

BIOLOGICAL AND ELECTRICAL RESISTANCE OF ACETYLATED FLAX FIBRE REINFORCED POLYPROPYLENE COMPOSITES

Andrzej Korneliusz Bledzki,^a Marta Lucka,^a Abdullah Al Mamun,*^a and Janusz Michalski^b

Flax fibre reinforced polypropylene composites were fabricated using a high speed mixer followed by injection moulding. Prior to composite production, the fibre was modified by acetylation in the presence of perchloric acid. The effect of acetylation of the fibre was assessed on the basis of moisture resistance and dielectric properties of the resulting composites. It was found that the moisture absorption and swelling properties of the composites were reduced respectively by 60% and 30% due to acetylation. Two different types of biocide were mixed with untreated flax fibre, and the samples were exposed to decay fungi for up to 3 months along with control polypropylene. The composites with acetylated fibres showed good protection against fungi, and biocide had less effect on biological resistance. The dielectric properties of the flax-polypropylene composites were also estimated as a function of aging period. The composites with modified fibre showed more improvement in dielectric properties compared to the composites with untreated fibres. The mechanical properties were investigated for those composites. Tensile and flexural strengths of composites were found to be increased following acetylation due to modification, and strength properties of both untreated and acetylated flax fibre reinforced polypropylene composites decreased with respect to aging period. The Charpy impact strengths of composites were found to increase with increasing aging periods.

Keywords: Flax fibre; Acetylation; Adhesion; Swelling; Biological resistance; Dielectric properties; Mechanical properties

Contact information: a: Institute für Werkstofftechnik, University of Kassel, Mönchebergstraße 3, 34125 Kassel, Germany; b: Department of Electrical Engineering, Technical University of Szczecin, 70-313 Szczecin, Poland; *Corresponding author: kutech@uni-kassel.de

INTRODUCTION

The utilization of biomass has gained increased importance due to the threats of uncertain petroleum supply and concerns about environmental pollution. Green, ecologically friendly, sustainable, renewable, biodegradable composites from plant-derived fibre and crop-derived plastic polymers are the most keenly required materials for the twenty first century (Mohanty et al. 2002; Bledzki et al. 2002; Nishino 2004; Bledzki et al. 2007). However, natural fibres typically exhibit a high hydrophilicity due to affinity or interaction between the hydroxyl groups of fibre components and water molecules. The high moisture sensitivity of lignocellulosic fibre has the potential to cause dimensional instability, and this phenomenon limits the use of such fibre as a reinforcement in composite materials. Low interfacial bonding between a natural fibre and polymer matrix

often reduces its potential as a reinforcing agent due to its hydrophilic nature; chemical modifications are considered to optimize the interface (Bledzki et al. 1999, 2008).

The diffusion of water into fibres apparently decreases the biological resistance and dielectric properties of the composites. Fungi need water to break down the cell wall of plant fibres. It has been found that the moisture content of natural fibre (i.e. wood fibre) needs to be above the saturation point for decay to occur (Carll et al. 1999). By chemically modifying wood, it can still be exposed to water or wet conditions, but it will sorb little cell wall water. Previous research has shown a decrease in the equilibrium moisture content and an increase in fungal resistance of southern pine solid wood in cases where fibre had been chemically modified by acetic anhydride or butylene oxide, indicating a mechanism of efficacy by lowering the moisture content of wood below the level required for microorganism attack. By contrast, fibre modified by propylene oxide did not lower the equilibrium moisture content, yet it still provided some fungal resistance in soil block testing. The results suggested a substrate modification mechanism, thus rendering the wood as a non-food source (Ibach et al. 2002; Clemons et al. 2004).

Enhancement of biological resistance of different natural fibres by chemical modification is believed to result from alteration of the chemical structure of the cell wall polymers, making them protectable from microbial decay enzymes. It was also reported that acetylated cellulose fibres exhibited good resistance to brown rot, white rot, and soft rot fungal decay (Timer et al. 1999; Rowell 2005). In an innovative study, the biodegradability of protein-filled polymer composites using dielectric measurements was investigated by Tchmutin et al. (2004). The biodegradation experiment was conducted in an aqueous phase of malt extract medium by an enzyme, *Aspergillus oryzae*, for 3 weeks. The authors observed that the biodegradation coefficients determined using dielectric measurements were in good agreement with the weight loss of the composites (Tchmutina et al. 2004).

During the last couple of years natural fibre composites have received considerable attention in the automotive, packaging, and construction industries. The uses of natural fibre composites are expanding day by day, opening the new application windows. In recent years, low cost natural fibre composites proved of interest for dielectric applications, showing also some potential for future application as dielectric materials in microchips, parts of transformers, and circuit boards. These materials can also be easily employed for multi-function applications, as the hollow cellular structure of plant fibres proved effective in providing insulation against heat and noise (Hong et al. 2004). Therefore, studies on the electrical properties of natural fibre-reinforced thermoplastic composites are very important. Dry cellulose is a good insulator, having a specific direct current resistivity of about 10^{18} Ω cm, and has been used in fibre or sheet form for this purpose, for example as insulating paper in electrical condensers. The dielectric properties decrease significantly with increasing water content and increasing content of ions or ionic sites (Klemm et al. 1998; Wang et al. 2008).

