Journal of Polymers and the Environment (2021) 29:892–899
https://doi.org/10.1007/s10924-020-01889-3

ORIGINAL PAPER

Flexural Characteristics and Impact Rupture Stress Investigations of Sustainable Green Olive Leaves Bio-composite Materials

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Accepted: 14 September 2020 / Published online: 19 October 2020

Abstract

The desired features of the green composite materials including the low cost, environmentally friendly and degradation characteristics have made them very suitable for wide industrial applications. However, wide extensive investigations about various agro waste fibers are still required to expand the utilization of such fibers into green materials to enhance the overall desired properties of green composites. Thus, in this work, investigations of the effects of fiber loading of olive fibers in the low density polyethylene (LDPE) green composites were carried out. The overall bio-composite flexural properties and rupture stress under impact environment were studied. All of the applied force, flexural strain, flexural strength, flexural moduli and maximum stress at fracture under impact environment of various designed composites were studied to determine the optimal reinforcing conditions of such green composites. Various composites with different fiber loading (20 wt%, 30 wt%, 40 wt%, and 50 wt%) were designed and fabricated to investigate the effect of fibers on the considered properties of the composites. Results have revealed the optimum reinforcing conditions to enhance the overall flexural property of the composites. The maximum flexural strength of the green bio-composites with various fiber loadings was found at the 40 wt% fiber content case with 34.6 MPa. It was also shown that both 30 wt% and 40 wt% fiber loading cases had the highest flexural moduli of more than 800 MPa. All of flexural strength, flexural modulus and impact rupture stress properties were dramatically enhanced with fiber loading up to 40 wt%, but decreased after that due to agglomeration of fibers. Moreover, olive fibers were capable of improving the flexural modulus of LDPE of more than 200%.

Keywords Green composites · Olive · Flexural strength · Sustainability · Bio-materials

Introduction

Natural composite materials have been enormously utilized over the previous decades due to their proper mechanical properties compared to their low weight. They are being developed for wide range of engineering fields [1–4]. The lightweight property of green composite materials plays in fact a key role in the design of green products as they were highly adopted by proper material selection tools. Moreover, the low cost, environmentally friendly, availability and degradation features of the natural fibers that reinforced polymers to make green composite materials are also key drivers for implementing such materials in various sustainable industries [5–7]. Designers, in addition, try to find suitable types of materials that are organically degradable so as to decompose into the soil after their end of use via living organisms to reduce the accumulation of wastes and thus, pollution [8]. Green composites are classified as bio-composites if they are combined by natural fibers with biodegradable resins. They are called green composites mainly because of their degradable and sustainable properties, which can be easily disposed without harming the environment. Because of their durability, green composites are mainly used to enhance the life cycle of the green products [9, 10]. However, polymers are being utilized for the natural fiber composites to reduce the amount plastics as such composites are filled with agro waste fibers from cellulosic materials to end up with alternative solutions for the environment as well as reduce waste and achieve proper mechanical properties for various applications [11–14]. Such natural fiber composites are usually low cost, low density, none abrasive with low energy consumption during manufacturing.

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Several types of polymers can be used for producing natural fiber composites such as low-density polyethylene, high density polyethylene, polypropylene, and polyester as well as others. The selection of the composite constituents (fibers and polymers) has several points of view according to the final desired properties including the physical, chemical, mechanical, and cost [15–17]. In fact, several of such standpoints are conflicting and required means of optimization in order to make successful green products. Polyethylene, on the other hand, is one of the most suitable polymers for the natural fiber composites as it has several proper physical, chemical and mechanical properties. It is a thermoplastic polymer with variable crystalline structure and an extremely large range of applications depending on the particular type. Low density polyethylene (LDPE) is a very flexible material with very unique flow properties that makes it particularly suitable for plastic films. It has in addition, high ductility but low tensile strength. LDPE is utilized in several industrial applications including toys, squeeze bottles, chemical tank linings, carrier bags, high frequency insulations, general packaging, and water pipes [18–21].

