline treatment, Mater. Today Proc. 2 (2015) 2757–2766, https://doi.org/10.1016/j.matpr.2015.07.269.
[18] E. Jayamani, S. Hamdan, M.R. Rahman, M.K.B. Bakri, Comparative study of dielectric properties of hybrid natural fiber composites, Procedia Eng. 97 (2014) 536–544, https://doi.org/10.1016/j.proeng.2014.12.280.
[19] E. Jayamani, G. Anil Nair, K. Soon, Investigation of the dielectric properties of natural fibre and conductive filler reinforced polymer composites, Mater. Today Proc. 22 (2020) 162–171, https://doi.org/10.1016/j.matpr.2019.08.032.
[20] E. Jayamani, S. Hamdan, P. Ezhumalai, M.K. Bakri, Investigation on dielectric and sound absorption properties of banana fibers reinforced epoxy composites, J. Teknol. 78 (2016), https://doi.org/10.11113/jt.v78.9195.
[21] T. Jackson Singh, S. Samanta, H. Singh, Influence of Kevlar hybridization on dielectric and conductivity of bamboo fiber reinforced epoxy composite, J. Nat. Fibers 14 (2017) 837–845, https://doi.org/10.1080/15440478.2017.1279583.
[22] C. Dash, D.K. Bisoyi, Significance of dielectric and mechanical properties on fiber load-ing of sunn hemp reinforced epoxy composite. In: Proceedings of the Third International Conference on Condensed Matter and Applied Physics (ICC-2019), Bikaner, India, AIP Publishing, 2020,80027.
[23] D. Pathania, D. Singh, A review on electrical properties of fiber reinforced polymer composites, Int. J. Theor. Appl. Sci. (2009) 34–37.
[24] E. Rodríguez, R. Petrucci, D. Puglia, J.M. Kenny, A. Vázquez, Characterization of COmposites Based on Natural and Glass Fibers Obtained by Vacuum Infusion, J. Compos. Mater. 39 (2005) 265–282, https://doi.org/10.1177/0021998305046450.
[25] Bcomp Ltd., ampliTex Art. No. 5025. https://pdf.nauticexpo.com/pdf/bcomp-ltd/amplitex-5025/51016–100536.html.
[26] Cloeren Technology GmbH, CEM100 Blue. https://www.cloeren.de/metallographie-shop/kalteinbettmittel-cem1000-blue-set.
[27] M.E. Messiry, Theoretical analysis of natural fiber volume fraction of reinforced composites, Alex. Eng. J. 52 (2013) 301–306, https://doi.org/10.1016/j.aej.2013.01.006.
[28] JEOL Ltd., JSM-6610 Series Scanning Electron. https://www.jeol.com/products/scientific/sem/JSM-6610series.php.
[29] DIN Deutsches Institut für Normung e. V., DIN EN 62631–2-1: Dielektrische und resistive Eigenschaften fester Elektroisolierstoffe.
[30] Rohde & Schwarz, HM8118 LCR Bridge/Meter. https://www.rohde-schwarz.com/de/produkte/messtechnik/lcr-meter-und-impedanzanalysatoren/rs-hm8118-lcr-messbruecken-messer_63493–44101.html.
[31] J.S. Bobowski, A.P. Clements, Permittivity and conductivity measured using a novel toroidal split-ring resonator, IEEE Trans. Microw. Theory Technol. 65 (2017) 2132–2138, https://doi.org/10.1109/TMTT.2016.2645147.
[32] Binder GmbH, Model MK 115. https://www.binder-world.com/int-en/product/mk-115.
[33] Keysight, E4991A Impedance/Material Analyzer. https://www.keysight.com/de/de/product/E4991A/rf-impedancematerial-analyzer.html.
[34] Keysight, 16453A Dielectric Material Test Fixture. https://www.keysight.com/de/de/product/16453A/dielectric-material-test-fixture.html.
[35] I.B. Amor, Z. Ghallabi, H. Kaddami, M. Raihane, M. Arous, A. Kallel, Experimental study of relaxation process in unidirectional (epoxy/palm tree fiber) composite, J. Mol. Liq. 154 (2010) 61–68, https://doi.org/10.1016/j.molliq.2010.04.006.
[36] V. Kumar, P.K. Kushwaha, R. Kumar, Impedance-spectroscopy analysis of oriented and mercerized bamboo fiber-reinforced epoxy composite, J. Mater. Sci. 46 (2011) 3445–3451, https://doi.org/10.1007/s10853-011-5249-6.
[37] A. Triki, M. Guicha, M. Ben Hassen, M. Arous, Comparative study of the dielectric properties of natural-fiber-matrix composites and E-glass-matrix composites, J. Appl. Polym. Sci. 129 (2013) 487–498, https://doi.org/10.1002/app.38499.
[38] M. Arous, I.B. Amor, S. Boufi, A. Kallel, Experimental study on dielectric relaxation in alfa fiber reinforced epoxy composites, J. Appl. Polym. Sci. 106 (2007) 3631–3640, https://doi.org/10.1002/app.26885.
[39] M.A. Omri, M.R. Sanjay, A. Triki, B. Yogesha, A. Kallel, Dielectric properties and interfacial adhesion of jute, kenaf and E-glass fabrics reinforcing epoxy composites, Polym. Compos. 40 (2019) 2142–2153, https://doi.org/10.1002/pc.25001.
[40] M.M. Adnan, I.-E. Nylund, A. Jaworski, S. Hvidsten, M.-H.G. Ese, J. Glaum, M.-A. Einarsrud, The structure, morphology, and complex permittivity of epoxy nanodielectrics with in situ synthesized surface-functionalized SiO2, Polymers 13 (2021), https://doi.org/10.3390/polym13091469.
[41] T.T.M. Phan, N.C. Chu, B. van Luu, H. Nguyen Xuan, D.T. Pham, I. Martin, P. Carrière, Enhancement of polarization property of silane-modified BaTiO 3 nanoparticles and its effect in increasing dielectric property of epoxy/BaTiO 3 nanocomposites, J. Sci. Adv. Mater. Devices 1 (2016) 90–97, https://doi.org/10.1016/j.jsamd.2016.04.005.
[42] Z. Wang, G. Meng, L. Wang, L. Tian, S. Chen, G. Wu, B. Kong, Y. Cheng, Simultaneously enhanced dielectric properties and through-plane thermal conductivity of epoxy composites with alumina and boron nitride nanosheets, Sci. Rep. 11 (2021) 2495, https://doi.org/10.1038/s41598-021-81925-x.
[43] S. Singha, M. Thomas, Permittivity and tan delta characteristics of epoxy nanocomposites in the frequency range of 1 MHz-1 GHz, IEEE Trans. Dielectr. Electr. Insul. 15 (2008) 2–11, https://doi.org/10.1109/T-DEI.2008.4446731.
[44] N. Hasan, N.S.M. Hussain, A.A.M. Faudzi, S.M. Shaharum, N.A.T. Yusof, N. H. Noordin, N.A.A. Mohtadzar, M.S.A. Karim, Cured epoxy resin dielectric characterization based on accurate waveguide technique. Applied Physics of Condensed Matter (APCOM 2019), Strbske Pleso, Slovak Republic, AIP Publishing, 2019, p. 20080.
[45] M.K. Hassan, S.J. Tucker, A. Abukmail, J.S. Wiggins, K.A. Mauritz, Polymer chain dynamics in epoxy based composites as investigated by broadband dielectric spectroscopy, Arab. J. Chem. 9 (2016) 305–315, https://doi.org/10.1016/j.arabjc.2015.07.016.