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Development and Performance Optimization of A Low-Carbon Environment Friendly Bio-Based Material

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Abstract

Global warming and environmental deterioration are common problems faced by human beings at present. Therefore, environmental protection and sustainable development have become the mainstream of development in the world today. Greenhouse gas emissions are the main cause of global warming (Sun et al,2013), while 80% of the world's energy demand is still supplied by traditional fossil fuels such as coal, oil and natural gas (Woolf et al,2010). In order to reduce the use of fossil fuels, we developed a new 3D printing material made of biochar and polylactic acid, which is expected to replace traditional petroleum-based materials in various fields. The purpose of this study is to improve the properties of materials by adding PBAT and modified biochar, and to evaluate the life cycle of composites.

We prepared the above composite materials and characterized them by SEM, FTIR and mechanical tests. The experimental results show that the addition of PBAT can increase the amount of biochar in the composite by 50%, increase the hardness of the material by 0.4%, and increase the tensile modulus by 69.12%; Sample S3 has the maximum bending strength of 61.89 MPa. Electron microscopy and functional group analysis showed that PBAT/PLA was well embedded and bound in biochar. This indicates that PBAT can increase the amount of biochar added to the composite and improve the interfacial compatibility of the composite.

Silane coupling agents can graft onto biochar, enhancing the polarity of biochar and reducing its water absorption by 50.32%; Modified biochar was applied to the composite, and functional group analysis showed that the characteristic peaks of silane coupling agent grafted biochar existed in the composite. The hardness of the composite material increased by 0.13%, the tensile strength increased by 9.83%, and the bending strength increased by 34.97%. The addition of PBAT reduced the total environmental impact potential value of the composite by 23.32%, and the modification process of biochar reduced it by 12.87%.

Keywords: Biochar; polylactic acid; PBAT; KH570; 3D printing; composite material; LCA.

The extensive use of traditional fossil energy is an important cause of global warming and environmental deterioration, which obviously does not conform to the theme of green sustainable development. Therefore, the replacement of traditional fossil energy by renewable biomass resources has become a hot spot of social research (Babu et al,2013). Biomass has the characteristics of large reserves, renewability, good cleaning performance and low price. However, the conventional way of using biomass is easy to cause resource waste and secondary pollution. Further conversion of biomass into biochar can greatly reduce the environmental problems caused by direct combustion of biomass (Bolan et al,2021). We have developed a new 3D printing material with biochar as raw material (Patent Number: 202210209057.7), which is expected to replace traditional petroleum-based materials in various fields. The purpose of this study is to improve the properties of materials by adding compatibilizer and modified biochar. On the one hand, it can solve the problem of poor mechanical properties of single polymer, improve material properties and realize the substitution of traditional petroleum-based materials; On the other hand, through the rational use of biochar, carbon sequestration can be realized and carbon neutrality can be achieved. This research is of great significance to the global carbon neutrality process.

1. The role of PBAT with different proportions in increasing the amount of biochar in composites and the effect of improving the interface compatibility of composites

1.1. Introduction

Poly (butylene adipate-terephthalate) (PBAT) is an aliphatic aromatic copolymer composed of aliphatic hydrocarbons and aromatic compounds. Because of its excellent mechanical and thermodynamic properties, it is considered as the best biodegradable material to toughen PLA (Wang et al.,2011). Many researchers have reported the blending of PBAT with polylactic acid (Pietrpsanto et al., 2020; Yu et al., 2019). PLA/PBAT blends were prepared by melt blending technology. It was found that with the increase of PBAT loading, the impact strength and tensile properties of PLA matrix were improved. The elongation at break of PLA/PBAT blends was 3%, which was lower than that of pure PLA (4.5%) and PBAT (500%) (Kumar et al.,2020). In the molten state, the interfacial transesterification reaction of PLA/PBAT blends (Coltelli et al.,2011) mixed two kinds of biodegradable materials, and the biodegradable materials with better performance and wider application range could be obtained. Therefore, the compatibilization modification of PLA/PBAT composites has become a research hotspot.

1.2. Methods

This experiment mainly explores the influence of PBAT on the addition of biochar, mechanical properties and water absorption of composites, so as to prove the improvement effect of PBAT on the compatibility of composites and determine its applicability as a compatibilizer for polylactic acid composites.

In this study, two factors and three levels L9 (3×3) orthogonal experimental design was adopted to explore the role of different proportions of PBAT in increasing the amount of biochar added in the composites and improving the interface compatibility of the composites. The experimental materials are shown in Table 1.The preparation method of that carbon/polylactic acid composite material is as following.

Pretreatment of Raw Materials. The biomass is dried at 105°C for 24h, then added into a pyrolysis furnace, and pyrolyzed in an oxygen-free environment to prepare biochar; After cooling, the biochar is put into a ball mill for crushing, then passed through a 200-300 mesh sieve, and the biochar sieve is collected and dried at 45-50℃ to constant weight.

Mixing raw materials. Mix the dried PLA raw materials, PBAT and biochar sieve according to the proportion in the orthogonal experimental table in Table 2, and place them on a shaker to vibrate uniformly for 60s.

Preparation of Composite Wire. The evenly mixed raw materials are added into the barrel of the extruder, and melted and extruded under the electric heating action of the screw of the extruder and the heating device.

FDM 3D Printing. According to the standards of GBT1040-2006 and GBT9341-2008, draw the tensile standard.

specimen and the bending standard specimen for printing, and set the packing density at 20%, the printing speed at 60mm/s, the nozzle temperature at 200°C and the hot bed temperature at 60 °C. After setting, print out the required tensile standard specimen and bending standard specimen entities for tensile, bending and hardness testing. The experiment scheme is shown in Table 2.

1.3. Testing and characterization

According to the national standard GB/T1040-2006 for tensile property measurement, the 1BA dumbbell tensile test standard sample was made by 3D printing, and then the tensile test was carried out on a universal testing machine (TY8000, CN) with 50 KN tensile sensor at room temperature at a speed of 5mm/min. According to the national standard GB/T9341-2008 for measuring bending performance, the standard bending test sample was made by 3D printing, and then the bending test was carried out at room temperature at the speed of 2mm/min on universal testing machine (TY8000, CN) with 50 KN compression sensor. Shore hardness gauges are used to measure the hardness of composite materials by measuring at least five samples and reporting their mean values and their standard deviations.