Natural fibers in addition, come from three main sources: vegetal, animal and mineral. The vegetal one is the most used and it comes from plants or wood. They are considered as one of the most abundant materials in the world. They mainly consist of cellulose, hemicellulose and lignin. Cellulose is important for water up-take because of its hydrophilic properties, it's also important for the strength of plants structure [22–24]. Hemicellulose on the other hand, is an amorphous polysaccharide, and it is partially soluble in water. The binder of the components is the lignin.

The olive tree (Olea europaea L.) is a long-lived evergreen Mediterranean tree that grows in other various regions worldwide including Argentina, Norfolk Island, and California. Olive trees are grown because of their olive oil that is very beneficial type of food for human beings. It was reported that approximately 900 million olive trees over more than 10 million hectares worldwide are available. About 98% of which are in the Mediterranean basin [25]. The chemical compositions of olive tree leaf are Cellulose 11.28 wt%, Hemicellulose 14.73 wt%, Lignin 16.33 wt% and Others 57.66 wt% [25]. The large numbers of olive trees makes huge available row materials for green composite, as plenty of wastes accumulated yearly as a result of cultivation.

Several studies had investigated the effect of fiber types, fiber loading and chemical treatments on the on the mechanical properties of the final produced composites [26]. Hoverer, several unrevealed fiber types are ignored as they are not properly investigated in such types of composite materials including olive fibers [27]. Therefore, wide extensive investigations about various agro waste fibers are still required to expand the utilization of such fibers and to reveal their capabilities of producing new green materials to enhance the overall desired properties as well as reduce the amount of accumulated waste to enhance better environmental indices [28–33].

Accordingly, this work aimed to investigate the utilization of olive fibers in low-density polyethylene to produce low cost green materials as a limited number of studies had considered olives in such green composites, taking into considerations that olives are abundant in the Middle East and produce large amounts of wastes after cultivation every year. This in fact, would enhance producing low cost materials for various industrial applications. The work also aimed to systematically study the effects of fiber loading of the overall mechanical properties of the olive/LDPE composites to reveal their desired mechanical performance as wide extensive investigations about various agro waste fibers are still required to expand the utilization of such fibers into green materials. Moreover, the work aimed to enhance the reliability of utilizing such green composites as a sustainable alternative solution for wide industrial applications via revealing robust mechanical performance characteristics.

Materials and Methods

Low-density polyethylene was purchased from SABIC company, with tensile strength of 0.40 N/mm^2, thermal coefficient of expansion of 220×10^{-6} and density of 0.930 g/cm^3. Moreover, olive leaves from olive trees in Jordan were collected, washed with distilled water, air-dried at room temperature, and cut into small pieces. A digital caliper was used to measure the olive pieces to keep them of about 2 to 2.8 mm length and not more 1.7 mm wide. The olive fibers were then mixed with the LDPE in hot double screw mixer machine with various fiber loadings. Mixing time was 7 minutes with a speed of 40 rpm and 190 °C temperature. Chemical compositions of olive tree are demonstrated in Table 1, and the low density polyethylene polymer properties are demonstrated in Table 2. It can be seen that olive big branches contain more cellulose and hemicellulose than both small branches and leaf. However, olive leaf has the highest lignin content than the others.

Table 1 Chemical compositions of olive tree

Chemical composition	Olive leaf	Olive small branches	Olive big branches
Cellulose (wt%)	11.28	39.07	39.42
Hemicellulose (wt%)	14.73	23.62	24.23
Lignin (wt%)	16.33	13.26	14
Other (wt%)	57.66	24.05	22.35

Table 2 Properties of LDPE

Property	LDPE
Density (g/cm^3)	0.930
Melt Flow Rate at 190 °C and 2.16 kg (dg/min)	22
Melt volume rate (MVR) at 190 °C and 2.16 kg (ml/10 min)	29
Melting point (°C)	107
Tensile modulus (MPa)	175
Tensile strain at break (%)	400

Table 3 Composites configurations

Composite (wt%)	LDPE (wt%)	Olive (wt%)
10	90	10
20	80	20
30	70	30
40	60	40
50	50	50

Several fiber loading were designed to be 10 wt%, 20 wt%, 30 wt%, 40 wt%, and 50 wt%. The fiber-polymer mixtures were then pressed under hot press for about 15 mints to ensure minimal thermal shrinkage and then cut into various specimens with 80 × 20 × 10 mm. The composites configurations are illustrated in Table 3. The prepared samples were then tested for flexural mechanical properties using three point bending test according to ASTM D790 standard procedure.