Studies involving dielectric properties of waste paper and newsprint fibre-reinforced natural rubber composites have also been reported. Electrical properties of waste paper and newsprint improved due to modification with sodium silicate and magnesium chloride (Agrawal et al 2004; Nashar et al. 2004).

In an interesting study, the dielectric properties of sisal and oil palm hybrid bio-fibre reinforced natural rubber bio composites was investigated by Jacob et al. (2006). The dielectric constant was seen to increase with fibre loading. This was attributed to the increase in orientation polarization of the polar groups present in lignocellulosic fibres. The authors also observed that volume resistivity of composites increased upon chemical modification. This implies that incorporation of lignocellulosic fibres can increase the conductivity of composites (Jacob et al. 2006).

Hong developed a new low dielectric constant material suited to electronic materials applications, using hollow keratin fibres and chemically modified soybean oil. The unusual low value of dielectric constant obtained was due to the air present in the hollow microcrystalline keratin fibres and the triglyceride molecules. It has been expected that the low-cost composite made from avian sources and plant oil has the potential to replace the dielectrics in microchips and circuit boards in the ever-growing electronics materials field (Hong et al. 2004).

Pothan reported mechanical and electrical properties of modified banana fibre polyester composites. The dielectric constant values of the treated fibre composites were found to be lower than the untreated fibre composites (Pothan et al. 2007).

In this present article the effect of acetylation of flax fibre on the biological resistance, dielectrics, and mechanical properties of composites was investigated.

Theoretical Background

The dielectric constant is an essential piece of information when designing capacitors and in other circumstances where a material might be expected to introduce capacitance into a circuit. If a material with a high dielectric constant is placed in an electric field, the magnitude of that field will be measurably reduced within the volume of the dielectric. This fact is commonly used to increase the capacitance of a particular capacitor design. The dielectric constant or relative permittivity ϵ of a material is defined as the ratio of the capacitance of a condenser containing the material as dielectric to that of the same condenser in vacuum without a material dielectric. The capacitance of a condenser measures the extent to which it is able to store charges.

The dielectric constant ϵ can be calculated from the capacitance using the equation

$$\epsilon = Ct / \epsilon_0 A , \quad (1)$$

where C is the capacitance of the material, t is the thickness of the sample, ϵ_0 is the permittivity of free space ($8.85 \times 10^{-12} \text{ C}^2/\text{Nm}^2$), and A is the area of sample under electrode. For a specific sample, the dielectric constant varies in direct proportion with the capacitance.

The volume resistivity is the resistance to leakage current through the body of an insulating material. It is define as the electrical resistance between opposite faces of a unit volume of insulating material, commonly expressed in ohm-centimeters. The ratio of the potential gradient is parallel to the current in a material to the current density. The insulation resistance of a material depends on its volume resistance; thus the volume resistivity (ρ) can be calculated by using the equation

$$\rho = RA / t , \quad (2)$$

where R is the volume resistance (Ω), A is the area of cross-section, and t is the thickness of the samples. Measurements of volume resistance of flax fibre reinforced composites are often complicated by the rectification and contact resistance at the boundary between the composites and the electrode. To overcome the problem, the volume conductivity was determined by measurement of the damping in a high frequency resonant circuit in which the composites material is the dielectric in the condenser of the LC circuit.

The ratio of the imaginary to the real dielectric loss angle constants $\varepsilon''/\varepsilon'$, or the tangent of the dielectric loss angle is commonly employed as a direct measure of the dielectric loss. It is also known as the dissipation factor and is a measure of the power dissipated. The dissipation factor $\tan \delta$ can be calculated from the equation

$$\tan \delta = \varepsilon'' / \varepsilon' , \quad (3)$$

where ε'' is the loss factor and ε' is the dielectric constant.

EXPERIMENTAL

Materials

Green flax fibre (6-8 mm) was obtained from Mühlmeier GmbH, Germany. Two different types of biocide have been used for this investigation,

The water-dispersion biocide SRP 21-06 was obtained from Clariant International Ltd, Muttenz, Switzerland. Flax fibres were treated with 0.5 wt % of biocide. Solid biocide was obtained from Ciba Specialist Chemie GmbH, Lampertheim, Germany. 0.5 wt % biocide was taken in the agglomeration process. Both biocides are commonly prepared at laboratory scale and used for bio resistance of textile fibre; they are temperature sensitive.

Acetic anhydride (analytical grade) was obtained from Merck KGaA, Darmstadt, Germany.