The functional groups on the surface of the composite were determined by Fourier transform infrared spectroscopy (FTIR). The test mode was ATR mode, and the scanning wave number ranged from 600 cm^{-1} to 4 000 cm^{-1}.

The density test method of composite material is to intercept 2cm length wires in different parts, measure their diameters and weights, repeat at least three times, and calculate the density of composite material. The test results are expressed as the arithmetic average of at least three results obtained under the same conditions.

According to the National Standard GB/T1034-2008 of plastic water absorption, dry the sample to be tested at 50℃ for at least 24h, and control the room temperature at 23℃-25℃. Then, after immersing the sample in water for 24h±1h, take out the sample and dry the water droplets attached to the surface of the sample. Weigh the weight changes of the sample before and after immersion, and calculate the water absorption mass fraction of the composite material. The test results are expressed as the arithmetic average of the three results obtained under the same exposure conditions.

1.4. Results

Figure 1 shows the scanning electron microscope of the composite material, and the fracture cross section SEM of the composite material prepared with different PBAT additions is shown in Figure 1. When the content of PBAT is low, there are more irregular pores in the composite, and the interfacial adhesion between PBAT and PLA is poor, which can be seen from a large number of voids penetrating the fracture surface of the composite. With the increase of PBAT content, the composite material has a strong bond between the filler and the matrix. There is no significant phase separation between the biochar and the matrix. With the increase of PBAT content, the interfacial interaction between PBAT/PLA and biochar is improved, and PBAT/PLA is well embedded and bound in biochar.Figure 2 shows the FTIR spectrum of the composite. The FTIR results show that the characteristic peaks of PLA (1745cm^{-1}, 1180cm^{-1}, 1040 cm^{-1}) and PBAT (730cm^{-1}, 870cm^{-1}, 1392cm^{-1}) are the main features of the composite. The peak at 590 cm^{-1} is caused by the tensile vibration of the carbon-carbon double bond (C=C) of biochar (characteristic peak of biochar), which indicates that PBAT, PLA and biochar are well combined in the composite.Figure 3 shows the mechanical properties of the composite. The results show that the content of biochar in the composite is increased by 50% by adding PBAT. The mechanical properties test showed that the hardness of the material increased by 4% compared with that when the addition of biochar was 10%. The tensile modulus, bending strength and bending modulus of the material increased. Figure 4 shows the density and water absorption of the composite. The results show that the addition of PBAT improves the density and water absorption of the composite.

2. Effect of silane coupling agent (KH570) modified biochar on compat ibility of composites

2.1. Introduction

The properties of composites are highly related to biochar, PLA matrix and the interface compatibility between them. Increasing the polarity of biochar in a certain range can effectively improve the mechanical properties of composites (Sudarisman et al.2015). The interface compatibility between carbon and PLA matrix is the key factor to determine whether the composite can bear the load. Modification of biochar is an effective way to obtain good interface compatibility of composites. Conventional modification methods, such as heat treatment (Sreekumars et al.2009) and alkali treatment of fiber (Huang et al.2019), can enhance the compatibility between biochar and composites, but at the same time, it will also damage the structure of biochar. Silane coupling agent can produce more chemical bonds between composites without damaging the structure of biochar, thus enhancing the interface compatibility of composites (Xie et al.2010). In this study, silane coupling agent KH570 was selected to modify biochar, so as to improve the surface adhesion between biochar and composites, improve its mechanical properties and make the material system have good stability.

2.2. Methods

The purpose of this study is to explore the effect of silane coupling agent (KH570) on modified biochar and the influence of modified biochar on the compatibility of composites.

Silane Coupling Agent Modified Biochar. Biochar was modified by KH570. The mass percentages of the modified solution were KH 570: 20%, absolute ethanol: 70% and water: 10%, respectively. The mass ratio of KH570 to biochar is 5: 1, 10: 1 and 15: 1, respectively. Immerse the pretreated biochar (300 mesh) in the modified solution, put it in a magnetic stirrer, stir for 1h, rotate at 20r/min at 30℃, fully mix the biochar with the modified solution, and stand at 30℃ for 12h; Standing and filtering the modified solution with a funnel, and washing the modified biochar with absolute ethanol to remove the residual silane coupling agent, so that the modified biochar is dispersed; The washed biochar was placed in an oven and dried for 4.5h, and the obtained samples were subjected to subsequent analysis, test and characterization experiments. Table 3 shows the experimental proportions and numbers.

Preparation of Composite Materials. The modified biochar was used to prepare composite materials, which was the same as the first part of the experiment, the prepared material is denoted by S9-2, and the composite materials were tested and characterized.

2.3. Testing and characterization

The water absorption and surface groups of biochar modified by silane coupling agent were tested to explore their physical and chemical characteristics and microstructure evolution. The influence on the compatibility between biochar and composites was analyzed, and its influence on the surface groups, mechanical properties, water absorption and density of composites was tested. The specific test method is the same as the first part.

2.4. Rusults

Figure 5 shows the FTIR spectrum and water absorption rate of the modified biochar. The results show that the peak at 1150 cm^{-1} is the stretching vibration peak of Si-O-C, which is the covalent bond formed by the condensation reaction between silanol and hydroxyl groups in biochar during heating, and the asymmetric vibration peak at 1 078 cm-1 is the overlap of Si−O−C and C−O−C. Compared with untreated biochar, the intensity of this peak is higher. The new peak at 795 cm-1 is the stretching vibration peak of Si−O−Si. These two characteristic peaks indicate that silane coupling agent has been successfully grafted onto the surface of biochar. The process of biochar modification enhanced the polarity of biochar, which reduced the water absorption rate and density of biochar by 58%. Figure 6 is a scanning electron microscope image of a biochar modified composite material. It can be seen that the composite material without modification by silane coupling agent is easily distinguished from each other and has many pores. This is because biochar is a polar molecule, while PLA is a nonpolar molecule. Due to differences in polarity, the compatibility between biochar and the matrix is poor. The modification with silane coupling agents has changed

the polarity of biochar, resulting in a tighter binding of the components of the composite material, a large number of pores, and a uniform dispersion of PLA between the pores of biochar. This is also the reason why the addition of silane coupling agents enhances the mechanical properties of the composite. Figure 7 shows the FTIR spectrum of biochar-modified composites. The results show that the characteristic peaks of silane coupling agent grafted biochar exist in the composites. The results of composite performance test in Figure 8 show that the repulsion between the modified biochar and the components of the composites is weakened, and the compatibility between biochar and matrix is improved. The maximum tensile strength and bending strength of the composites reach 34.3MPa and 71.1MPa respectively. The maximum tensile strength reached 78.6HD, and the tensile strength, bending strength and hardness of the composites increased by 9.83%, 34.97% and 0.13% respectively. Figure 9 shows that the density and water absorption of the composite material are reduced by modification treatment.