Results and Discussion

Flexural strength is a critical mechanical property to determine whether a green composite material will fail under pressure and are especially important in any construction process involving ductile materials loaded with bending forces. If a green composite material starts to fracture or completely fractures during a three point bend test, then it is valid to accept that the material will fail under a similar case in any application, which may lead to catastrophic failure. Here various samples of low density polyethylene with olive fibers were designed and tested under various fiber loading particularly 10 wt%, 20 wt%, 30 wt%, 40 wt%, and 50 wt%. Figure 1 shows the mold used for producing the composites

Fig. 1 The green fibers, composite mold, and plate composite for the prepared materials and the performed test

as well as the plate of composite for making the samples under investigations in addition to the performed test. The flexural strengths and flexural moduli were studies for the considered green composite materials.

The load applied for each composite type is illustrated in Fig. 2, where close deflections for all specimens of different fiber loading were found before applying 0.4 Kn. However applying more load values demonstrates various deflections according to their fiber contents. This may be caused as the strain was initiated by the matrix which is similar to all specimens. However, with increasing load values, the stress efficiency between the matrix and the fiber varies resulting in various deflection resistances according to the composites ingredients and fiber loadings.

Flexural stress σ_f can be found on any point on the load deflection curve by utilizing Eq. (1)

$$\sigma_f = \frac{3FL}{2bd^2} \quad (1)$$

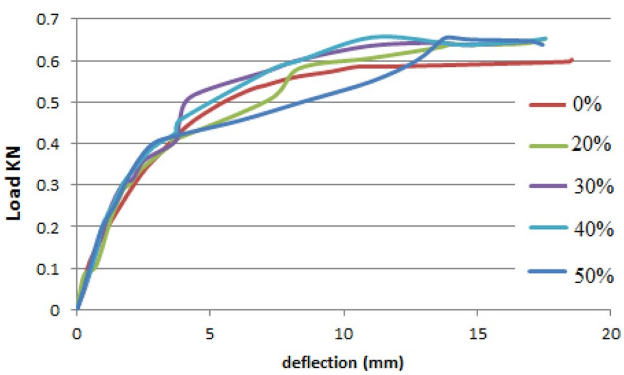


Fig. 2 Load /deflection behavior of the composites with various fiber loading

where σ_f is the flexural stress in [MPa] and F is the applied load at a given point in [N]. L, b and d are the length of the support span in [mm], width and thickness of the specimen in [mm] respectively.

Flexural strength is then the maximum capability of the composite material to resist the plastic deformation and can be found at the yield point in the flexural stress-deflection curve.

Flexural strain (ε_f) can then be found as in Eq. (2).

$$\varepsilon_f = \frac{6Dd}{L^2} \quad (2)$$

where D is the maximum deflection of the center of the specimen in (mm). L and d are the length of the support span and thickness of the specimen in [mm] respectively.