Polypropylene (Sabic PP 575P) was provided as granules by Sabic Deutschland GmbH & Co.KG, Duesseldorf, Germany. Its melting temperature was 173°C; melting index was 10.5 g/10 min at 230°C; density at room temperature was 0.905 g/cm³; stress at break was 34 MPa; Strain at break was 600%; and flexural modulus was 1.8 GPa.

Acetylation Process

The flax fibres were soaked in demineralised water for an hour, filtered, and placed in a round-bottom flask containing acetylating solution. Acetylating solution consisted of 250 ml toluene, 125 ml acetic anhydride, and a small amount of the catalyst perchloric acid (60%). The process temperature of acetylation was 60°C, and the duration was 1 to 3 hour. After modification the fibre was washed periodically with distilled water until it was acid-free. Finally modified flax fibres were air dried for a certain time before investigation. Our previous paper reported that composites from modified flax with

degree of acetylation of 18 % had better thermal and mechanical properties. Therefore, flax fibre with a degree of acetylation of 18% was used for this investigation (Bledzki et al. 2008).

Processing of Composites by Mixer-Injection Moulding Methods

Treated or untreated flax fibres with polypropylene were mixed with a high speed cascade mixer (Henschel heat-cooling mixer system, type HM40-KM120). Flax fibres were dried at 80°C in an air circulating oven for 24 hours (moisture content <1%) before mixing. The flax fibre at 30 wt% proportion and polypropylene was placed into a hot mixer and heated to the melting temperature of polypropylene (173°C), and then hot agglomerate granules were transferred to the cool mixer, where hot agglomerate granules were cooled down to room temperature by the cold water. Then, cold agglomerate granules were dried again (80°C, 24 hours) before the sample preparation by an injection-moulding process. Test samples were prepared from dried agglomerate by injection-moulding process within the temperature zone 150°C-180°C, at a mould temperature of 80°C with an injection pressure of 20 kN/mm².

Characterization of Composites

Water absorption

Water absorption studies were performed following the EN ISO 62 standard test method for water absorption of plastics. Five specimens of tensile and flexural from every batch were submerged in distilled water at 40°C. The specimens were removed from the water after certain periods of time, weighed with a high precision balance to find the amount of water taken up, and then resubmerged in water.

Biological tests

A laboratory decay test was undertaken to assess biological resistance of the modified and non-modified flax fibre composites. All samples, previously oven dried at 105°C to constant weight, were sterilised with radiation and exposed at a relative humidity of 95% and of temperature 23°C for 90days prior to the test. Samples were supported on sterile plastic mesh to prevent contact with the agar and incubated. After this period, hyphae adhering to sample surfaces were removed and the samples were dried at 105°C to constant weight. The weight losses (*WL*) for individual samples and mean percentage weight losses for each sample set were determined using Equation (4),

$$WL = 100 * (M_0 - M_f) / M_0 \quad (4)$$

where M_0 is mass of oven-dried sample prior to the test, and M_f is mass of oven-dried sample after the test.

Fungal colonisation and decay of experimental and reference (control) surface was also examined using microscopy.

Microscope

The morphology of treated and untreated flax fibre composites were investigated using a light microscope, Carl Zeiss, Axioplan, Germany. Samples were fixed on a metal surface before investigation.

Dielectric properties

Samples were prepared by cutting from the composite specimens using a die. Rectangular specimens of 2 mm thickness were used. The test samples were coated with silver paste on either side, and copper wires were fixed on both sides of the samples as electrodes. The capacitance, resistance, and dielectric loss factor were measured directly at room temperature, using an impedance analyzer by constant frequency. These properties were estimated according to Polish Standard PN-88 / E-04405.

Mechanical properties

Tensile, flexural, and impact tests were carried out for characterization of composites. Tensile and flexural tests were performed at a test speed of 2 mm/min according to EN ISO 527 and EN ISO 178, using a Zwick UPM 1446 machine. All tests were performed at room temperature (23°C) and at a relative humidity of 50%. The Charpy impact test was carried out using 10 notched samples according to EN ISO 179, using a Zwick Charpy impact machine. In each case a standard deviation < 5% (drop weight) was used to calculate the Charpy impact strength.

RESULTS AND DISCUSSION

Water Absorption and Swelling Properties

The water absorption and swelling properties of treated and untreated flax fibre composites that were submerged at 40°C are illustrated in Fig. 1. It was observed that the moisture absorption increased with increased aging periods for both cases. The water uptake for both composites was found to be equilibrium after 100 days of aging periods. The equilibrium moisture content of treated flax composites decreased about 60% compared to untreated fibre composites. These effects were attributed to acetylation, and are most likely due to reduction of hydrophobicity of fibre.

The swelling properties of treated and untreated flax fibre composites also increased with the increased aging periods. The swelling properties of treated and untreated flax composites increased rapidly with increased aging periods till 20 days, and after that the swelling properties increased slowly with increased aging periods. It was also found that the swelling property seemed to approach an equilibrium state after 100 days of aging periods. The swelling properties decreased about 30% due to modification, which is likely associated with a reduction of water uptake.