3. Life cycle assessment of composite materials

3.1. Research objective

Through the life cycle assessment of composite materials, we can calculate the carbon footprint of products and provide a reliable scientific basis for the environmental sustainability of products. Make a reasonable explanation of the main stages and substances in the processing and production process that cause pollution, and then put forward a reasonable optimization and improvement scheme to promote the green and sustainable development in the field of composite materials.

The benchmark flow of the life cycle assessment model is "producing and printing 1kg composite materials", and the system boundary is "cradle to gate". The relevant data of polylactic acid particle production process, PBAT particle production process, biochar powder preparation process, wire processing and printing process and raw material transportation process are collected, and the environmental impact potential of adding PBAT and biochar modification process on composite materials production is analyzed.

3.2. Life cycle inventory analysis

In this study, the relevant data of biochar powder preparation process, wire processing process, wire printing process and raw material transportation process were collected, and the life cycle list was established. The input raw materials mainly include: production raw materials; electricity and diesel oil. Energy consumption mainly involves electric energy consumption. The output mainly includes: waste water, waste gas, solid waste, etc. During the operation of the equipment, it is inevitable to produce certain noise pollution, which is temporarily ignored in this research model. The resource consumption, energy consumption and emission data of PLA and PBAT are all from Swiss Ecoinvent database; Background data about electricity, diesel oil and production process come from CLCD (Chinese life cycle database) database and Ecoinvent database developed by e-Footprint online platform; The energy consumption emission in the biological production process comes from the experimental field records. The life cycle assessment models of PLA/biochar composite (S0), PLA/PBAT/biochar composite (S9) and PLA/PBAT/modified biochar composite (S9-2) were established, which is used to explore the environmental impact of the modification process of PBAT and biochar on the composites.

3.3. Life cycle impact assessment

Life cycle impact assessment (LCA) refers to the transformation of input and output data in the whole production process of biochar/polylactic acid composites into designated environmental impact categories to determine the impact on the environment. Characterization is to classify the input and output according to the environmental impact category. According to the selected calculation model, multiply it by the corresponding characterization factor, and the data in the life cycle list will be converted into the corresponding environmental impact characterization indicators. In the characteristic index analysis, the most important environmental problem in the production process of biochar/polylactic acid composites is the high energy consumption. Therefore, 13 environmental impact

characterization indicators, such as global warming potential (GWP), primary energy consumption (PED) and water resource consumption (WU), were selected in the study, and the environmental impact of composite materials was comprehensively analyzed. The basic principle of normalization is to make the environmental impact indicators dimensionless by introducing the corresponding national benchmark values, so that different environmental impact categories can be compared and the main environmental impact types can be determined. The normalized benchmark value in this paper comes from the per capita data calculation (He et al.,2016) of China National Bureau of Statistics (NBS 2000), and is calculated by the weight factor (Wang et al.,2006). The weight factor is determined by the expert group appraisal method, and the weight factor has been approved by the expert group and widely used. Divide the characterization index by the corresponding normalized reference value to get the normalized value, multiply the normalized value by the corresponding weighting factor, and then add all the weighted results in each scene to get the comprehensive environmental impact potential value. The larger the normalized value is, the more serious the environmental impact of this category is. Similarly, the higher the comprehensive environmental impact potential in which scenario is, the greater the potential impact of this scenario on the environment is (Ding et al.,2022). Five environmental impact indicators, namely global warming potential (GWP), primary energy consumption (PED), water resources consumption (WU), acidification (AP) and eutrophication (EP), were selected for normalization and weighted analysis. Sensitivity analysis is mainly used to characterize the key part of the impact of input and output data in the life cycle assessment system on the results of LCA characterization indicators. Sensitivity analysis is conducted according to the percentage change of input and output data during the main process changes (Guo et al.,2021). The greater the sensitivity of the process, the greater the contribution of the environmental load generated by the process to the environmental impact characterization index. If corresponding improvement is needed, this process unit can be selected first.

3.4. Explanation of life cycle assessment results

The characterization results in Figure 8 show that the electrical energy loss in the production process of composite materials is the main contribution to the composition of life cycle results; The production loss of poly (lactic acid) particles is second only to the power loss in the life cycle results of S0 and S9. The contribution rate of production loss of polylactic acid particles in S9-2 to the life cycle results of composites decreased significantly. The normalized results in Figure 9 show that the total environmental impact potential of the composite material decreases by 23.32% after adding PBAT, that is, adding PBAT can effectively reduce the environmental impact of the composite material production; Compared with PLA/biochar composites, the total environmental impact potential of the composites decreased by 12.87% during the process of carbon modification. The sensitivity results in Figure 10 show that the process that contributes the most to the environmental impact is the production of composite materials; Under the production scenarios of three materials, the sensitivity of electric energy consumption in the production process of composite materials is the highest, so reducing the electric energy consumption in the production process of composite materials and biochar is the key to reduce the overall environmental burden.

4. Conclusion

By adding PBAT, the biochar content in the composite was increased by 50%. As the content of PBAT increases, the PBAT/PLA melt is well embedded and bound in the biochar, resulting in an increase in the density of the composite material and no obvious fracture through holes on the surface; The mechanical properties were improved, and the hardness of the material increased by 4% compared to that when the amount of biochar added was 10%. The tensile modulus, flexural strength, and flexural modulus of the material increase.