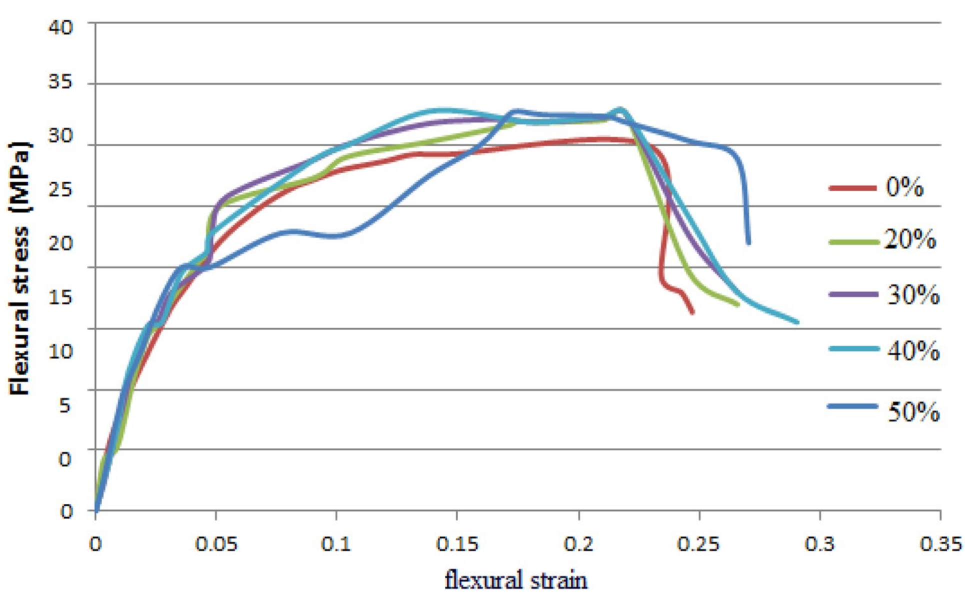
And the flexural modulus (E) can be found as in Eq. (3).

$$E = \frac{\sigma_f}{\varepsilon_f} \quad (3)$$

The experimental flexural stress–flexural deflection behavior of the various fiber loading of green composites are demonstrated in in Fig. 3. It can be demonstrated that the flexural moduli of elasticity are of the same order and close to each other with small variations. However, their maximum flexural strengths are different and vary with the fiber loading of the composites.

The maximum flexural strength of the olives/ LDPE composites with various fiber loadings is presented in Fig. 4. It can be demonstrated that the flexural strength increases with fiber loading until 40 wt% as fibers enhance the matrix flexural strength due to the compatibility of the fibers with the matrix. That is; this indicates to the good interfacial bonding between the olive fiber and the matrix. In addition, the

Fig. 3 Flexural stress–flexural strain behavior of the green bio-composites



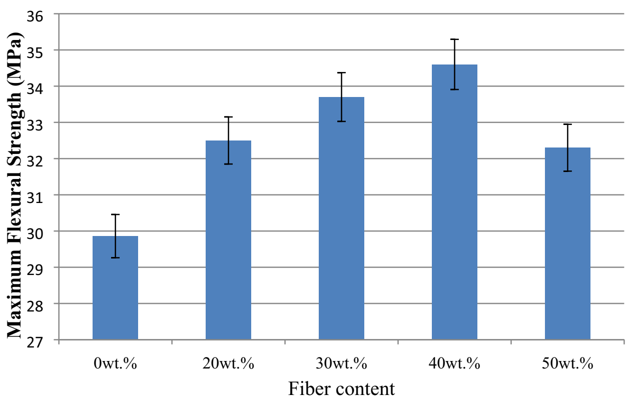


Fig. 4 The maximum flexural strengths of the green bio-composites with various fiber loading

surface roughness of the olive fiber could provide mechanical interlocking within the composite resulting in the good interfacial bonding between the olive fiber and the matrix. It also displays that with increasing fiber loading, fibers effectively participate in the stress transfer [34]. However, for the case of 50 wt% fiber loading, flexural strength decreased due to the fact that the high volume fraction of fibers has made agglomeration, and the matrix couldn't wet the whole fibers, leading to fiber pull and causing more holes and stress concentrations [21, 34, 35].

On the other hand, the maximum flexural strain of the composites with various fiber loadings is demonstrated in Fig. 5. It can be seen that flexural strain also increases with fiber loading but decreases for the case of 50 wt% fiber loading. This includes that olives fibers were able to enhance the flexural strain of the composites that enables wider applications for the composites than the matrix itself, particularly, for structural applications. The 40 wt% was the best among all fiber loadings as increasing fiber loading up to certain limit would make composites to have better stress transfer efficiency leading to enhance the flexural strain. It also displays that at increasing fiber loading, fibers effectively participate in the stress transfer [34]. In addition, the surface roughness of the olive fiber could provide mechanical interlocking within the composite resulting in good interfacial bonding between the olive fiber and the matrix. However, increasing the fibers will reduce the overall strain of the composite due to the same reason that made the composite strength weaker. That is; agglomeration of fibers occurred, and this usually leads to a performance deterioration in the composites.