Biological Resistance

Decay fungi obtain nourishment by digesting plant cell walls, thus causing deterioration of the fibre. Fungal hyphae secrete extracellular enzymes and other agents that depolymerise cell wall materials; these depolymerised materials are then absorbed

into the fungal hyphae, where they are assimilated and further metabolized. Mold and stain fungi derive their food from materials stored in cell cavities or from nutrients on the fibre surface and have little influence on the strength of fibre. Mold and stain fungi primarily colonize sapwood plant fibre. They differ, however, in that mold fungi have colourless hyphae, while stain fungi have pigmented hyphae that may cause a stain throughout the affected sap plant fibre. The life cycle of a fungus consists of a vegetative phase and a fruiting phase. Plant fibre becomes infected either by spores produced during the fruiting phase, which under favourable conditions germinate on the fibre surface and produce filaments called hyphae that invade the fibre, or by spread of hyphae (collectively known as mycelium) from a source of previous infection (Zabel et al. 1992).

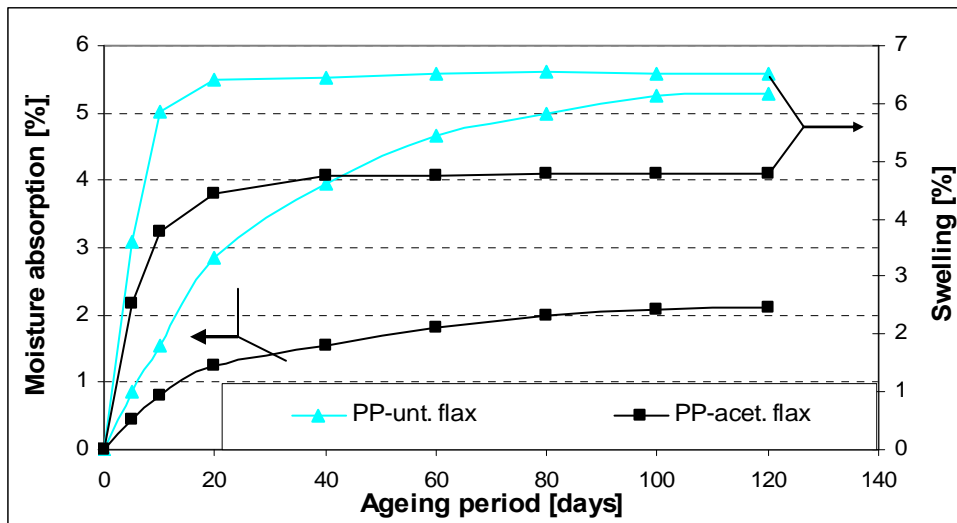
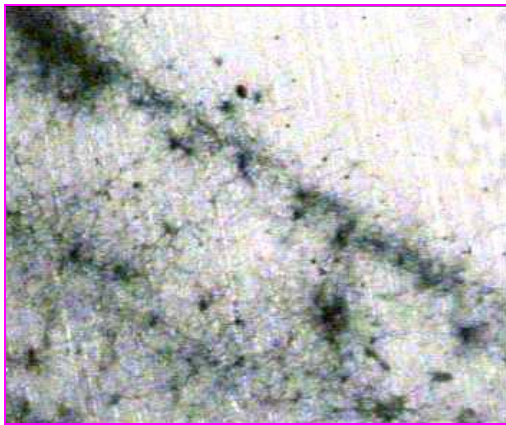


Fig. 1. Influence of aging period on the moisture absorption and swelling of flax composites

Decay fungi are classified as *brown rots*, *white rots*, or *soft rots*. Brown-rot and white-rot fungi are principally *Basidiomycete* fungi, and under favourable conditions they can rapidly disintegrate plant substances. White-rot fungi cause plant fibre to become pale, eventually reducing it to a fibrous whitish mass. Brown-rot fungi cause the wood to darken, shrink, and break into cubicles that are easily crumbled. Brown-rot decay fungi preferentially attack plant fibre (Carll et al. 1999).

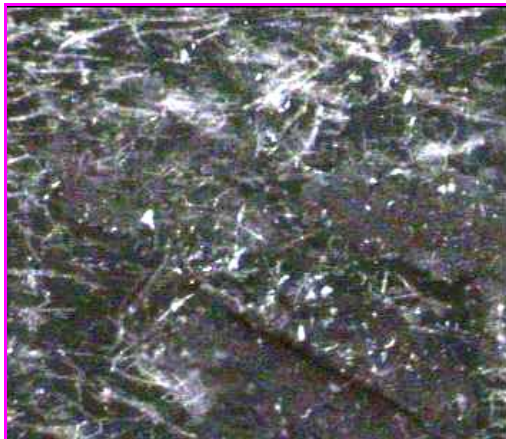
The effects of the fibre modification on the biological resistance of flax-polypropylene composites were compared to unmodified polypropylene and untreated flax polypropylene composites, and the results are shown in Fig. 2. It was observed that the composites with acetylated fibres exhibited a good resistance against micro fungi. It was observed that the attack of the *Aspergillus niger* was less than 10% on the sample surface and 8% weight losses were registered after 3-months of storage. On the other hand it was detected that more than 50% of the surface area of the untreated flax composites was covered by micro-fungi (some darkened and coloured fungi were also observed) and 41% weight losses were registered. Both types of biocide did not appear to reduce the growth of micro-organisms significantly. It was also shown that about 40% of biocide treated flax composites were attacked by micro-fungi, and weight losses of 33% were recorded. They may become deactivated during the composite processing.