During the modification process of biochar, silane alcohol and hydroxyl groups in biochar undergo a condensation reaction during heating to form a covalent bond, and KH570 is successfully grafted onto the surface of biochar; After preparing the composite, infrared spectroscopy analysis showed that the modified carbon was well present in the composite; Scanning electron microscopy (SEM) showed that the components of the composite modified by silane coupling agent formed a closer bond, and PLA was evenly dispersed among the pores of the

biochar; The results of mechanical properties show that the modified biochar improves the tensile strength, flexural strength, and hardness of the composite by 9.83%, 34.97%, and 0.13%, respectively, compared to the untreated composite; Water absorption decreased by 34.29% and density decreased by 8.98%.

By comparing the environmental characteristics of composite materials under three different production scenarios, the following conclusions were obtained: the addition of PBAT reduced the total environmental impact potential value of the composite material by 23.32%, and the biochar modification process increased the total environmental impact potential value of the composite material by 13.67%, but compared to the PLA/biochar composite material, it still decreased by 12.87%.

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Table 1 Experimental materials

Material name	Specification/Model	Manufacturer
Mongolian pine- biochar	300	control oneself
polylactic acid	210	Zhejiang Haizheng
Poly- butyleneadipate-co- terephthalate	801t	Xinjiang lanshan tunhe- polyester co., ltd

Table 2 Sample proportion and number

PBAT addition ratio	Carbon addition ratio	Number
Pure polylactic acid		S0
5%	5%	S1
	10%	S2
	15%	S3
7.5%	5%	S4
	10%	S5
	15%	S6
10%	5%	S7
	10%	S8
	15%	S9

Table 3 Sample proportion and number of biochar modified by silane coupling agent

Number	KH570/g	Carbon/g	Ethanol/g	Deionized water/g
K0	-	-	-	-
K1	5	1	17.5	2.5
K2	10	1	35	5
K3	15	1	52.5	7.5

