Effect of epoxidized soybean oil on mechanical properties of woven jute fabric reinforced aromatic and aliphatic epoxy resin composites

Péter Niedermann[1]
Gábor Szebényi[1]
Andrea Toldy[1]

Corresponding author:
Andrea Toldy
e-mail: atoldy@mail.bme.hu
Tel: +3614632462
Műegyetem rkp. 3-9. H-1111, Budapest, Hungary

[1] Department of Polymer Engineering, Faculty of Mechanical Engineering, Budapest University of Technology and Economics, Műegyetem rkp. 3-9. H-1111, Budapest, Hungary

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e-mail: atoldy@mail.bme.hu
Tel: +3614632462
Műegyetem rkp. 3-9. H-1111, Budapest, Hungary

[1]Department of Polymer Engineering, Faculty of Mechanical Engineering, Budapest University of Technology and Economics, Műegyetem rkp. 3-9. H-1111, Budapest, Hungary

Abstract

A systematic study was carried out to describe the effect of epoxidized soybean oil (ESO) on storage modulus, glass transition temperature (T_g) and mechanical properties in epoxy resin composites reinforced by jute fabric. In addition to aromatic diglycidylether of bisphenol-A (DGEBA) resin, a glycerol (GER) - and a pentaerythritol (PER)-based aliphatic resin was applied as base resin, which can be also synthesized from renewable feedstock. Based on strip tensile test results the usual alkali treatment of jute fabric was avoided. By increasing the ESO-content in aliphatic composites the T_g increases, while in case of DGEBA it decreases. The results indicate that although ESO has a significant softening effect, the jute fiber reinforced DGEBA composite can be replaced without significant compromise in mechanical properties by a potentially fully bio-based composite consisting of 25 mass% ESO-containing aliphatic PER reinforced by jute fibers.

Introduction

With the depleting mineral oil stock the interest for the renewable sourced structural materials is growing rapidly in polymer composite applications as well. In case of epoxy resins (EP) composite matrices epoxidized vegetable oils (EVOs), which can be synthesized by the epoxidation of different bio-based oils [1, 2], are one of the most researched renewable sourced resin components. Among these, epoxidized soybean oil (ESO) is possibly the most investigated one, as it is applied in large quantities as PVC plasticizer as well. In order to ameliorate the properties of EVOs, their blends with synthetic EP components and different types of curing agents are also considered [3-7].

To achieve fully bio-based composite systems, not only the matrix, but also the reinforcing fiber must be renewable sourced. Among the large variety of natural fibers available for this purpose, jute is one of the most promising renewable sourced reinforcing materials, thanks to its high cellulose content and relatively good mechanical properties compared to other natural fibers. According to the literature elemental jute fibers have the following characteristics: density: 1.3 g/cm^3; cellulose content: 61-71 mass%; hemicelluloses content: 14-20 mass%; lignin content: 12-13 mass%, wax content: 0.5 mass%. Furthermore, jute is produced in large quantities (2300 kt/year), so its structural application is not depending from the availability [8].

In case of natural fibers as jute the need for chemical treatment before composite preparation is an often discussed issue. Most frequently sodium hydroxide (NaOH) alkali treatment of the fibers is applied aiming at improving their mechanical properties by removing the non-cellulosic materials (lignin, hemicellulose) from the fibers. Several fiber treatment methods were published in the literature, however the results are contradictory, both improvement and worsening of the mechanical properties of the fibers is reported. Saha et al. [9] have reported almost 50% increase of jute fibers mechanical properties due to a treatment with 4 mass% NaOH solution at room temperature for 0.5 h, but above this specific treatment time and NaOH solution concentration the mechanical properties decreased in comparison to the untreated fibers. Similar effect was observed by Chakraborty et al. [10] with 0.5 mass% NaOH solution at room temperature and 24 h treatment time, the reported increment was 82% in this case. Gassan and Bledzki [11] examined the influence of alkali treatment on jute/EP unidirectional (UD) composites' mechanical properties with different NaOH solutions and treatment times. They observed 120% increment in the composites' tensile strength due to a treatment with 25 mass% NaOH solution for 20 min. at 20°C. Three different types of treatments were investigated on jute/EP composites in Doan's research [12]. NaOH treatment (1), 3-aminopropyl-triethoxy-silane solution and NaOH treatment (2) and 3-phenyl-aminopropyl-trimethoxy-silane solution and NaOH treatment (3). The highest improvement was observed in case of the latter (2) treatment. Pinto et al. [13] carried out mode I tests on jute/EP composites, where fibers were previously treated with 5 mass% NaOH for 2 h additionally to silane fiber pre-treatment. The specimens reinforced with treated fibers had better mechanical properties than the untreated ones, and with z-axis reinforcement the G_{IC} values improved as well.

