PhD Theses

Sustainable Polymeric Composites Reinforced by Fiber-Fabric Materials for Advanced Application

By

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Introduction

Cellulosic/artificial fiber reinforced composites are produced through reinforcing the respective fibers with suitable polymers/additives in the composite system. When there is at least one of the constituents in the composite systems are derived naturally can be called biocomposites. Additionally, the incorporation of nanoparticles in the composite is termed as nanocomposites. Composite materials have become a more fascinated product in this century for numerous potentialities. Natural fibers reinforced with polymeric materials like thermoplastic, thermosetting, and cementitious matrix are becoming popular significantly, especially for their light weight, strength, and sustainability features. The fiber-based composite materials provide exceptional stiffness, tensile and flexural properties, longevity, corrosion, and fire-resistant properties. Nanomaterials will also be a crucial part nowadays of developing composites with intensified performance characteristics (reducing weight, increasing mechanical properties with different improved functionalities). These remarkable characteristics have made composite materials convenient to apply for aerospace, construction, automotive, marine, biomedical, packaging, electronics, defense, furniture industries, and so on [1, 2]. Anyhow, the performances of the composite materials depend on the manufacturing processes, types of fibers used, and the polymer matrix/resin used. After all, considering the environmental sustainability and feasibility, there is a huge potentiality to develop composite panels or sheets with innovative and attractive features. Therefore, four different types of novel composites are produced and reported in this thesis report under four different work packages. In case of first work package, novel panels were produced through reinforcing rice straw and energy reed materials with phenol formaldehyde (PF) resin. The second work package entailed about the reinforcement of scots pine particle and coir fibers with OPC (ordinary Portland cement) matrix. In third work package, the possibilities of reinforcing flax/glass with MDI (Methylene diphenyl diisocyanate) resin were reported. Fourthly, the green synthesis of silver nanoparticles over glass/hemp woven fabrics were studied which were then finally reinforced with epoxy resin.

Objectives

- Developing natural fiber/particle/fabric reinforced composite materials
- Developing hybrid composite panels and nanocomposites
- Enhancing mechanical properties of composites
- To develop thermal conductive materials
- Improving thermal properties of developed materials
- Green synthesis of silver nanoparticles utilizing different parts of plants
- Developing lightweight and biodegradable materials
- Producing sustainable products for automotives, marine, aeronautical, defence, furniture, and construction and buildings sector

Work packages

The overall research reported in this thesis are divided into four work packages as we produced various types of composite panels from different polymers and cements which are partly published in different journal (mentioned in the list of publications). The work packages are named in terms of work package 1, 2, 3, and 4. The title of each work packages are provided bellow:

Work package 1: Rice straw and energy reed materials reinforced phenol formaldehyde resin polymeric biocomposites.

Both the rice straw and energy reeds are collected from central European regions to find out more diversified renewable materials for medium density biocomposite panels production. The incorporations of energy reeds with the rice straw enhanced the performances of developed biocomposite panels due to the attribution of positive hybrid reinforcement effects. Moreover, this research work would facilitate the biocomposite panel manufacturers with a sustainable and novel materials from renewable sources, where energy reed fibers could function as a new reinforcement biomaterial. Still now, the research on hybrid composites development through utilizing energy reeds and rice straws are not studied yet to make them usable as a prominent biocomposite material.

Work package 2: Semi-dry technology-mediated coir fiber and scots pine particle-reinforced sustainable cementitious composite panels.

The utilization of renewable lignocellulosic materials for flooring, wall, and roofing application in the building and construction sector could expedite lower costs, lower energy consumption, lower environmental burdens, and thermal comfort. Consequently, cementitious materials developed from renewable plant-derived resources and OPC could encourage construction that is more environmentally sustainable. Though forested areas in the world continue to shrink, the combination of natural fibers with plants [3] could accelerate potential alternative lignocellulosic materials for OPC bonding via hybrid composite production. The current study implemented a semi-dry technology to produce composite panels from coir fibers and Scots pine particles and their associated composites. These are similar to the wood particle/cement composite panels used by the manufacturing industry. Comparatively, semi-dry technology is cheaper and more convenient as it is less capital intensive and less labor intensive [4, 5]. However, research on semi-dry technology oriented natural fiber-reinforced cementitious materials remains limited. In this regard, the current research developed cementitious composite panels using semi-dry technology, which is a novel and innovative fabrication method for scots pine/coir fiber and OPC that, to our knowledge, has not been reported yet. Moreover, this research will facilitate manufacturers of engineered construction materials by means of an economic, green, and feasible method of hybrid production as well as composite panel production.