Moreover, the flexural moduli of the composites under the effect of fiber loading are shown in Fig. 6. It can be revealed that fibers can enhance the flexural modulus of the composite as fiber loading increases. The increase in modulus is caused by increase of fiber content, which has a sufficient stiffness than the matrix [34, 35]. This is due to the fact that increasing fiber loading up to certain limit would make composites to have better stress transfer efficiency leading to enhance the flexural modulus. It also displays that at increasing fiber loading, fibers effectively participate in the stress transfer due to the good interfacial bonding between the fibers and the matrix [34]. It can also be seen that 30 wt% and 40 wt% fiber loadings are almost of the same order of flexural moduli. However, increasing the fiber loading to 50 wt% will decrease the flexural modulus due to fiber pull out from the composite as excess of fibers would find no enough matrix to properly wet the fibers and inappropriate adhesive force would occur comparable to the 30 wt% and 40 wt% cases [34–36].

Figure 7 in addition, demonstrates the various flexural characterisitcs of the examined composites with various fiber loadings. It can be demonstrated that the 30 wt% type as well as 40 wt% one of composites are the best among the overall flexural characteristics simeltenously. This in order can reveal the importance of investigating such bio-materials for considering various applications in order to obtimize the best desired properites for a certain application.

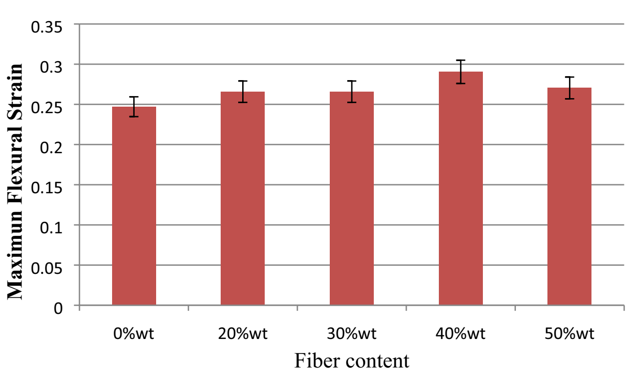


Fig. 5 The maximum flexural strain of the green bio-composites with various fiber loadings

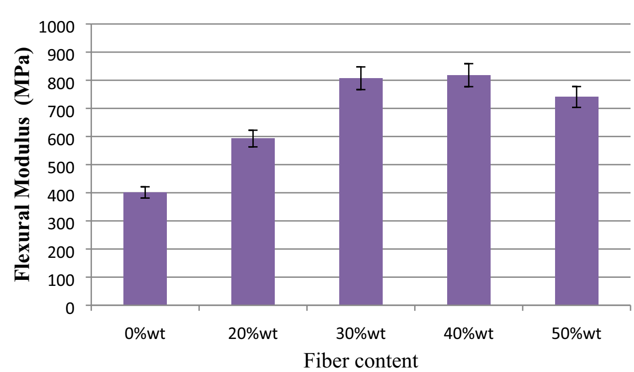


Fig. 6 Flexural moduli of the green bio-composites under the effect of fiber loading