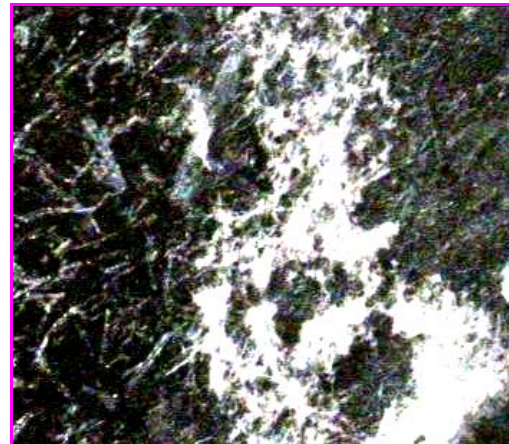
(2a)



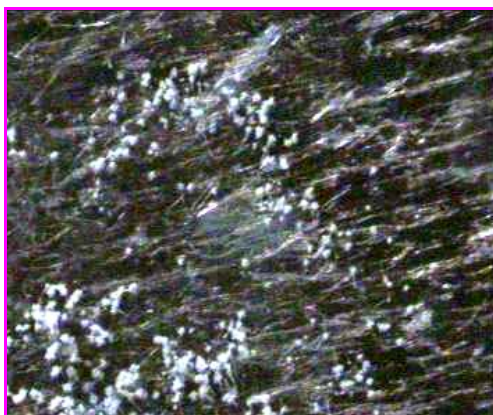
(2b)



(2c)



(2d)



(2e)

Fig. 2. Photomicrographs (magnification 150X) of the biological resistance of flax composites: (2a) control polypropylene, (2b) untreated flax composites, (2c) acetylated flax composites, (2d) biocide treated flax composites, and (2e) untreated flax-polypropylene/ biocide composites

Dielectric Properties

Dielectric constant

Figure 3 shows the influence of acetylation of flax fibres on the dielectric constant values of flax-reinforced polypropylene composites. At the initial stage it can be seen that the dielectric constants of fibre loaded composites were lower than those of the control polypropylene composites. The presence of natural fibre in composite materials increases the air content of a composite due to the hollow space in the middle of each fibre, termed the lumen. The air trapped in the lumen results in a lower dielectric value.

Another observation is that the dielectric constant of treated and untreated fibre composites increased with increased aging periods. This is because water molecule is very polar. It increases the surface polarity of the composite materials.

The acetylation modification of flax fibres composite resulted in lowering of dielectric constant. This is due to the decrease of orientation polarization of composites containing treated fibres. Acetylation treatment results in reduction of moisture absorption capacity of fibres due to the reduction in interaction between polar $-OH$ groups of flax fibres and water molecules. The resultant decrease of hydrophilicity of the fibres leads to lowering of orientation polarization and subsequently dielectric constant (Pothan et al. 2007; Pothan et al 2006).

