

Fabrication and experimental study of corn starch based vetiver cellulose reinforced bio-composite film for food packaging application

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Research Article

Keywords: Starch, Cellulose, Food packaging, Mechanical testing, Bio-composite

Posted Date: February 18th, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-202867/v1>

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Abstract

Established applicability of biocomposites in the field of automotive, construction and structural industries has led to a hope to utilize them in the near future for further applications such as food packaging and lightweight coatings. Due to their global abundance, natural fibers and starch have been the most widely tested and used raw materials for use as the bio-composite reinforcements. Here in this work, composites based on corn starch (CS) and vetiver cellulose fibers (VCF) are fabricated using the solution casting method. Alpha-cellulose was synthesized from the vetiver roots post converted into nano-cellulose using the ball milling. Various compositions of 0%, 5%, 10%, 15% and 20% (by weight) of this α -cellulose were introduced as the reinforcement to commercially available corn starch matrix. Tensile tests for the fabricated composites were done in accordance with the American Standard for Testing and Materials standard procedure. Crystallinity and failure morphology of the composites was studied using the X-Ray Diffraction techniques and scanning electron microscopy, respectively. Water absorption capacity test, Fourier-transform infrared spectroscopy analysis, and determination of contact angle of the films were also done to evaluate the film properties. Results reveal that the incorporation of vetiver cellulose fiber in the starch enhances the mechanical properties of bio-composites. Also, the water absorption capacity and the contact angle decreased and increased, respectively; thereby predicting the composites application as a lightweight food packaging material.

1. Introduction

Nanocomposites based on biopolymers have gained interest in the research community because they are sustainable and green in nature; moreover they are being employed to build a broad range of constructive products ranging from coatings to therapeutics to packaging materials [1]. In the recent years, an issue of pertinent importance has raised that with economic development there should be no harm to the property of globe. As various new merchandise (particularly plastics) are being progressively more developed, the wastes ensuing from their process, usage and disposal creates a serious threat to the atmosphere because of their non-degradability and resistance to most micro-organisms. The time needed for a lot of synthetic polymers to completely decompose is anticipated to be around hundreds of years, meaning that during the lengthy provisional natural phenomena would be influenced by the occurrence of these materials. As a result, the research community has made various attempts to devise innovative ways of fabricating green polymers, in particular from the natural sources such as starch [2–11]. Nor et al. [12] studied the mechanical properties of starch filled polypropylene (PP) under coverage of the hydrothermal conditions. The PP based composites were compounded using the Brabender Plasti-Corder machine, and the composites samples were fabricated by the compression molding technique. Their results exposed that mechanical properties of PP based composites worsen after a period of immersion time and the hygrothermal aging of composites accelerated as the temperature increased. Mousavi et al. studied on modifying the properties of polypropylene-wood composite by eggshell nano-particles and natural polymers. They showed that on adding the eggshell nano-particles and maleic anhydride instigate enhancement in the mechanical properties in conjunction with better adhesion between the corn particles,

polymer and starch. Najaf et al. [14] evaluated the mechanical properties of composites made from recycled plastics and sawdust. Results show that mechanical properties of the samples having recycled plastics were equivalent with those of composites made from virgin plastics; and this was measured as an opportunity to develop the use of recycled plastics in the fabrication of composites. Sabetzadeh et al. [15] examined the thermoplastic corn starch/low-density polyethylene (LDPE) compounds containing the maximum quantity of starch for biodegradable applications. Their results show that the apparent viscosity of blends decreases with the increase in shear rate; and the addition of starch to LDPE also results in decreasing of the melt flow index values of specimens. Kim et al [16] studied the characteristics of cross-linked potato starch and starch-filled linear LDPE films. Mechanical properties of these films were measured and compared to those of the films containing native potato starch, and results showed that the molar degree of cross-linking of the cross-linked starch increased proportionally as epichlorohydrin concentration increased; and the tensile strength, strain energy and percent elongation of the films containing cross-linked starch were superior than those of the films containing native starch. Generally, the injection molding techniques are used to manufacture the starch based reinforced composites. Ke et al. [17] worked on blending of starches and poly lactic acid, and they have prepared four dry corn starches with diverse ratios of poly lactic acid using lab scale twin screw extruder. Starch was blended with poly lactic acid and tensile bars were produced by the injection molding technique. Cellulose and starch based materials are amongst the most abundantly available organic materials on the earth because of their structural integrity and stable properties. Starch can be either used as the reinforcement material or matrix material when it is in powder form or gelatinized form. Bio-composites are made of organic biodegradable matrix and agriculture based reinforcements, and these are used for many applications like packaging, food containers and use and throw cups because these bio composites are degradable after some days in soil at particular temperature and time so it minimizes the waste on earth.