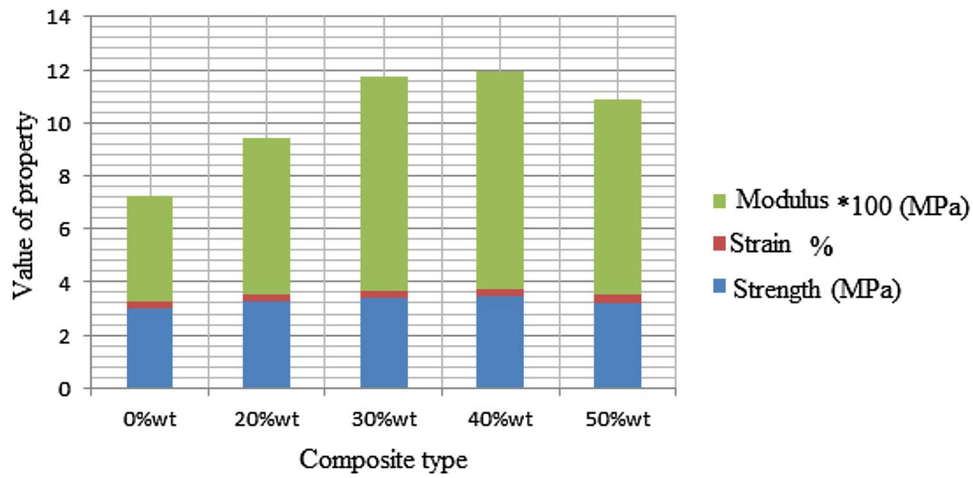


Fig. 7 The overall flexural characterisitcs of the examined green bio-composites

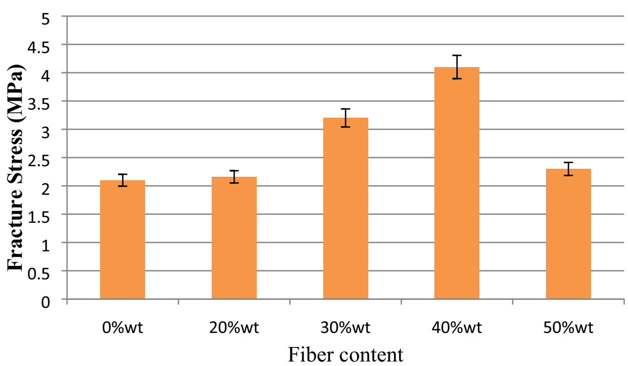


Fig. 8 Impact fracture stress of the green bio-composites

Table 4 Enhancement of rupture stress under impact loading

Fiber loading (wt%)	Rupture stress (MPa)	Stress enhancement (%)
0	2.1	–
20	2.16	2.857
30	3.2	52.380
40	4.1	95.238
50	2.3	9.523

On the other hand, impact is of principal for the bio-composites as impact strength generally measures the toughness of the materials when it is suddenly stressed. Thus, the materials maximum stress at rupture while impact loading is applied can give an idea about the biomaterials appropriateness for various applications when they are utilized in conditions with sudden impact like that of automotive applications. In fact, for polymeric bio-based materials, the major impact resistance comes from the matrix itself. However, green fibers usually reduce their impact capabilities as they enhance internal gaps and inclusions inside the matrix [35, 7]. Therefore, bio-composites usually suffer from their resistance to the impact loads, and composites with higher impact resistance are more desirable in their overall performance [37]. The impact test of the composites were conducted to demonstrate the maximum stress (fracture stress) the materials can attain under an impact environment. This is illustrated in Fig. 8.

The investigations of the materials maximum stress at rupture while impact loading is applied demonstrated that olive fibers had enhanced this quantity. It can be seen that the 20 wt% case did not dramatically enhance the maximum stress at rupture while impact loading was applied, whereas the cases of 30 wt% and 40 wt% fiber contents have improved that significantly. This is due to the fact that adding 20 wt% fiber loading would not be enough to make proper stress transfer efficiency inside the composite under impact loading [34, 35]. However, in cases of 30 wt% and 40 wt% fiber loading, composites had better stress transfer efficiency leading to enhance the maximum stress the materials can attain, under an impact environment, before rupture to about 52% and 95% respectively. The enhancement percentages of the fracture stress under impact loading are tabulated in Table 4.

Nevertheless, adding excess fibers to the matrix would make agglomeration inside the matrix without proper adhesive forces between the matrix and the fiber resulting in reducing the maximum stress comparing with the cases of 30 wt% and 40 wt%.

Conclusions

The utilization of olive fibers in low-density polyethylene to produce low cost green materials was performed. Olive fibers were capable of enhancing the flexural properties of the low density polyethylene. The trends of all composites with various fiber loadings were clear in improving the performance of the matrix. The optimal fiber loading for reinforcing the LDPE with olives was revealed to be 40 wt%. Olive fibers were capable of improving all of flexural strength, flexural strain, flexural modulus and impact flexural stress of LDPE. Flexural modulus was enhanced of about 200% at the optimal fiber loading, revealing the potential of olive fibers for the green composites as a reliable low cost ecofriendly alternative material for green products.