Work package 3: Thermomechanical behavior of Methylene Diphenyl Diisocyanate-bonded flax/glass woven fabric reinforced laminated composites.

Glass and flax woven fabrics were selected for this research. Some studies also revealed, that pretreatment of both the flax and glass fibers could improve the interfacial bonding in the polymeric composite [6]; so both the flax and glass woven fabrics were treated with NaOH and silane, respectively before producing the hybrid composite. Different percentages of flax and glass woven fabrics were used as reinforcement to fabricate the composites through reinforcing with MDI matrix. According to our knowledge, no research have been conducted yet on pretreated flax and glass woven fabric-based laminated composites reinforced with MDI polymeric resin. As MDI is used widely in industrial particle board manufacturing process, I hope this current research could facilitate the bulk productions of glass/flax reinforced MDI composites. The mechanical, physical, morphological, thermal, and statistical analysis have provided significant information on the produced hybrid composites.

Work package 4: Hemp/glass woven fabric reinforced laminated nanocomposites via in-situ synthesized silver nanoparticles from *Tilia cordata* leaf extract.

The hybridization of natural fibers with synthetic materials like glass could solve the problems of lower mechanical/physical properties obtained through cellulosic fibers reinforcements. The woven fabrics had been treated with AgNPs first before going to composites production which could bring a revolutionary development on the colorful laminated composites manufacturing houses. The presence of green AgNP was tested and confirmed by using XRF and ICP OES test. The developed hybrid composites were characterized for thermal, mechanical, physical, and morphological performance and found satisfactory. The colorimetric studies also ensured the presence of brilliant color appearances on the developed composite materials. Mechanical properties of these composites were tested on specimens prepared from the panels. Statistical analysis of the test results confirmed the positive influence of nanosilver loading on hybrid composites. According to our knowledge however, until now no research has been performed on biosynthesized AgNP treated thermoset polymeric hybrid composites with glass and hemp woven fabrics reinforcements. Moreover, the synthesis of nanoparticles from *Tilia cordata* leaf extracts to treat glass and hemp woven fabrics also not yet studied for producing hybrid nanocomposites although having superior potentiality. Hence, there is an urgent necessity is urged to explore the reinforcement effects of nanosilver treated glass and hemp woven fabric reinforced epoxy composites. The current research could facilitate the industrial production units with new route of nanosilver loaded laminated hybrid composite products.

Materials and Methods

Work package 1

The energy reeds were obtained from Energianövény-Team Kft., company located in Lengyeltóti, Hungary. The rice straw was received from local areas of central Europe (Hungary). However, both the rice straw and energy reeds were dried at ambient temperature and defibrated using a defibrating machine. The fiber materials were sieved for ensuring homogeneous fiber dimensions before going to composite productions. Phenolic resin-like PF was supplied by Chemco, a. s. in Slovakia for the purpose of research. The PF resin is reddish-brown in appearance and liquid. The dynamic viscosity of the PF resin was within 240 to 1080 mPa.s, density 1210±20 kg/m^3, dry matter content minimum 48 wt%, pH 10–12, and maximum free phenol content 0.1 (wt%). Both rice straw and energy reeds were cut into smaller pieces and the defibrated. The defibrated materials were then sived and used to produce biocomposite panels. The hot pressing technology was implemented for manufacturing biocomposites from rice straw and energy reed fibers. A moisture analyzer (Kern ULB 50–3 N, KERN AND SOHN GmbH, Germany) was used for investigating the moisture contents of fiber materials. However, accuracy of the equipment was 0.001 g, whereas the temperature was 105±0.3 °C. The standard EN 322:1993 was followed for moisture content investigations. Nearly around 1 g of scots pine and coir materials each were taken for this test. Thermal conductivity of rice straw and energy reed fiber reinforced PF composites were investigated as per EN ISO 10456:2012 standard through following hot plate method at ambient atmospheric conditions (relative humidity 65±5% and temperature 20±2 °C). The size of the tested samples was 350 mm by 350 mm, whereas the thickness was 10 mm. 100 readings were taken for each plates. The panels were placed between the hot and cold plates. However, the temperature gradient was kept 10 °C during the test. The mechanical performances of biocomposite panels were tested in terms of flexural properties (strength and modulus) and internal bonding strengths through Instron testing machine (4208, United States). The size of samples for flexural properties were determined by 20t+50, where t is thickness. In this regard, the length of the samples was 10×20+50 = 250 mm and the width was 50 mm. There were six samples from each composite type prepared and taken for the respective tests as per the standards. The standard EN 310 was followed for flexural properties investigation and EN 319 for internal bonding strength. The length and width of the samples prepared for internal bonding strength and physical properties were 50 mm by 50 mm whereas thickness was constant (similar with composite plate). Furthermore, morphological studies were conducted through a SEM equipment (S 3400 N, High Technologies Co., Ltd., Hitachi, Japan) by means of 100x and 200x times magnifications at 15.0 kV for both unfractured and fractured samples. The sizes of the test samples for SEM were nearly around 5 × 5 × 2 mm^3. In order to test FTIR, the samples were prepared nearly around 10 × 10 mm^2 dimension (length × width).