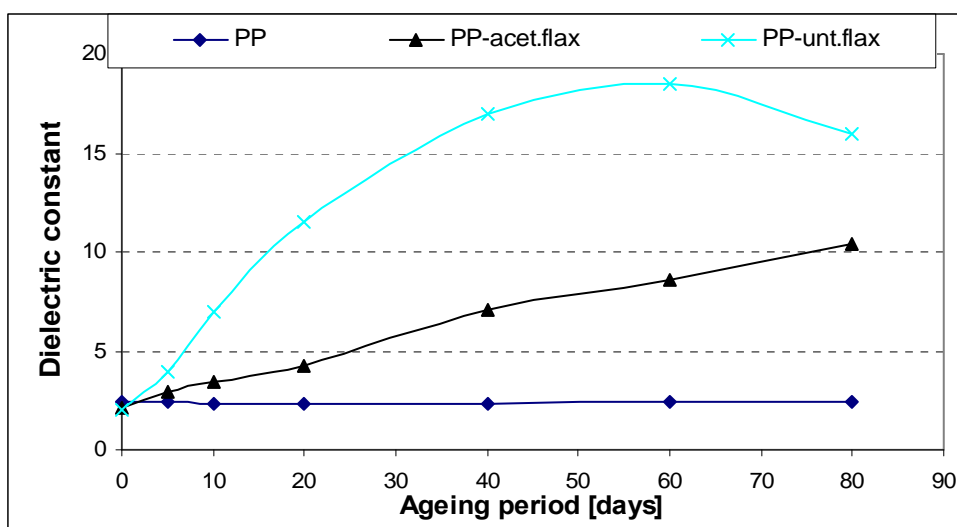


Fig. 3. Influence of aging period on the dielectric constant of flax composites

Specific volume resistivity

The study of the specific volume resistivity of an insulating material is important because the most desirable property of an insulator is its ability to resist the leakage of electric current. Figure 4 shows the variation of volume resistivity with aging period. An interesting observation is that volume resistivity decreased with fibre loading compared to the control polypropylene. This implies that the conductivity increased upon addition of lignocellulosic fibres. This is due to the presence of polar groups, which facilitate the flow of current. In polymers it is well known that most of the current flows through the crystalline regions, and the passage of current in the amorphous regions are due to the

presence of moisture. The presence of a lignocellulosic flax fibre increases the moisture content and hence increases the conductivity of the system. The specific volume resistivity reduced swiftly with increased aging periods in the initial stage and then decreased slowly with increased aging periods. The rapidly decreasing volume resistivity was associated with the water uptake. In comparison to untreated flax composites the volume resistivity of acetylated flax composites was found to increase with increasing aging period. At the initial aging period the changes in volume resistivity were similar to each other, but with increasing aging period the difference increased. As described earlier, chemical modification of fibres resulted in lowering of moisture content and increased interfacial adhesion, leading to increased resistivity values.

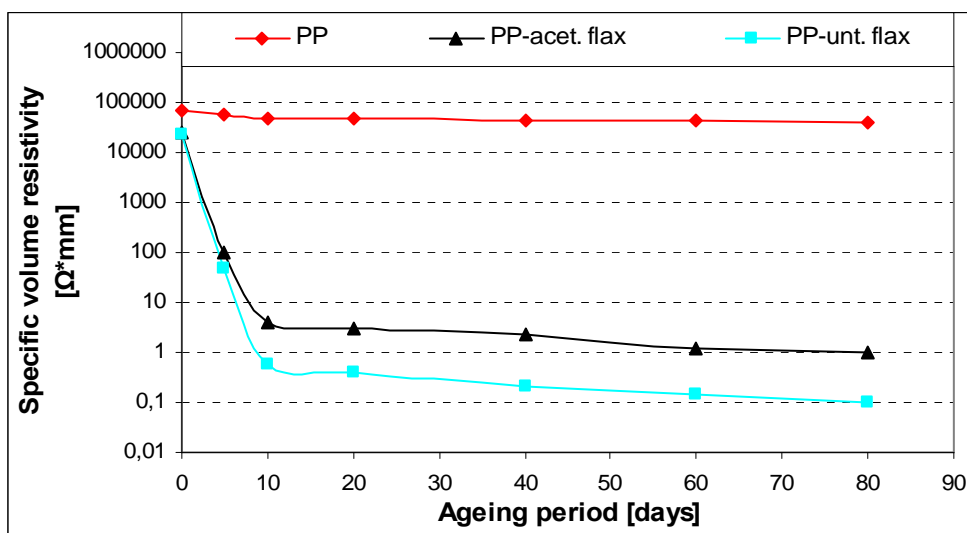


Fig. 4. Influence of aging period on the specific volume resistivity of flax composites

Dissipation factor

Dissipation factor or loss tangent is defined as the ratio of the electrical power dissipated in a material to the total power circulating in the circuit. The viscoelastic nature of the polymer creates responses in the material to both mechanical and electrical stimuli. The measurements of dissipation factor ($\tan \delta$) and loss factor of an insulating material are important, since the loss tangent is a measure of the electrical energy that is converted to heat in an insulator. The contrasting dissipation factors of treated and untreated flax fibre composites at different aging periods are presented in Fig. 5. It can be seen that the dissipation factor increased with aging period for both cases. This is because the presence of flax fibre facilitated more storage of electric current due to the presence of polar functional groups in the composites. During the initial aging period the dissipation factor of untreated flax composites increased rapidly, and after a certain period it decreased slowly. The rapidly increasing dissipation factor was associated with the water uptake. It can be said that the acetylation of flax does not affect the relaxation mechanism and after long exposure to moisture the acetylated fibre may be close to untreated fibre in polarity. This is because only 18% of functional groups of flax fibre

were chemically bonded by acetylation, while the rest functional groups would still absorb moisture like those of untreated fibre. Moreover, $\tan \delta$ is very sensitive to the changes of ϵ'' and ϵ' , especially in the case of the acetylated flax fibre.