2. Experimental Methodology

2.1. Materials

Starch, sodium chlorite (NaClO_2), acetic acid (CH_3COOH) and sodium hydroxide (NaOH) were purchased from Bangalore Scientific and Industrial Supplies, India. Acetone, glycerol and distilled water were acquired from the commercial sources and utilized without any further refinement.

2.2. Preparation of Holocellulose and Alpha-Cellulose

Preparation of holocellulose was performed by the Wise et al. [18] method. Initially, the vetiver sawdust of 10 grams and distilled water of 320 mL were poured into the flask and kept on water bath at 80°C for 20 minutes. Next, 4 grams of sodium chlorite and 2 mL of acetic acid were added to solution to initiate the delignification reaction. Same process was repeated second and third time in one hour intervals. Mixture was then cooled, filtered, treated with water and acetone; and the resulting product was oven-dried. The resulting holocellulose yield was found to be 60%.

Alkaline treatment was used to adjust the hemicellulose content in the holocellulose [19]. First, 8 grams of the holocellulose were poured into 40 mL of 17.5% of sodium hydroxide solution. Next, the reactant was further reacted at 80°C for 2 hours to extract the hemicellulose [20]. The addition was done four more times in the interval of 5 minutes. The resultant product was filtered and washed with 8.3% NaOH, water, acetic acid and acetone. Sample was dried overnight and the yield of cellulose was found to be 47%.

2.3. Preparation of the Films

Starch-cellulose films were prepared by using solution casting technique; where the starch was dissolved in water with continuous stirring for 20 minutes at 90°C. The gelatinized starch solutions were adjusted with different pH values and the solution was poured on metal plate, and then dried at room temperature for 48 hours. Four different films of 5%, 10%, 15% and 20% (by weight) of cellulose based starch films were then fabricated.

2.4. Characterization methods

2.4.1. Tensile test

Tensile strength, % elongation and the elastic modulus of the films were evaluated by means of the Universal Testing Machine (INSTRON: Electroplus E1000 and E3000 test system) with sample specifications as per the ASTM D882-18 standards method [21]. The cross-head speed was maintained at 1 mm/min.

2.4.2. X-Ray Diffraction (XRD) test

The crystalline structure of films was investigated using a BRUKER AXS, Model D8® advance diffractometer with the SSD-160 detector. The diffractogram was recorded from 5° to 50° angle range at a scanning rate of 0.5° per minute [22].

2.4.3. Fourier-transform infrared (FTIR) spectroscopy

Infrared Spectra (IR) of films was measured by the FTIR spectrophotometer. The infrared spectrum was taken in transmittance mode at a resolution of 5 cm⁻¹ ranging from 500–4000 cm⁻¹.

2.4.4. Surface Morphology

Surface topology studies of all the films were studied using a Scanning Electron Microscope (SEM) (JEOL JSM-6084LV). SEM was run at an accelerating voltage of 10 kV.

2.4.5. Water Absorption test

Capacity of water absorption of the film was carried out according to the ASTM D570-98 standard [23] where the specimen were conditioned in a hot air oven for 2 hours at a temperature of 60°C and weighed (W_i). Dried films were kept in distilled water at the room temperature for 24 hours. After that, the samples were removed from the water environment, dried by wiping gently with the help of a blotting paper and weighted (W_f) to conclude the water absorbed by films. Water absorption capacity was determined by the

Eq. 1 [24–34]:

$$\% \text{ Water Absorption} = \frac{(W_f - W_i)}{W_i} * 100 \quad (1)$$

2.4.6. Contact angle measurement

Contact angle of the prepared samples were measured at the room temperature using a contact angle analyzer. A drop of water was dropped on the films and the contact angle is measured with the help of software and images were taken for 0.013 seconds to 1 second. Contact angle of CS film and VCF added films were then analyzed.

3. Results And Discussion

3.1. Tensile test

The tensile strength and % elongation of corn starch film and its biocomposites are showcased in the Figs. 1 and 2, respectively. The tensile strength of films increased with the addition of cellulose microfibrils from vetiver roots in corn starch matrix. It can be observed that the tensile strength of neat corn starch film is 3.60 MPa. When cellulose was added to the corn starch, the tensile strength value enhanced to a maximum value of 32.90 MPa for the 10 % composition, and after that the tensile strength value again started decreasing (up to 19.06 MPa for the 20% of cellulose composition). This enhanced tensile strength is mainly due to a good interaction between the cellulose fibers and starch matrix. But, due to the inhomogeneous distribution of fibers in starch matrix, tensile strength values started deteriorating (for the 15 and 20% samples). This was analyzed with the SEM micrographs, which will be discussed further in this article.