Work package 2

The Scots pine (*Pinus sylvestris*) particles were provided by FALCO Woodworking Co., in Szombathely, Hungary. The coir (*cocos nucifera*) fibrous chips were obtained from a local company named Pro Horto Ltd., located in Szentes, Hungary. The scots pine particles were used as received from the company, while the coir fibrous chips were defibrated before they were fabricated into composite panels. Typical OPC CEM I 42.5 was used as the cement matrix for reinforcing Scots pine and coir fibers in order to manufacture the composite panels. The FALCO Wood working Co., also provided the cement, which was manufactured by Duna-Dráva Cement Kft., in Vác, Hungary. Error! Reference source not found. presents the physical and morphological tphoographs/micrographs of the Scots pine, coir fibers, and OPC. The additive reagent (water glass, Na_2SiO_3) was procured from Sigma Aldrich, Hungary. Both the materials were sieved and used for producing panel productions using pressing technology. All the products are characterized using nearly same protocols with work package 1.

Work package 3

The flax woven fabrics (article number: LV06506, density: 230 g/m^2, composition: 100% flax, and Twill structure) were purchased from Malitext (Pécs, Hungary). The glass woven fabric (with measured density of 255 g/m^2, 100% glass, plain weave structure (grid size 4.4 × 4.2 mm^2)) was procured from Tolnatext company located in Tolna, Hungary. The alkaline NaOH was bought from VWR international Kft., (Debrecen, Hungary) and Vinyltrimethoxysilane, $C_5H_{12}O_3Si$ (L#MKBZ5796V, 98%, molecular weight (M_W) 148.23 g/mol) from Sigma-Aldrich Co., (St. Luis, MO, USA). The MDI (Ongronat XP-1161) was purchased from Borsodchem Zrt. (Kazincbarcika, Hungary). The Formula Five type mold release wax was procured from Novia (Hungary) to use as a coating material between the composites and teflon sheet to avoid stickiness of the resin with the teflon. Initially, the flax woven fabrics were pretreated with 0.5% NaOH solution (material: liquor ratio was 1: 20) for 30 min at 100 °C temperature to enhance the fibers interaction to polymeric resin. The glass fabrics were treated by vinyltrimethoxysilane at room temperature for 30 min (material: liquor ratio was 1: 10). After the pretreatment, the fabrics were rinsed and washed three times to remove the alkaline mucus, Vinyltrimethoxysilane solutions, and other impurities from the surface. The fabric samples were then dried in an oven at 60 °C for 6 min. After that, six layers of glass (G)/flax (F) woven fabrics (G1, GF2, GF3, and F4) coated with MDI resin were stacked up by hand-layup method. The sequence of layers in the laminates were (G,G,G,G,G,G/ G,G,F,G,G,G/ G,F,G,F,G,F/ F,F,F,F,F,F) with thicknesses of 1.56, 1.9, 2.58, and 3.6 mm for G1, GF2, GF3, and F4 composites, respectively. The produced laminates were pressed (3.5 MPa pressure) by a pressing machine for 15 min at room temperature. Later on, the composites were then cured for 24 h at ambient conditions.