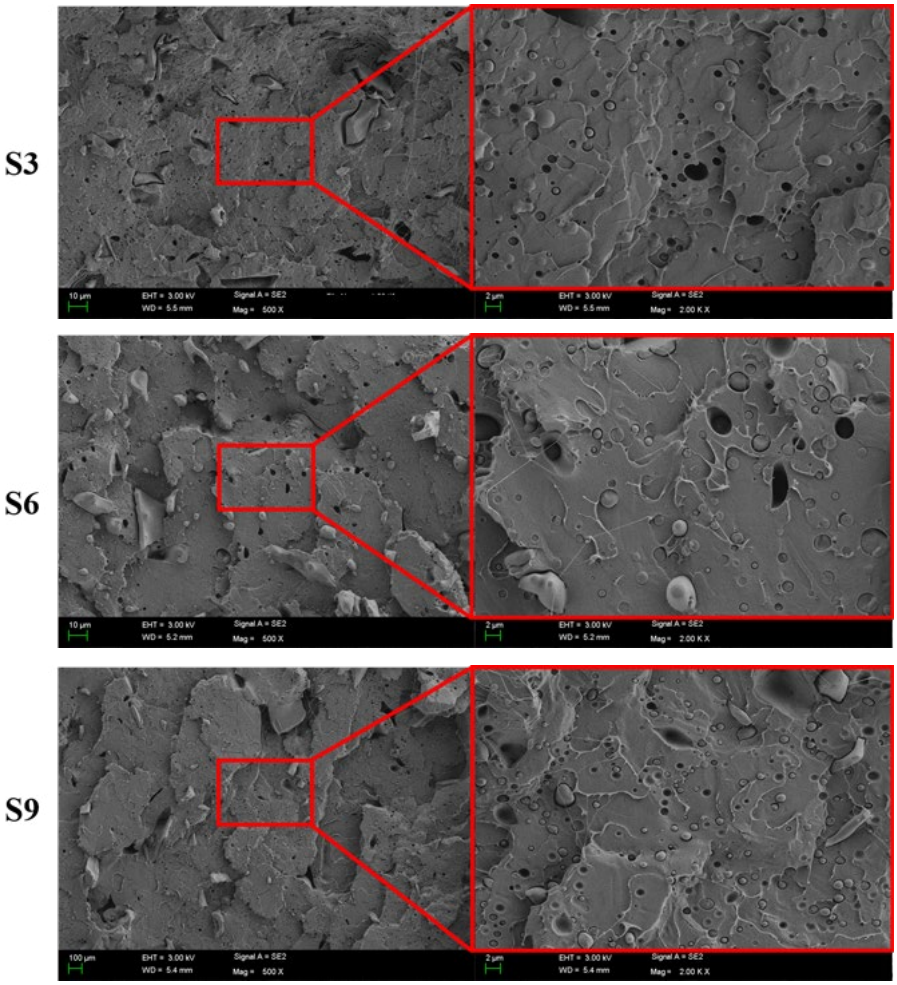


Figure 1 SEM of composite materials

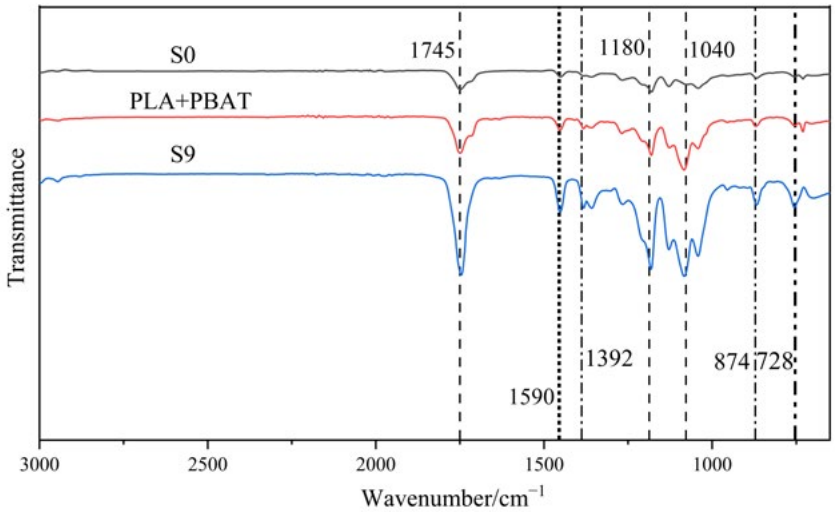


Figure 2 FTIR spectra of composites

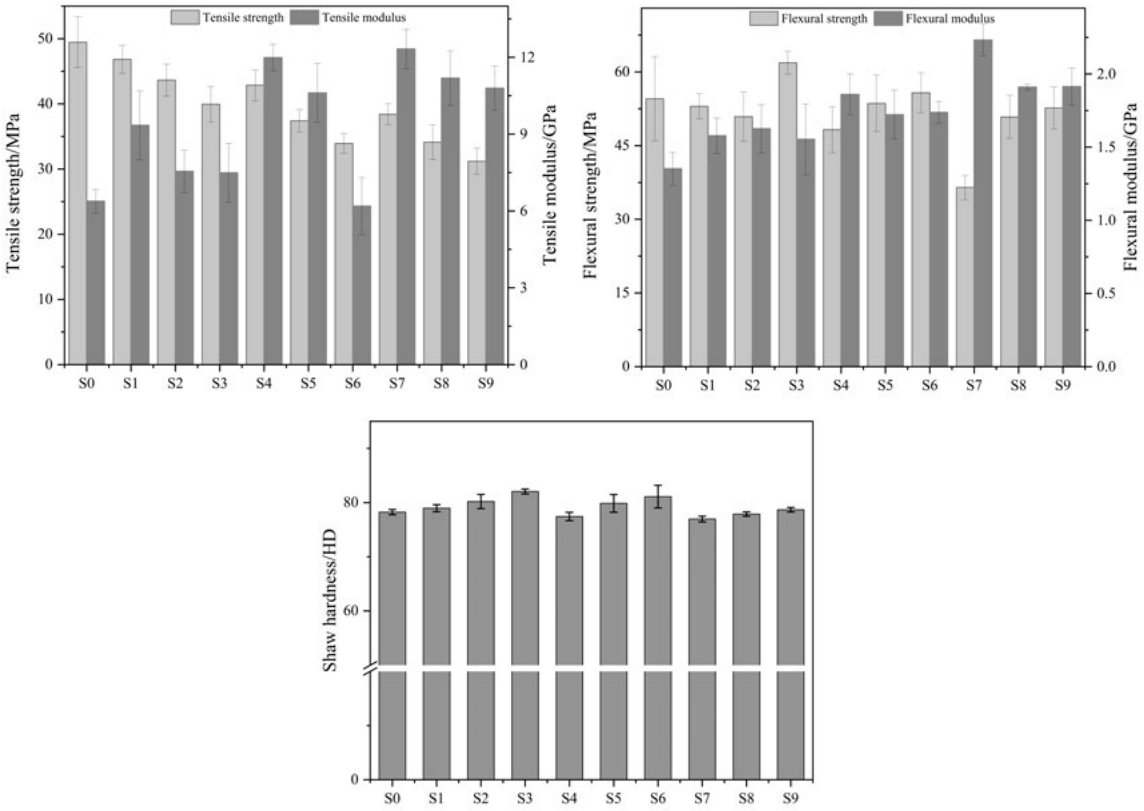


Figure 3 Mechanical properties of composites

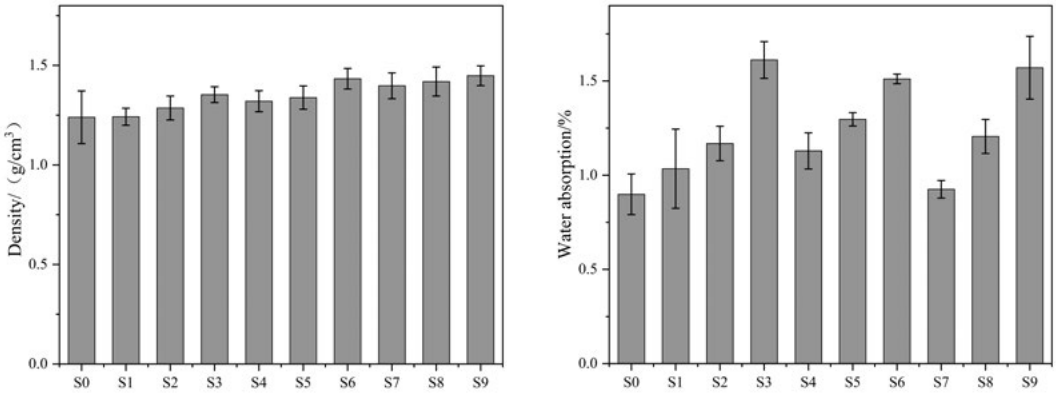


Figure 4 Density and water absorption of composite materials