The reinforcing effect of jute fibers is widely investigated in different EP matrices. Hossain et al. [14] investigated the effect of the fiber reinforcing direction on the jute/EP laminates mechanical properties, and concluded that in case of the 0°-0° reinforcing direction tensile and bending strength were the highest compared to the 0°-45° and 0°-90° reinforcing directions. Mishra and Biswas [15] found that in case of jute/EP composites, the hardness, tensile properties and impact strength increased and the void content decreased by increasing the fiber content. Several hybrid bio-based or bio-based/synthetic composites were investigated as well [16-24].

The applicability of natural sourced matrix and reinforcing materials instead of synthetic ones in the structural composite applications is a widely investigated area. Given that the composition and therefore the properties of plant oil based polymers and natural fibers is less constant than in case of synthetic ones, usually partial replacement is applied, especially in high-tech applications with strict safety standards, e.g. electrical, automotive and aeronautical industry. Other possibility is to apply these fully bio-based composites at first in less demanding areas of these advanced sectors, as aircraft interior applications. In order to map the application possibilities of bio-based resins in these more demanding sectors, in the present work a systematic research was performed to describe the effect of ESO addition in various EP systems reinforced by jute fibers. The expected outcome of this work was to define the composition of a potentially fully bio-based jute fiber reinforced composite, which contains the highest amount of ESO, with acceptable reduction in the T_g and mechanical properties compared to those of the neat EP composites.

In this research work the widely investigated and commonly used aromatic DGEBA resin, a glycerol (GER)- and a pentaerythritol-based (PER) aliphatic resin was chosen as base resin.

These aliphatic resins are currently produced on petrol oil base, however their synthesis is feasible from renewable feedstock as well (glycerol is available in large quantities from natural fatty acids, while pentaerythritol can be synthesized from bio-based methanol as well), eventually resulting in complete replacement of mineral oil based resins by renewable ones. As hardener a phthalic anhydride based component was used, which can be potentially also synthesized from natural sources [25], offering the possibility of a fully bio-based matrix material. Woven jute fabric reinforcement was chosen as natural sourced reinforcing material because of its accessibility in large quantities and promising mechanical properties according to the literature [8]. Given that the literature results on the effect of NaOH alkali treatment of natural fibers were found to be rather contradictory [9-13], prior to composite preparation, a systematic study was carried out to choose the optimal fiber treatment circumstances resulting in the best fiber mechanical properties.

Experimental

Materials

As matrix material of woven jute fabric reinforced epoxy resins blends of epoxidized soybean oil (ESO) with glycerol-based (GER) and pentaerythritol-based (PER) aliphatic EPs and with bisphenol-A based aromatic EP (DGEBA) were tested. In each case 25, 50 and 75 mass% ESO was added to the synthetic resin. The characteristics of the applied EP components are summarized in Table 1.

Table 1 Characteristics of the applied EP components

As hardener anhydride type curing agent was used with accelerator. The curing agent was methyl-tetrahydrophtalic-anhydride with minimal tetrahydrophtalic-anhydride content (Aradur 917) by Huntsman Advanced Materials (Basel, Switzerland) with the following characteristics: density at 25°C: 1.22 g/cm^3; viscosity at 25°C: 75 mPas. The accelerator was 1-methylimidazole (DY070) also by Huntsman Advanced Materials (Basel, Switzerland) with the following characteristics: density at 25°C: 0.99 g/cm^3; viscosity at 25°C: $\leq$ 50 mPas. During the sample preparation in all cases stoichiometric ratio of epoxy resin component and hardener was used. The accelerator was applied in 2 mass% related to the mass of the EP component.

Raw linen woven jute fabric with approximately 270 g/m^2 areal weight was applied as reinforcement in the composite systems. The fabric was provided by Műszaki Konfekció Kft. (Szeged, Hungary). To reach a minimum value of humidity content in the jute fibers, before the composite preparation the fabrics were dried in 80°C for 2 h under 50 mbar vacuum in Sheldon Manufacturing (Cornelius, USA) 1465 vacuum oven in each case.

For the alkali treatment of jute fibers NaOH was acquired from Sigma Aldrich in form of pellets with ≥97% purity. 0.5; 1; 2; 4 and 8 mass% NaOH solutions were prepared by dissolving NaOH pellets in distilled water. The solutions were let to cool down to room temperature for 2 hours.