The tensile and flexural properties of the produced composites were measured by using universal testing equipment Instron 4208 (Instron corporation, USA). The tensile test was conducted as per ISO 527–1:11996 procedures whereas flexural properties were adopted from the ISO 178:2019 standard. The dumbbell shaped test specimens was used for tensile testing (length 175 mm and width 20 mm), whereas sample sizes of 85 mm in length and 10 mm in width were used for flexural properties test. Six samples from each composite were selected for conducting the test. The crosshead movement of tensile test was 10 mm/min and flexural test 5 mm/min. The FTIR characterization of the composites were performed using FTIR–6300 (Jasco, Japan) spectrometer at 4000–500 cm^{-1}. The morphological investigation was performed by using a SEM equipment (SEM, S 3400N, Hitachi, Japan) at 15.0 kV voltage with 2000x and 1000x magnifications. The similar testing protocol and dimensions of work packages 1 and 2 were followed for SEM, TGA/DTG, and FTIR tests. The TGA and DTG analysis was conducted using Themys thermal analyzer (Setaram Instrumentation, France) within 25 °C to 850 °C at 10 °C/min temperature gradient under nitrogen (N_2) atmosphere. The water absorbency was tested at 2 h, 24 h, and 240 h time intervals as per MSZ 13336-4:13379 method. Samples of 50 mm by 50 mm dimensions were prepared to execute this test. The composite samples were immerged into 30 mm depth of water. The moisture content of the composite boards was investigated in line with EN 322 methods. The dimensions of the samples were kept the same (50 mm by 50 mm).

Work package 4

Glass and hemp woven fabrics being prominent reinforcement materials and epoxy resin polymeric matrix materials were selected for our experiments. The images/micrographs of physical and morphological appearance of the glass and hemp woven fabrics. Hand woven hemp fabrics (density: 75 g/yard, organic fiber, and milky grey color) were purchased from Rambutan, Vietnam. The fabrics were made by tribes women in northern Vietnam. The E-glass plain woven fabrics as mentioned in work package 3 were also used as reinforcement material. Metallic silver precursor ($AgNO_3$, purity 99.98%) was purchased from Sigma Aldrich (St. Luis, MA, United States). Leaves of *Tilia cordata* tree were collected from the garden of University of Sopron, Sopron, Hungary. The epoxy resin (modified bisphenol A) and crosslinkers (hardener, modified cycloaliphatic amine) were purchased from Novia Kft., located in Hungary. The viscosity of the epoxy resin is within 800 to 1200 mPa.S, equivalent weight 175 to 190 g/equiv., and density 1100 to 1110 kg/m^3, whereas the hardener contains 40 to 70 mPa.S viscosity and 950 kg/m^3 density, equivalent weight 60 g/equiv. The epoxy resin and hardener were mixed by proportion of 100 to

33 in laboratory standard environment (65% relative humidity and 25 °C temperature). All the products were characterized using nearly same protocols with work package 3.

Results and Dissertations of the thesis

Thesis 1

Novel hybrid composites were developed from rice straw and energy reeds. According to our knowledge, no research were conducted before regarding the fabrication of rice straw and energy reed materials with PF resin through applying hot-pressing technology. Novel findings are, that 100% energy reed panels are suitable for structural purposes because of high strength and good moisture resistance, and 100% rice straw panels are rather suitable for thermal insulation purposes. There was an increased pattern of mechanical properties found with the increase in energy reed materials in the composite. The highest performances against internal bonding failure are displayed by the 100% energy reed fiber reinforced composite panels (0.52 (0.04) MPa) compared to all other types of panel (0.25 (0.02), 0.31 (0.09), and 0.34 (0.04) MPa) for composite 1, 2, and 3). The maximum value for MOR was seen for 100% energy reeds reinforced composites by 21.47 (2.12) MPa, whereas the lowest value was observed for 100% rice straw reinforced panels just only 11.18 (1.91) MPa with a 47.9 % decline from the highest one. Obtained better fiber to matrix interaction in the composite systems as seen in morphological studies. The SEM micrographs shown the strongly interacted lignocellulosic materials. 100% rice straw reinforced composite displayed lowest values of thermal conductivity (0.061 (0.00083)) W/(m.K), whereas the highest values found for 100% energy reed fibers reinforced composites (0.104790 (0.000571)) demonstrating rice straw materials exist better thermal insulation properties than that of energy reeds. The water absorption, thickness swelling, and moisture content studies of the developed hybrid biocomposite panels shown that 100% rice straw reinforced composites provided the maximum values (54.284 (2.6580)% for water absorbency, 38.572 (0.1744)% for thickness swelling, and 5.92 (0.6464)% for moisture content) whereas the energy reed fiber (100%) reinforced composites provided the lowest values (7.746 (0.3391)% for water absorbency, 9.383 (0.5115)% for thickness swelling, and 5.11 (0.2423)% for moisture content) after 2 h. the similar trend is also noticed after 24 h although the absorption rates started to decline gradually after 2 h. The sequence of the physical properties in terms of higher values is for the composites are clearly noticed in the figure which demonstrates that the values start to increase with the increase in rice straw content in composite system.