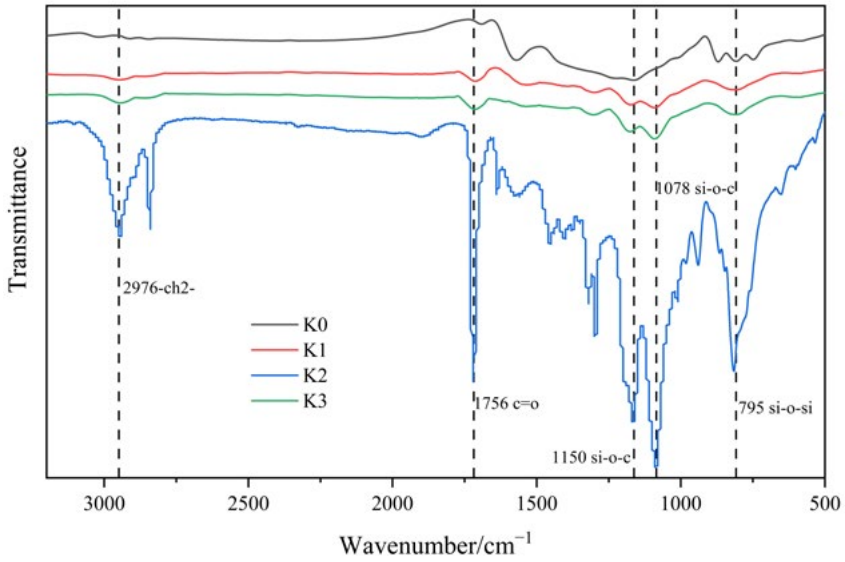


Figure 5 FTIR spectra and water absorption of modified biochar

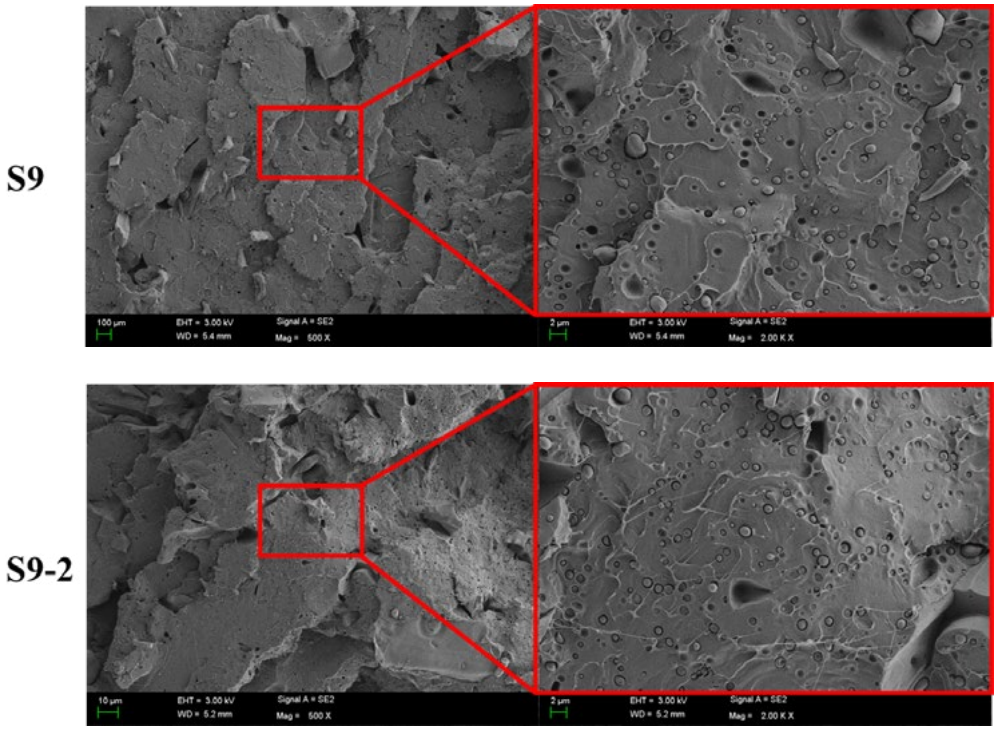


Figure 6 SEM scanning of modified composite materia

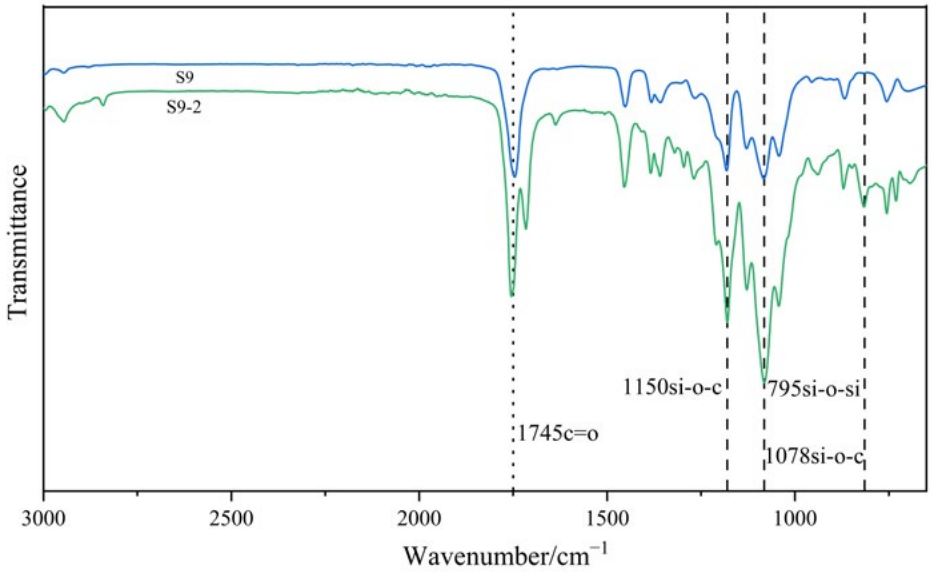


Figure 7 FTIR spectra of biochar modified composites

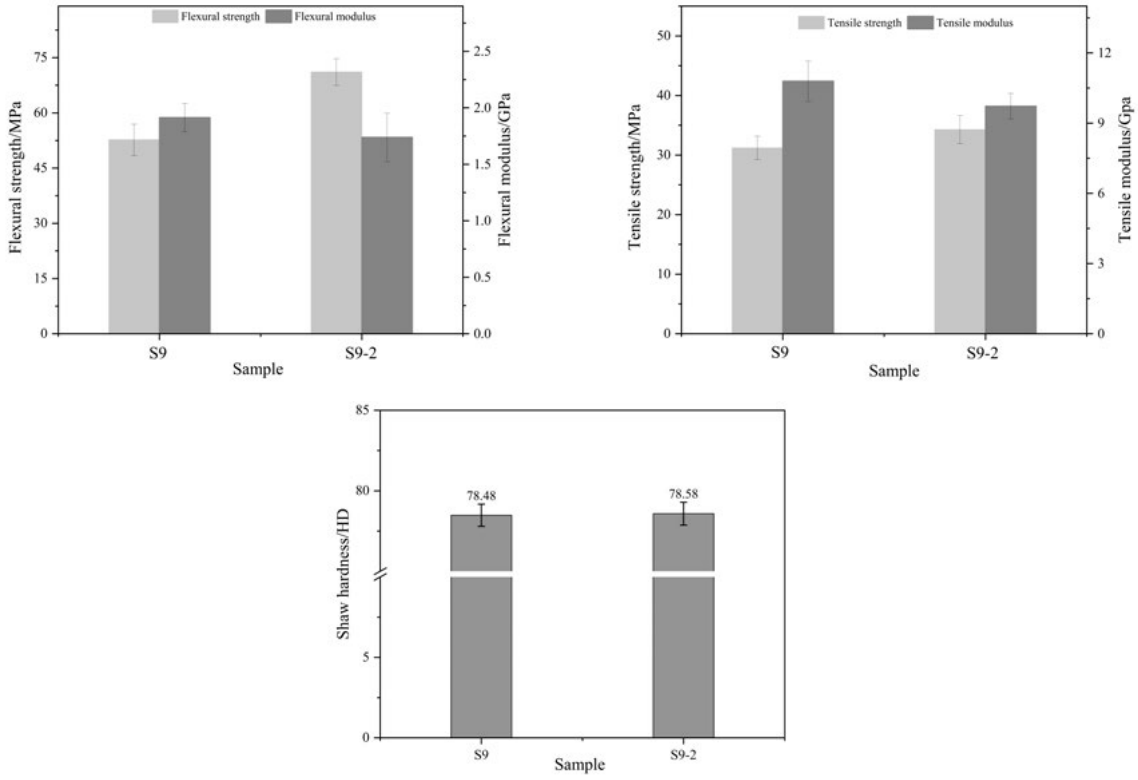