Fiber treatment

5 pieces were cut from the jute fabric stored at 25°C, 53% relative humidity for each NaOH concentration. The jute fabric pieces were immersed into the previously prepared NaOH solutions and the pieces were removed subsequently after 0.5; 1; 2; 4 and 8 hours. The removal was followed by a three-step washing with distilled water to adjust the pH, controlled by universal pH paper. The washed jute fabric pieces were dried for 1 week at 25°C, 53% relative humidity, and afterwards for 1 day at 80°C under 50 mbar vacuum in Sheldon Manufacturing (Cornelius, USA) 1465 vacuum oven. The dried samples were consequently kept for 1 week at 25°C, 53% relative humidity in order to reach saturation, which was controlled by mass measurement (Radwag PS 200/2000.R1, Radom, Poland).

Composite preparation

The composite laminates were made by hand lamination in a press tool and then these laminates were put under compression with 200 bar hydraulic pressure in a Collin Teach-Line Platen Press 200E (Dr. Collin GmbH, Munich, Germany) press to achieve high and uniform fiber content in the composites. The fabric has higher specific tensile strength in weft direction. For composite preparation 6 layers of jute fabric were used for every composite resulting in 2 mm thick composite laminates. All the 6 layers were laid, and the specimens were machined in weft direction. During the composite pressing, heat treatment was applied. All laminates were heat treated for 2h at 100°C and 2h at 140°C for proper EP conversion. Fiber content measured on the basis of the fabrics areal weight was approximately 62 mass% in every case.

Determination of the effect of alkali treatment on jute fabric mechanical properties

Strip tensile test

For the strip tensile test 300 mm long pieces were cut from the jute fabric, each containing 20 yarns in warp direction. 5 samples were tested at each concentration and during each treatment time. The treated and untreated samples were subjected to strip tensile tests according to the ISO 13934-1:2013 standard using Zwick Z020 (Ulm, Germany) instrumented tensile tester with a grip distance of 200 mm and a cross-head speed of 10 mm/min 50 mm width. The applied force was parallel to the warp direction of the fabrics. The measured maximal force values were divided by the number of yarns, resulting in specific maximal force values, which were used for the comparison of the differently treated fabrics

Characterization of composites

Dynamic mechanical analysis (DMA).

DMA measurements were carried out to determine the dynamic mechanical properties and the T_g values in three point bending setup, with DMA Q800 device from TA Instruments (New Castle, DE, USA). The temperature range was 0-200°C with 3°C/min heat rate and 1 Hz frequency. The size of the specimens was 55x10x2 mm (length x width x thickness), and the support span was 50 mm. The amplitude was strain controlled with 0.1% relative strain.

Tensile test.

To determine the EP systems’ tensile strength and Young’s modulus a Zwick Z020 (Ulm, Germany) type computer controlled universal tester was used with a 20 kN load cell. The specimen dimensions were 150x10x2 mm (length x width x thickness) according to EN ISO 527. The initial test length was 100 mm. The test speed was 2 mm/min. The temperature was 22°C and the relative humidity was 59%.

Bending test.

To determine the bending strength and bending modulus values of the EP systems three point bending tests were carried out according to EN ISO 14125 with a Zwick Z020 universal tester with 20 kN load cell. The specimen size was 100x10x2 mm (length x width x thickness), and the support span was 80 mm. The test speed was 2 mm/min. The temperature was 22°C and the relative humidity was 59%.

Scanning electron microscopy (SEM).

Micrographs were taken of the fracture surfaces of characteristic tensile test specimens with JEOL (Tokyo, Japan) JSM 6380LA type SEM with 1000x magnification. To prepare a conductive surface the specimens were gold spur coated with a JEOL (Tokyo, Japan) JFC1200 device before the investigations.

Results and discussion

Effect of the alkali treatment on the mechanical properties of the reinforcing fabric

Strip tensile test

Prior to composite preparation, a systematic study was carried out to select the optimal fiber treatment conditions resulting in the best fiber mechanical properties. The maximal force values per fiber (N) determined from strip tensile tests of differently treated jute fabrics are displayed in Table 2.