References

1. Zini E, Scandola M (2011) Green composites: an overview. Polym Compos 32(12):1905–1915
2. Thakur VK, Ding G, Ma J, Lee PS, Lu X (2012) Hybrid materials and polymer electrolytes for electrochromic device applications. Adv Mat 24(30):4071–4096
3. AL-Oqla FM (2017) Investigating the mechanical performance deterioration of Mediterranean cellulosic cypress and pine/polyethylene composites. Cellulose 24(6):2523–2530
4. Fares O, AL-Oqla FM, Hayajneh MT, (2019) Dielectric relaxation of Mediterranean lignocellulosic fibers for sustainable functional biomaterials. Mater Chem Phys 229:174–182
5. AL-Oqla FM, Salit MS, (2017) Materials selection for natural fiber composites. WoodHead Publishing, Cambridge
6. Lagel M, Hai L, Pizzi A, Basso M, Delmotte L, Abdalla S, Zahed A, Al-Marzouki F (2016) Automotive brake pads made with a bioresin matrix. Ind Crops Prod 85:372–381
7. AL-Oqla FM, El-Shekeil Y (2019) Investigating and predicting the performance deteriorations and trends of polyurethane biocomposites for more realistic sustainable design possibilities. J Cleaner Prod 222:865–870
8. AL-Oqla FM, Sapuan S (2017) Investigating the inherent characteristic/performance deterioration interactions of natural fibers in bio-composites for better utilization of resources. J Polym Environ 26(3):1–7
9. Almagableh A, AL-Oqla FM, Omari MA (2017) Predicting the effect of nano-structural parameters on the elastic properties of carbon nanotube-polymeric based composites. Int J Perform Eng 13(1):73
10. AL-Oqla FM, Omar AA, Fares O (2018) Evaluating sustainable energy harvesting systems for human implantable sensors. Int J Electron 105(3):504–517
11. AL-Oqla FM, Alothman OY, Jawaid M, Sapuan S, Es-Saheb M (2014) Processing and properties of date palm fibers and its composites. Biomass Bioenergy. Springer, Cham, pp 1–25
12. Yan F, Lin Z, Wang X, Azarmi F, Sobolev K (2017) Evaluation and prediction of bond strength of GFRP-bar reinforced concrete using artificial neural network optimized with genetic algorithm. Compos Struct 161:441–452
13. Bhatti HN, Safa Y, Yakout SM, Shair OH, Iqbal M, Nazir A (2020) Efficient removal of dyes using carboxymethyl cellulose/alginate/polyvinyl alcohol/rice husk composite: adsorption/desorption, kinetics and recycling studies. Int JBioMacro 150:861–870
14. Kausar A, Naeem K, Hussain T, Bhatti HN, Jubeen F, Nazir A, Iqbal M (2019) Preparation and characterization of chitosan/clay composite for direct Rose FRN dye removal from aqueous media: comparison of linear and non-linear regression methods. J Mater Res Technol 8(1):1161–1174
15. Agoudjil B, Benchabane A, Boudenne A, Ibos L, Fois M (2011) Renewable materials to reduce building heat loss: characterization of date palm wood. Energy Build 43(2):491–497
16. AL-Oqla FM, Salit MS (2017) Material selection of natural fiber composites using the analytical hierarchy process. Woodhead Publishing, Elsevier, Cambridge, pp 169–234
17. Ishtiaq F, Bhatti HN, Khan A, Iqbal M, Kausar A (2020) Polypyrole, polyaniline and sodium alginate biocomposites and adsorption-desorption efficiency for imidacloprid insecticide. Int JBioMacro 147:217–232
18. Al-Oqla FM, Omar AA (2012) A decision-making model for selecting the GSM mobile phone antenna in the design phase to increase over all performance. Prog Electromagn Res 25:249–269. https://doi.org/10.2528/PIERC11102702
19. Al-Oqla FM, Omar AA (2015) An expert-based model for selecting the most suitable substrate material type for antenna circuits. Int J Electron 102(6):1044–1055