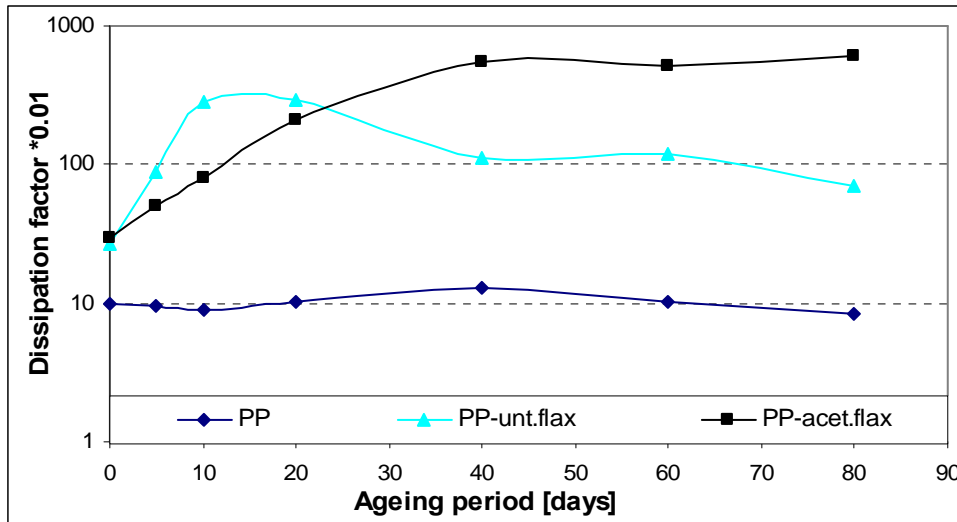


Fig. 5. Influence of aging period on the dissipation factor of flax composites

Mechanical Properties

Tensile strength

Figure 6 shows the tensile strength of flax composites as a function of aging period at 40°C. Tensile strength of composites was found to be decreased with increasing aging period for both cases. It was also found that initially the tensile strength decreased a little faster at the beginning, compared to later periods for both cases and after 40 days the tensile strength seemed to be constant with aging period. Initially the water absorption or diffusion kinetics was fast and the rates decreased gradually because the water molecule concentration increased with aging period in the composite materials. Compared to the initial value, the tensile strength decreased 5% and 20% for treated and untreated flax composites, respectively. The reduction of tensile properties is attributed to the diffusion of water molecules and the formation of a thin layer in between fibre and matrix, increasing the volume of materials. As a result of the initiation of micro-structural damage, micro-fungal action weakens the adhesion and causes macroscopic damage.

Flexural strength

Flexural strength is the ability of the material to withstand bending forces applied perpendicular to its longitudinal axis. The stresses induced due to the flexural load are a combination of compressive and tensile stresses. For polymeric materials that break easily under flexural load, the specimen is deflected until rupture occurs in outer fibres. The effects of acetylation of flax on the flexural properties as a function of aging period are given in Fig. 7. The flexural strengths are found to be deliberately decreased up to 40

days aging period, and after that the property decreased slowly. It was also observed that the flexural strength seemed to be constant with aging period after 80 days. Compared to the initial value, the flexural strength decreased 15% and 30% for treated and untreated flax composites, respectively. The reduction of flexural strength may be for the same reason which is associated with tensile strength.