Table 2 Effect of alkali concentration and treatment time on maximal force values per fiber (N) determined from strip tensile tests of jute fabrics

According to the results of Table 2 from the 25 different alkali treatment conditions only 4 resulted in a modest increase in the maximal force values (marked with grey background), in all other cases the tensile strength of the fabric decreased. The highest increase in maximal force was detected when the fabric was immersed into 1 mass% NaOH solution for 8 hours, however the increase is only around 5%, which taking into account the standard deviation cannot be considered as a significant amelioration. According to Kabir et al. [26] the non-cellulosic materials (hemicellulose and lignin) can be partially dissolved from natural fibers with the applied alkali treatment. As hemicellulose and lignin act as binding material in elementary jute fibers, less force is sufficient to move the elementary fibers from each other, resulting in a decrease in maximal force values. Based on these results we did not consider the alkali treatment of jute fabric justified, consequently untreated fabric was used for the preparation of composite specimens.

Characterization of composites

Dynamic mechanical properties

After preparation of jute fabric reinforced EP blends in order to determine the effect of the ESO content on the dynamic mechanical properties and the T_g values DMA measurements were carried out. Figure 1 shows the storage modulus curves as a function of temperature of all the hybrid and neat EP composite systems.

The composite systems' T_g values were determined by the tan δ peaks. Figure 2 shows the tan δ curves of all composite systems as a function of temperature.

Table 3 shows the T_g and storage modulus values at 0°C (below T_g) and 150°C (beyond T_g).

As it can be seen from Table 3, in most cases the storage modulus values decreased with the ESO addition in all composite systems. Three exceptions occurred: in case of PER with 25 and 50 mass% ESO content in glass transition range and in rubbery state, as well in case of DGEBA with 50 mass% ESO content in glassy and rubbery states the storage modulus values were higher than in case of the neat synthetic systems. The T_g values of the composites decreased approximately by 40°C compared to the corresponding neat EP systems (results previously published by the authors [7]). In the literature this effect is usually attributed to the reaction of hydroxyl (-OH) groups of the jute reinforcement and anhydride type curing agent [26]. As for the effect of ESO-addition on the T_g values of the composites, it can be concluded that in case of the aliphatic matrices, the T_g rather increases when the amount of ESO is increased. This synergistic T_g increase is in good correspondence with the behavior of the neat EP systems [7]. Also in good agreement with the neat resin, in case of aromatic DGEBA the T_g values rather show a decreasing tendency, nevertheless these values are still higher than in case of the aliphatic systems due to the more rigid aromatic backbone of DGEBA. Also, similarly to neat resins [7], peak doubling in case of tanδ curves of samples containing 75 mass% ESO in Figure 2 suggests that at higher ESO content phase separation occurred. In these cases dual T_g values could be determined as well (Table 3).

Figure 1 Storage modulus curves as a function of temperature of the jute reinforced EP/ESO composites

Figure 2 tanδ curves as a function of temperature of EP/ESO composites

Table 3 Storage modulus and T_g values of EP/ESO composites

Mechanical properties

Tensile and bending tests were carried out to investigate the effect of the ESO addition on the mechanical properties of the jute fiber reinforced epoxy resin composites. The mechanical properties of EP/ESO composites are shown in Table 4.

As the efficiency of the jute fiber reinforcement in woven structure was modest, the mechanical properties of the composite systems were rather determined by the mechanical properties of the matrices. In case of the aliphatic matrices the tensile strength showed a decreasing tendency due to the ESO-addition, above 50 mass% ESO-content the tensile strength values were lower than in case of the neat ESO composite, which could be the result of the phase separated structure [3-6]. The same tendency was observed in case of the Young’s modulus, but the values decreased below the level of neat ESO composite only from 75 mass% ESO content. It has to be emphasized that in case of natural fiber reinforced composites matrix properties have significantly higher effect on the tensile properties than in case of high performance fiber reinforced composites, because of the comparable fiber and matrix properties. The bending strength was in the range between the values of the neat aliphatic resin and neat ESO composites, in case of PER composite containing 25 mass%

ESO even a slight increase could be detected. As for the bending modulus, from 75 mass% ESO-content, the values decreased below the level of neat ESO composite.

In aromatic DGEBA systems all mechanical properties decreased with ESO addition, however they stayed above values of the neat ESO composites in all cases.

Table 4 Mechanical properties of the jute reinforced EP/ESO composites

Based on Table 4 among the hybrid ESO-containing epoxy resin composites the mechanical properties of the hybrid composites with 25 mass% ESO-content approach the properties of the reference DGEBA composite in the most values. In those applications where high T_g is not a crucial requirement, the jute fiber reinforced aromatic DGEBA epoxy resin composite can be replaced by 25 mass% ESO-containing hybrid epoxy components. Given that besides the natural jute fiber and ESO, both PER, GER and the anhydride based hardener can be potentially synthesized from bio-based sources as well, this leads to a replacement by a fully bio-based composite without significant compromise in mechanical properties.