Thesis 2

Thesis 2.1: A novel semi-dry fabrication technology was implemented for the production of cementitious composite panels from scots pine and coir fibers. Novel insulation panels were developed from scots pine and coir fibers reinforcing with the cement. The obtained thermal conductivity values are 0.17045 (0.00352) W/(m.K) for 100% pine reinforcements, whereas 60% pine and 40% coir reinforcement was 0.16651 (0.00265), 50% pine and 50% coir reinforcement was 0.11646 (0.00263), 40% pine and 60% coir reinforcement was 0.11265 (0.00409), and 100% coir reinforcement was 0.10547 (0.00657) W/(m.K). Interestingly, when the coir fiber was induced in composite systems, the thermal conductivity values started to decline. That is why our study reveals the 100% coir fiber-reinforced cementitious composite to be the best insulation material (according to our tested result), whereas 100% scots pine displayed the lowest thermal conductivity according to the performance perspectives.

Thesis 2.2: The developed panels providing superior mechanical performances needed for construction and building materials. The flexural properties obtained for five composite panels were 6.22 (0.78), 6.77 (0.12), 6.78 (0.73), 7.97 (0.8), and 8.02 (0.87) MPa, respectively for 100% pine reinforcements, 60% pine and 40% coir reinforcement 50% pine and 50% coir reinforcement, 40% pine and 60% coir reinforcement, and 100% coir reinforcement after 28 days of air curing in the laboratory. The flexural strengths of 100% scots pine reinforced cementitious matrix was the lowest, whereas the patterns increase with the increase of coir fiber contents. Additionally, the internal bonding strengths of P@C1 displayed 0.63 (0.08) MPa, whereas PC@C2 was 0.64 (0.03), PC@C3 was 0.66 (0.02), PC@C4 was 0.68 (0.03) MPa, and C@C5 was 0.72 (0.06) MPa. The properties shown improved performances when coir fiber was induced in the composite systems. Obtained significant dimensional stability (water absorbency, thickness swelling, and moisture content) of the developed sustainable products. The incorporation of coir fiber leads to decreased moisture content as well as decreased water uptake. 100% coir fiber-reinforced cementitious materials also exhibited better stability against water compared to 100% scots pine reinforced cementitious composite panels. The moisture content for composite 1 (100% scots pine reinforced cementitious composite) was 5.3 (0.4)% and 3.9 (0.9)% for composite 5 (100% coir reinforced cementitious composite).

Thesis 3

Novel composite panels were developed from pretreated flax and glass through reinforcing with MDI resin. Composite panels were developed with four different ratios of glass and flax woven fabric reinforcement (100/0, 83.33/16.67, 50/50, and 0/100) to investigate their performances with MDI resin.

Thesis III.1

Pretreatment of the fibers provided better fiber to matrix interactions and better thermomechanical performances. The tensile strengths of pure glass (G1), glass/flax (GF2 and GF3), and pure flax (F4) reinforced MDI composites take the values of 78.61 (8.2), 69.63 (2.77), 49.44 (2.05), and 21.19 (1.59) MPa, respectively. The flexural strengths followed the similar trend as the tensile characteristics. The perceived flexural strengths were 211.9 (17.9), 147.7 (18.5), 58.9 (9.5), and 43.9 (3.5) MPa, respectively for G1, GF2, GF3, and F4 composites. The regression analyses of all the composites mechanical performances were conducted in terms of glass fiber proportion in the composites. The R^2 values for all the composites are higher than 0.57, except for flexural strength with R^2=0.41. The presence of glass fiber results in better mechanical performances of all the hybrid composites. The mechanical features of the produced composites were further analyzed conducting one-way ANOVA with the type of composites as categorical factor. These tests showed that strength of the different composites are significantly different as the p values are less than the assumed level of significance of 0.05. Better dimensional stability. The G1 (pure glass) sample absorbed the lowest moisture whereas F4 (pure flax) attained the highest moisture content. The hybrid composites (GF2) attained moisture content of 1.34 (0.32) %, 1.88 (0.29) %, and 2.15 (0.09) % moisture content after 2 h, 24 h, and 240 h; whereas GF3 showed 3.76 (0.08) %, 3.84 (0.33) %, and 3.97 (0.04) % moisture content within the same time period.