Figure 8 Effect of modification treatment on mechanical properties of composites

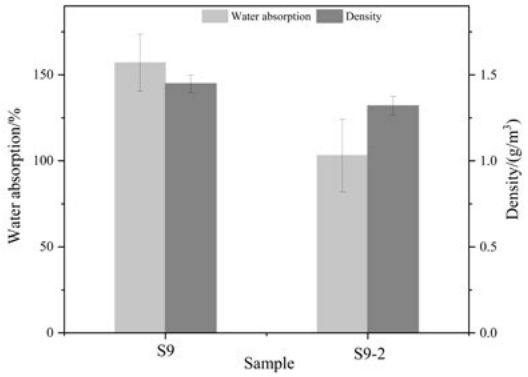


Figure 9 Effect of modification treatment on density and water absorption of composites

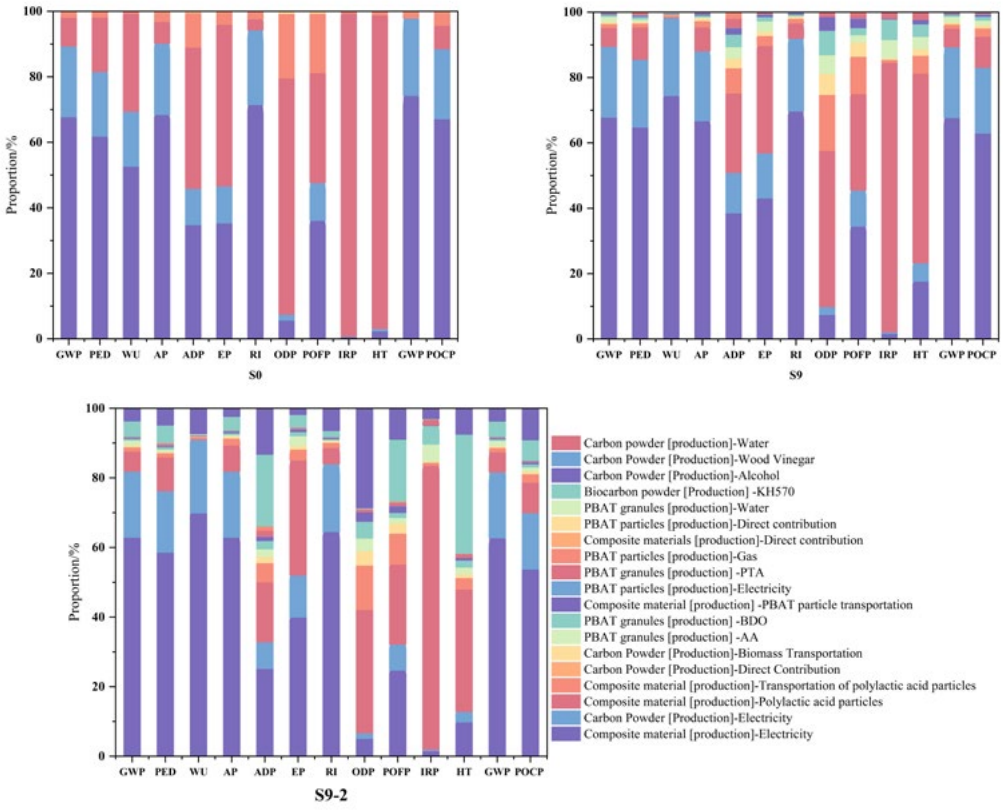


Figure 10 Cumulative contribution percentage of composite material characterization