Scanning electron microscopy (SEM)

In order to investigate the failure characteristics, the adhesion between the fibers and the matrix material, and to check the impregnation of the jute fabric, SEM investigation was carried out on the samples. The SEM images were taken from the tensile test specimens' fracture surface. As illustration, Figure 3 shows the aliphatic PER specimens fracture surfaces with different ESO-contents.

Figure 3 SEM micrographs of jute fiber reinforced PER/ESO composites in 1000x magnification (PER-ESO ratio: 100-0, 75-25, 50-50, 25-75, 0-100)

According to the SEM investigations some impregnation errors and in some cases debonding of the matrix from the jute bundles could be detected, but in general it can be stated that the impregnation of the fibers was acceptable. By increasing the ratio of ESO, no clear tendency of weaker fiber-matrix adhesion could be revealed, the weaker mechanical properties can be rather explained by the structure and properties of the ESO polymer matrix.

In order to study the effect of alkali treatment on the structure of the jute fabric one sample composite was prepared from jute fabric treated with 4 mass% NaOH solution for 2 hours and from the reference DGEBA-AR917 matrix. Figure 3 displays the fracture surface of this composite in 1000x magnification compared to the untreated jute reinforced DGEBA-AR917 matrix composites.

Figure 4 SEM micrographs of composite prepared from reference DGEBA-AR917 matrix and jute fabric in 1000x magnification (top: untreated fibers, bottom: fibers treated with 4 mass% NaOH solution for 2 hours)

Comparing the structure of the untreated fibers (top of Figure 4) and treated fibers (bottom of Figure 4), it can be stated that not only the binder between the elementary fibers (hemicellulose, lignin) was washed out, but also the hollow structure of these elementary fibers collapsed. This means that the structure of the fibers consisting of these elementary fibers degraded due to the alkali treatment, which explains the previously discussed decrease in tensile strength of the treated jute fabric (Table 2).

Conclusions

In order to facilitate the headway of natural fiber reinforced composites with bio-based resin matrix, a systematic research was performed to characterize the effect of epoxidized soybean oil (ESO) addition on the storage modulus, glass transition temperature and mechanical properties of jute fiber reinforced composites with a glycerol-based (GER), a pentaerythritol-based (PER) aliphatic EP and bisphenol-A based aromatic EP (DGEBA) matrix.

Prior to composite preparation, the effect of alkali treatment (in particular NaOH concentration and treatment time) was examined on raw linen woven jute fabric based on the literature on the alkali treatment of elementary fibers [9, 10]. According to the results above certain concentration and treatment time a significant decrease of specific maximal force values during the strip tensile tests could be detected, as the binder between the elementary fibers (hemicellulose, lignin) was washed out. Based on these results we did not consider the alkali treatment of jute fabric justified, therefore untreated jute fabric was used for the preparation of composite specimens.

Due to the addition of ESO the storage modulus decreased in almost all jute fabric reinforced composites. In aliphatic matrices the T_g rather increases when the amount of ESO is increased, while in aromatic DGEBA the T_g values rather show a decreasing tendency, nevertheless these values are still higher than in case of the aliphatic systems due to the more rigid aromatic backbone of DGEBA. This phenomena is in good accordance with the behavior neat epoxy resin systems (previously published by the authors [7]). The T_g values of the composites decreased approximately by 40°C compared to the corresponding neat epoxy resin, which is usually attributed to the reaction of hydroxyl (-OH) groups of the jute reinforcement and anhydride type curing agent.

As for the mechanical properties, significant weakening effect of ESO was detected in all composite specimens. In case of aliphatic PER and GER systems above 50 mass% ESO-content the tensile strength values were even lower than in case of the neat ESO composite, while the Young’s modulus values decreased below the level of neat ESO composite only from 75 mass% ESO content. The bending strength was in the range between the values of the neat aliphatic resin and neat ESO composites, in case of PER composite containing 25% ESO even a slight increase could be detected. In aromatic DGEBA systems all mechanical properties decreased with ESO addition, however they stayed above values of the neat ESO composites in all cases.

According to the results, in those applications where moderate glass transition temperature is acceptable, the jute fiber reinforced aromatic DGEBA epoxy resin composite can be replaced without significant compromise in mechanical properties by a potentially fully bio-based composite consisting of 25 mass% ESO-containing hybrid epoxy resin as matrix reinforced by jute fibers.

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