Thesis III.2

Better thermal stability of developed hybrid composites was also found. The maximum weight losses (10% to 60%) were occurred at temperatures ranging from 315 to 450 °C. Besides, residues of the composites G1, GF2, GF3, and F4 were amounted to 87.99, 36.01, 58.78, and 30.82%, respectively.

Thesis 4

Green silver nanoparticles were synthesized from *Tilia cordata* leaf extracts over hemp and glass woven materials. Novel nanocomposites were developed from nanosilver treated glass and hemp woven fabrics laminated with epoxy resin. The nanosilver content values measured by XRF tests (0, 655±23, and 1829±30 PPM) for the composite types CC3, NC1, and NC2, respectively. The developed products also provided better thermomechanical performances with the increase in silver precursor in the colloid system. The highest tensile strength of 40.9 MPa was found for NC1, whereas CC3 exhibited the lowest value of 16.2 MPa. NC2 provided moderate tensile strength (18.9 MPa) between the two extremes. In case of bending strength, NC1 displayed the highest performances compared to the NC2, and CC3. The 1 mM loading of silver precursor has yielded the highest mechanical performances, although a 5 mM loading may have generated an agglomeration problem hence the mechanical properties declined, still remaining higher than those of the control composite plates. It seems that the nanosilver treatment on the fabrics used for laminations positively influences the mechanical properties of the produced composite panels. All the properties were tested in terms of silver NPs loading on each composite panel. The R^2 values for all the composites was found the values above 0.93 for all the mechanical properties demonstrating that the presence of nanosilver results in significant changes of the mechanical performances. The homogeneity of variances was always found met through ANOVA test as well. The developed composites also shown brownish coloration effects, whereas color strengths increased with the increase in nanosilver loading (control sample shown the K/S values by 2.3, 1.0 mM and 5.0 mM $AgNO_3$ loaded composites provided 3.5 and 11.8 values, respectively).

References

[1] A. Gholampour and T. Ozbakkaloglu, "A review of natural fiber composites: properties, modification and processing techniques, characterization, applications," *Journal of Materials Science*, vol. 55, pp. 829–892, 2020, doi: 10.1007%2Fs10853-019-03990-y.

[2] S. Karade, "Cement-bonded composites from lignocellulosic wastes," *Construction and Building Materials,* vol. 24, no. 8, pp. 1323-1330, 2010, doi: 10.1016/j.conbuildmat.2010.02.003.

[3] H. Savastano Jr, S. Santos, J. Fiorelli, and V. Agopyan, "Sustainable use of vegetable fibres and particles in civil construction," in *Sustainability of Construction Materials*. Duxford, United Kingdom: Woodhead Publishing, 2016, pp. 477-520.

[4] K. F. Hasan, P. G. Horváth, and T. Alpár, "Development of lignocellulosic fiber reinforced cement composite panels using semi-dry technology," *Cellulose*, vol. 28, pp. 3631–3645, 2021, doi: https://doi.org/10.1007/s10570-021-03755-4.

[5] T. Evans, A. Majumdar, and J. Ryder, "A semi-dry method for the production of lightweight glass-fibre-reinforced gypsum," *International Journal of Cement Composites and Lightweight Concrete*, vol. 3, no. 1, pp. 41-44, 1981, doi: https://doi.org/10.1016/0262-5075(81)90021-X.

[6] K. M. F. Hasan, P. G. Horváth, and T. Alpár, "Potential natural fiber polymeric nanobiocomposites: A review," *Polymers*, vol. 12, no. 5, pp. 1-25, 2020, doi: 10.3390/polym12051072.