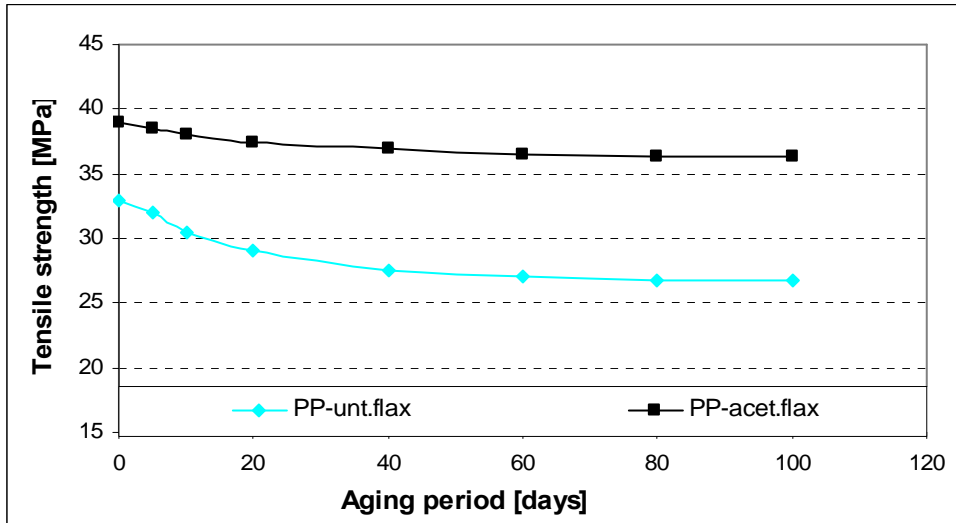


Fig. 6. Influence of aging period on the tensile strength of flax composites

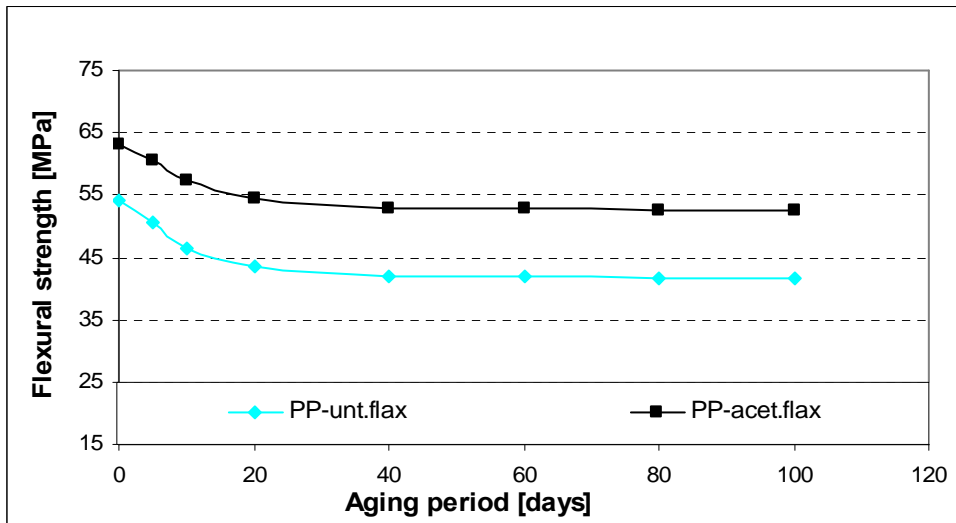


Fig.7. Influence of aging period on the flexural strength of flax composites

Charpy impact strength

The Charpy impact test is also known as a standardized high strain-rate test, which determines the amount of energy absorbed by a material during fracture. This absorbed energy is a measure of a given material's toughness and acts as a tool to study temperature-dependent brittle-ductile transition. Thus, composites having weak interfacial bonding can propagate cracks and undergo interfacial debonding. This leads to significant increase in the energy-absorbing capacity of the composites as a result of the large new surfaces produced and frictional work resulting from differential displacement between matrix and fibre, which increases the impact fatigue resistance of the composites (Bledzki et al. 2008). The variation of Charpy impact strength of flax composites as a function of aging period is presented in Fig. 8. It can be seen that the Charpy impact strength increased with increasing aging period for both cases.

Compared to the initial value, the Charpy impact strength increased 10% and 20% for treated and untreated flax composites, respectively. This is because of the weakening of interfacial bonding and increased energy absorption during the fracture due to the increasing influence of water absorption.

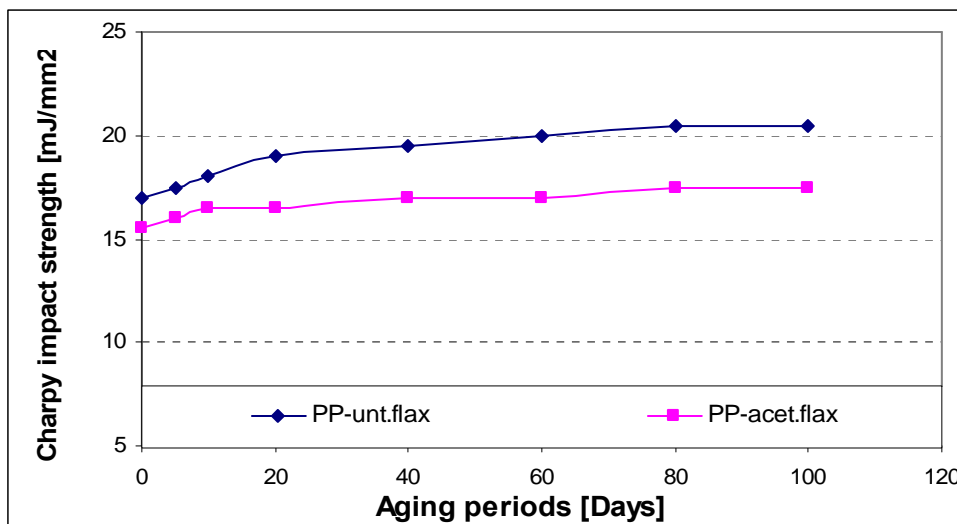


Fig. 8. Influence of aging period on the Charpy impact strength of flax composites

CONCLUSIONS

This study inspected the effect of acetylation of flax on the properties of reinforced polypropylene composites with respect to aging periods. The following conclusions could be drawn:

1. Moisture absorption and swelling properties of composites decreased by 60% and 30% respectively due to acetylation of flax fibre.
2. Biological resistance of composites improved by about 40% due to acetylation, and biocide had less effect on biological resistance.

3. Overall dielectric properties of composites improved due to acetylation, and water uptake (aging period) had a significant influence on dielectric properties.
4. Tensile and flexural strength properties increased by 15 to 20% due to acetylation and decreased 5% to 15% due to water uptake.
5. Charpy impact properties of treated and untreated flax composites increased by 10% to 20%, respectively, due to water uptake.

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