20. Aridi N, Sapuan S, Zainudin E, AL-Oqla FM (2016) Mechanical and morphological properties of injection-molded rice husk polypropylene composites. Int J Polym Anal Charact 21(4):305–313
21. Asim M, Jawaid M, Abdan K, Ishak M (2018) The effect of silane treated fibre loading on mechanical properties of pineapple leaf/kenaf fibre filler phenolic composites. J Polym Environ 26(4):1520–1527
22. AL-Oqla FM, Salit MS, (2017) Natural fiber composites. Woodhead Publishing, Cambridge, pp 23–48
23. Sapuan S, Haniffah W, AL-Oqla FM (2016) Effects of reinforcing elements on the performance of laser transmission welding process in polymer composites: a systematic review. Int J Perform Eng 12(6):553
24. Aridi N, Sapuan S, Zainudin E, AL-Oqla FM (2017) A review of rice husk bio-based composites. Curr Org Synth 14(2):263–271
25. Alshammari BA, Alotaibi MD, Alothman OY, Sanjay M, Kian LK, Almutairi Z, Jawaid M (2019) A new study on characterization and properties of natural fibers obtained from olive tree (*Olea europaea* L.) residues. J Polym Environ 27(11):2334–2340
26. Shah DU (2014) Natural fibre composites: comprehensive Ashby-type materials selection charts. Mater Des 62:21–31
27. AL-Oqla FM, Hayajneh MT (2007) A design decision-making support model for selecting suitable product color to increase probability. Paper presented at the Design Challenge Conference: Managing Creativity, Innovation, and Entrepreneurship, Amman, Jordan,
28. AL-Oqla FM, Sapuan S, Anwer T, Jawaid M, Hoque M (2015) Natural fiber reinforced conductive polymer composites as functional materials: a review. Synth Met 206:42–54
29. AL-Oqla FM, Sapuan S, Ishak M, Nuraini A (2015) A model for evaluating and determining the most appropriate polymer matrix type for natural fiber composites. Int J Polym Anal Charact 20(just-accepted):191–205
30. AL-Oqla FM, Sapuan S, Ishak MR, Nuraini AA (2014) A novel evaluation tool for enhancing the selection of natural fibers for polymeric composites based on fiber moisture content criterion. BioResources 10(1):299–312
31. Al-Widyan MI, Al-Oqla FM (2011) Utilization of supplementary energy sources for cooling in hot arid regions via decision-making model. Int J Eng Res Appl 1(4):1610–1622
32. Al-Widyan MI, Al-Oqla FM (2014) Selecting the most appropriate corrective actions for energy saving in existing buildings A/C in hot arid regions. Build Simul 7(5):537–545. https://doi.org/10.1007/s12273-013-0170-3
33. Zhang K, Wang F, Liang W, Wang Z, Duan Z, Yang B (2018) Thermal and mechanical properties of bamboo fiber reinforced epoxy composites. Polymers 10(6):608
34. El-Shekeil Y, Sapuan S, Abdan K, Zainudin E (2012) Influence of fiber content on the mechanical and thermal properties of Kenaf fiber reinforced thermoplastic polyurethane composites. Mater Des 40:299–303

35. El-Shekeil Y, AL-Oqla F, Sapuan S (2019) Performance tendency and morphological investigations of lignocellulosic tea/polyurethane bio-composite materials. Polym Bull 1–14
36. Noreen S, Bhatti HN, Iqbal M, Hussain F, Sarim FM (2020) Chitosan, starch, polyaniline and polypyrrole biocomposite with sugarcane bagasse for the efficient removal of Acid Black dye. Int JBioMacro 147:439–452
37. AL-Oqla FM, Sapuan S, Ishak MR, Nuraini A (2016) A decision-making model for selecting the most appropriate natural fiber–Polypropylene-based composites for automotive applications. J Compos Mater 50(4):543–556