List of publications

Journal publication (published)

1**. Hasan, K.M.F.**, P.G. Horváth, M. Bak, Z.M. Mucsi, L. Duong Hung Anh, and T. Alpár. Rice straw and energy reeds fiber reinforced phenol formaldehyde resin hybrid polymeric composite panels. Cellulose. **28** (2021) P. 7859–7875

2.**Hasan, K.M.F.**, P.G. Horváth, and T. Alpár. Development of lignocellulosic fiber reinforced cement composite panels using semi-dry technology. Cellulose. 28(6) (2021) P. 3631–3645.

3. **Hasan, K.M.F.**, P.G. Horváth, Z. Kóczán, and T. Alpár. Thermo-mechanical properties of pretreated coir fiber and fibrous chips reinforced multilayered composites. Scientific Reports. 11(1) (2021) P. 1-13.

4. **Hasan, K.M.F**, P.G. Horváth, and T. Alpár. Potential Natural Fiber Polymeric Nanobiocomposites: A Review. Polymers. 12(5) (2020) P. 1072.

5. **Hasan, K.F.**, et al., P.G. Horváth, A. Horváth, and T. Alpár. Coloration of woven glass fabric using biosynthesized silver nanoparticles from Fraxinus excelsior tree flower. Inorganic Chemistry Communications. 126 (2021) P. 108477.

6. **Hasan, K.M.F.**, P.G. Horváth, M. Bak, and T. Alpár. A state-of-the-art review on coir fiber-reinforced biocomposites. RSC Advance. 11(18) (2021) P. 10548-10571.

7. **Hasan, K.F.**, P.G. Horváth, T. Alpár. Thermomechanical Behavior of Methylene Diphenyl Diisocyanate-Bonded Flax/Glass Woven Fabric Reinforced Laminated Composites. ACS Omega. 6(9) (2021) P. 6124–6133.

8. **Hasan, K.M.F.**, P.G. Horváth, G. Markó, and T. Alpár. Thermomechanical characteristics of flax-woven-fabric-reinforced poly (lactic acid) and polypropylene biocomposites. Green Materials. 40(XXXX) (2021) P. 1-10.

10.Mahmud, S., **Hasan, K.M.F.** M.A. Jahid, K. Mohiuddin, R. Zhang, and J. Zhu. Comprehensive review on plant fiber-reinforced polymeric biocomposites. Journal of Materials Science. 56 (2021) P. 7231–7264.

11**. Hasan, K.M.F.**, P.G. Horváth, Z. Kóczán, M. Bak, A. Horváth, and T. Alpár. Coloration of flax woven fabric using *Taxus baccata* heartwood extract mediated nanosilver. Colour Technol. **00** (2021) P. 1-11.

12. **Hasan, K.M.F.**, P.G. Horváth, K. Zsolt, Z. Kóczán, M. Bak, A. Horváth, et al. Hemp/glass woven fabric reinforced laminated nanocomposites via in-situ synthesized silver nanoparticles from Tilia cordata leaf extract. Composite Interfaces. (2021) P. 1-19.

13. **Hasan, K.M.F.**, P.G. Horváth, Z. Kóczán, D.H.A. Le, M. Bak, L. Bejó, et al. Novel insulation panels development from multilayered coir short and long fiber reinforced phenol formaldehyde polymeric biocomposites. Journal of Polymer Research. 28(12) (2021) P. 1-16.

14. **Hasan, K.M.F.**, P.G. Horváth, Z. Kóczán, M. Bak, and T. Alpár. Colorful and facile in situ nanosilver coating on sisal/cotton interwoven fabrics mediated from European larch heartwood. Scientific Reports. 11(1) (2021) P. 1-13.
15. **Hasan, K.M.F.**, P.G. Horváth, Z. Kóczán, M. Bak, and T. Alpár. Semi-dry technology-mediated coir fiber and Scots pine particle-reinforced sustainable cementitious composite panels. Construction and Building Materials. 305 (2021) P. 124816.
16. **Hasan, K.M.F.**, X. Liu, Z. Kóczán, P.G. Horváth, M. Bak, L. Bejó, et al. Nanosilver coating on hemp/cotton blended woven fabrics mediated from mammoth pine bark with improved coloration and mechanical properties. The Journal of The Textile Institute (2021) P. 1-10.