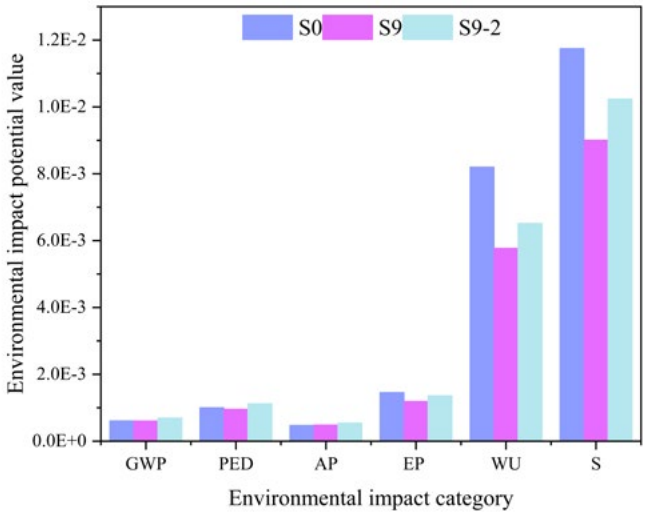
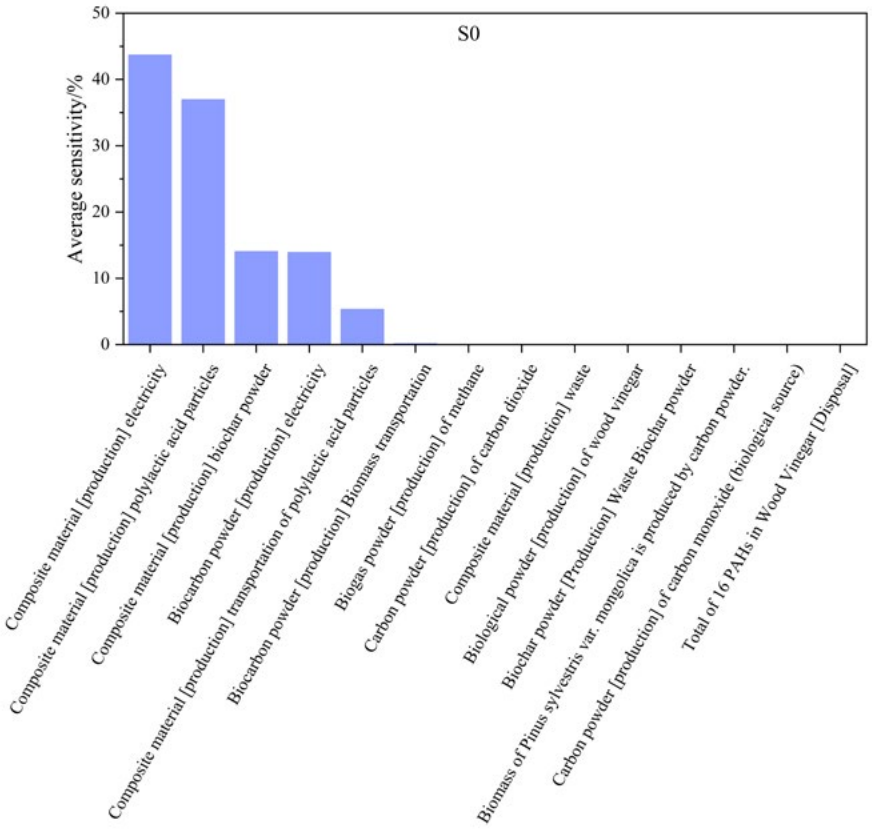


Figure 11 Normalized values of composite material



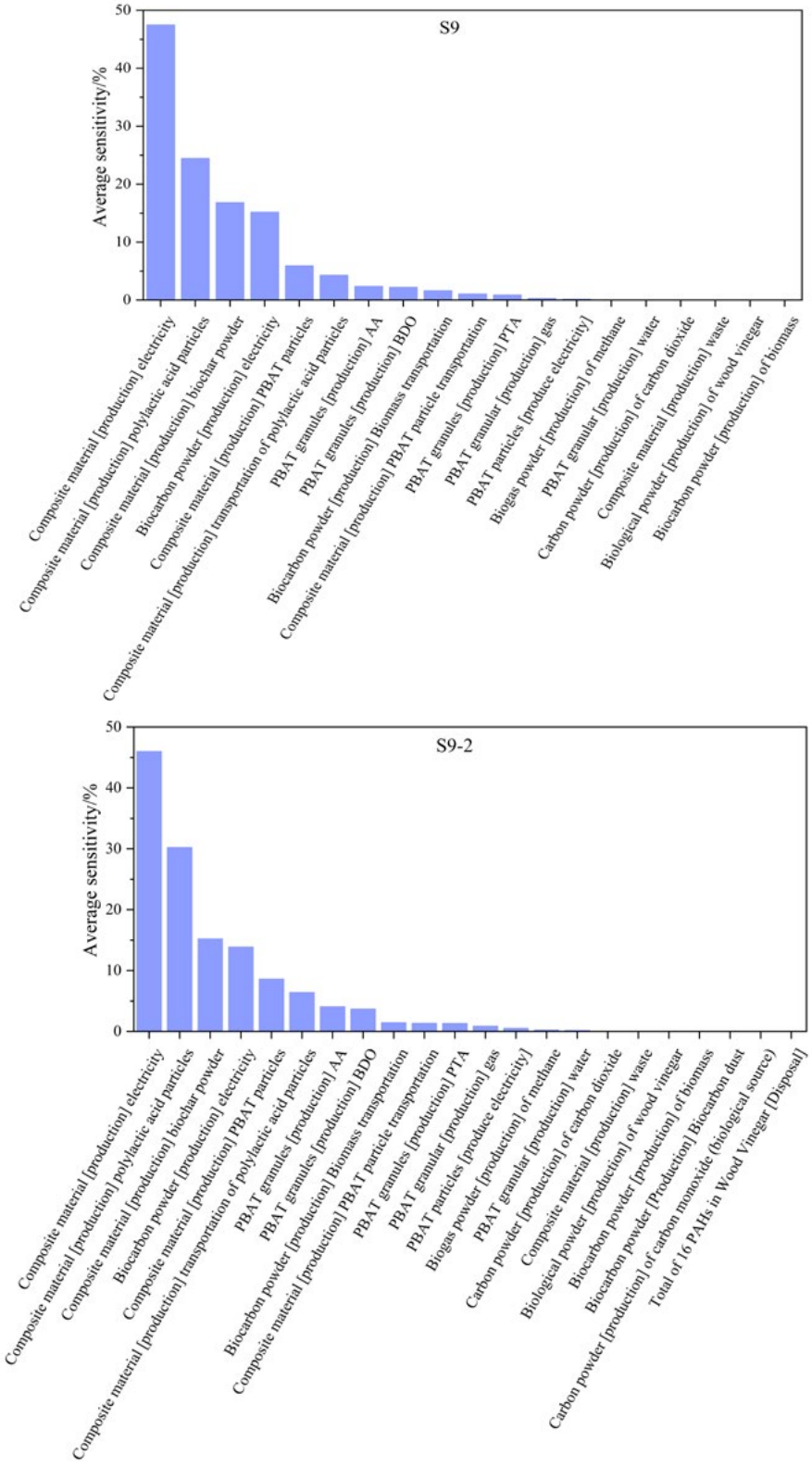


Figure 12 Sensitivity of composite materials