Journal publication (under consideration/review/ under preparations)

17. **Hasan, K.F.**, Hasan, K.M.F., P.G. Horváth, Z. Kóczán, M. Bak, and T. Alpár. Effects of Sisal/Cotton Interwoven Fabric and Jute Fibers Loading on Polylactide Reinforced Biocomposites. Fibers and Polymers. Accepted.

18. **Hasan, K.F.**, et al. Sustainable coloration of aramid fabrics with Fomes fomentarius mushroom extracted nanosilver. Textile Research Journal. Under review.

19. **Hasan, K.F.**, et al. Green silver nanoparticle synthesis from sustainable coloration, biocidal, UV-protection, and thermal point of view: 2000 to 2020, A comprehensive review on game changing material, Progress in Polymer Coating. Under consideration.

20. **Hasan, K.F.**, et al. Flame retardant hybrid composites manufacturing through reinforcing lignocellulosic and carbon fibers with epoxy resin (F@LC). Industrial Crops and Products. Under consideration.

21. **Hasan, K.F.**, et al. Potential flame retardant hybrid composite panels development from fibers defibrated from cones and poplar. Composites Part A: Applied Science and Manufacturing. Under consideration.

Book chapter publication (Published/ under processing)

1. Topic: Nanotechnology for waste wood recycling, Publisher: Elsevier.
2. Topic: Introduction to biomass and biocomposites, Publisher: CRC Press (Taylor and Francis).
3. Topic: Design and fabrication technology in biocomposite manufacturing, Publisher: CRC Press (Taylor and Francis).
4. Topic: Silk protein and its nanocomposites, Publisher: Elsevier.
5. Topic: Industrial Flame-Retardants for Polyurethanes, Publisher: American Chemical Society (ACS).
6. Topic: Coir Fibre: Geographic distribution, and cultivation, Publisher: Elsevier.
7. Topic: Physicochemical and Morphological Properties of Microcrystalline cellulose and Nanocellulose Extracted from Coir Fibres and its composites, Publisher: Elsevier.
8. Topic: Nanomaterials-based smart and sustainable protective textiles, Publisher: Elsevier.
9. Topic: Community Entrepreneurship and Environmental Sustainability of Handloom Sector, Publisher: Springer.
10. Topic: Natural fibre reinforced vinyl ester composites: Influence of hybridization on the mechanical and thermal properties, Publisher: CRC Press.

Conference paper

1. **Hasan, K.M.F.**, F.Z. Brahmia, M. Bak, P.G. Horváth, G. Markó, L. Dénes, et al. Effects of cement on lignocellulosic fibres. in 9TH HARDWOOD PROCEEDINGS PT. I. 2020. Sopron, Hungary.
2. **Hasan, K.M.F.**, P.G. Horváth, and T. Alpár. Effects of alkaline treatments on coconut fiber reinforced biocomposites in 9th Interdisciplinary Doctoral Conference. 2020. Pecs, Hungary: Doctoral Student Association of the University of Pécs.
3. **Hasan, K.M.F.**, P.G. Horváth, M. Bak, and T. Alpár, *Morphological study on composite materials developed through reinforcing natural and synthetic woven fabrics from glass and hemp*, in *ITMC Conference*. 2021: Montreal, canada.
4. **Hasan, K.M.F.**, P.G. Horváth, M. Bak, and T. Alpár, Energy reed fiber reinforced thermosetting polymeric biocomposite, in Springwind Conference. 2021: Sopron, Hungary.
5. Hasan, K.M.F., P.G. Horváth, M. Bak, and T. Alpár, Thermal and Mechanical Characterization of NaOH Treated Coir materials reinforced composites. Proceedings of the 2021 Society of Wood Science and Technology. International Convention, Flagstaff, Arizona. August 1-6, 2021.
6. **Hasan, K.M.F.**, P.G. Horváth, M. Bak, and T. Alpár, Green insulation panels development from industrial lignocellulosic materials reinforced cementitious composites. 5th International Conference on Building Energy and Environment. July 2022, Montreal, Canada.
7. **Hasan, K.M.F.**, P.G. Horváth, M. Bak, and T. Alpár, et al. Morphological analysis of carbon woven and nonwoven fabric reinforced composite products. Interdisciplinary Doctoral Conference. 2021. Pecs, Hungary: Doctoral Student Association of the University of Pécs.