3.2. X-Ray Diffraction test

XRD pattern of corn starch and cellulose are shown in the Fig. 3. XRD of corn starch contains diffraction peaks (2θ) at 15.05° and 22.08° , and a doublet at 17.96° and 20.02° that corresponds to the A-type crystalline structure. XRD of vetiver cellulose contains diffraction peaks at 13.06° , 14.09° and 29.89° , and a doublet at 23.01° , 23.98° and 24.01° that corresponds to the B-type crystalline structure.

3.3. Fourier-transform infrared (FTIR) spectroscopy

Structural modifications and chemical functional groups of starch and cellulose are presented in the Fig. 4. These modifications can be identified by the FTIR measurements. The spectrum of pure starch film shows many sharp edges. It can be observed that a well-defined band at 1001 cm^{-1} that corresponds to symmetric stretch of -OH functional groups. A large and high band spectrum indicates the presence of moisture content in the starch (can be observed in high band at 3300 cm^{-1} that indicates symmetric and

asymmetric stretch, and overtone of band). Furthermore, it is observed that there are no significant sharp edges in the cellulose, indicating the strong and broad of CO stretch that is a cellulose alcohol group.

3.4. Surface Morphology

Figure 5 represents the fracture morphology of corn starch film and VCF bio-composites obtained from the SEM technique. Fracture morphology of neat starch film composites with 0% cellulose fiber shows a uniform, homogeneous and less porosity formation (refer to the Fig. 5a). Figure 5b shows the fractured surface of 5% composite. Herein this figure, development of non-uniform mat between fibers and matrix was revealed, which results in a relatively high tensile strength (due to the interlocking/cross-linking phenomenon) in comparison to the neat matrix composition. Figure 5c showcases the 10% composition composite, where fiber breakage phenomenon can be observed resulting from the stress that has been transferred successfully to the fiber. Hence, this configuration resulted in the highest tensile strength amongst various composites tested. After that from the Figs. 5d, 5e and 5f, tensile property started deteriorating because of poor stress transfer capability.

3.5. Water Absorption test

Water absorption property is one of the most sensitive and usefulness parameter of starch based composites. Water absorption is mainly used to study the moisture absorption by the composite material. Figure 6 shows the moisture absorption percentage of all composites for 24 hours. It can be observed that there is a significant difference between starch and cellulose based composites. The moisture absorption percentages of composites are 27.37, 23.43, 23.51, 21.27 and 20.08% for starch and its cellulose based composites. It can be seen that the addition of cellulose into starch reduces the absorption of water content. This is mainly due to the cellulose property of being an organic substance and has less hydrophilic nature. This test confirms that our fabricated composite can be readily used for the food packaging application.

3.6. Contact angle

The contact angle of CS film and VCF reinforced bio-composites are shown in the Fig. 7. CS film exhibited a low contact angle of 20.95° (refer to Fig. 7a and b), and starch reinforced with VCF composites exhibited a relatively higher contact angle of 69.01° (Fig. 7c). This indicates the addition of vetiver cellulose in the starch destroying some of the hydroxyl groups in the bio-composites and also the hydrophobic nature of the bio-composite. This also confirms the fact that why the VCF based bio-composites have the lowest water absorption.

4. Conclusions

The bio-composites based on VCF and CS matrix was successfully fabricated through the solution casting method. The presence of VCF in CS improved the tensile property and crystallinity index of

composites. The maximum tensile strength was 32.90 MPa achieved by the 10% by weight of VCF in the CS matrix sample. There is no significant change in the functional group distribution with the addition of fiber in the matrix domain. VCF based bio-composites have the lower water absorption character and higher contact angle in comparison to the neat cellulose film. This bio-composite is suggested for the food packaging application due to its excellent mechanical property, environmentally friendly behavior and less water absorption characteristic.

Declarations

Acknowledgements

“Academic support from the Presidency University, Bengaluru, India; University of Petroleum and Energy Studies, Dehradun, India and King Mongkut's University of Technology North Bangkok, Bangkok, Thailand are gratefully appreciated.”

Competing Interests

“The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.”

Funding Info

“Monetary support from the Taif University Researchers Supporting Project number (TURSP-2020/244), Taif University, Taif, Saudi Arabia is gratefully appreciated.”

Author Contribution

“All the authors contributed equally for data curation, formal analysis, methodology, conceptualization, project administration, resources, investigation, supervision and writing.”

Data Availability

“All data generated or analysed during this study are included in this published article.”

Animal Research (Ethics)

“This article does not contain any studies with human participants or animals performed by the authors.”

Consent to Participate (Ethics)

“The authors give the consent to participate this research study.”

Consent to Publish (Ethics)

“The authors give the consent to publish this research work in the journal.”

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Figures

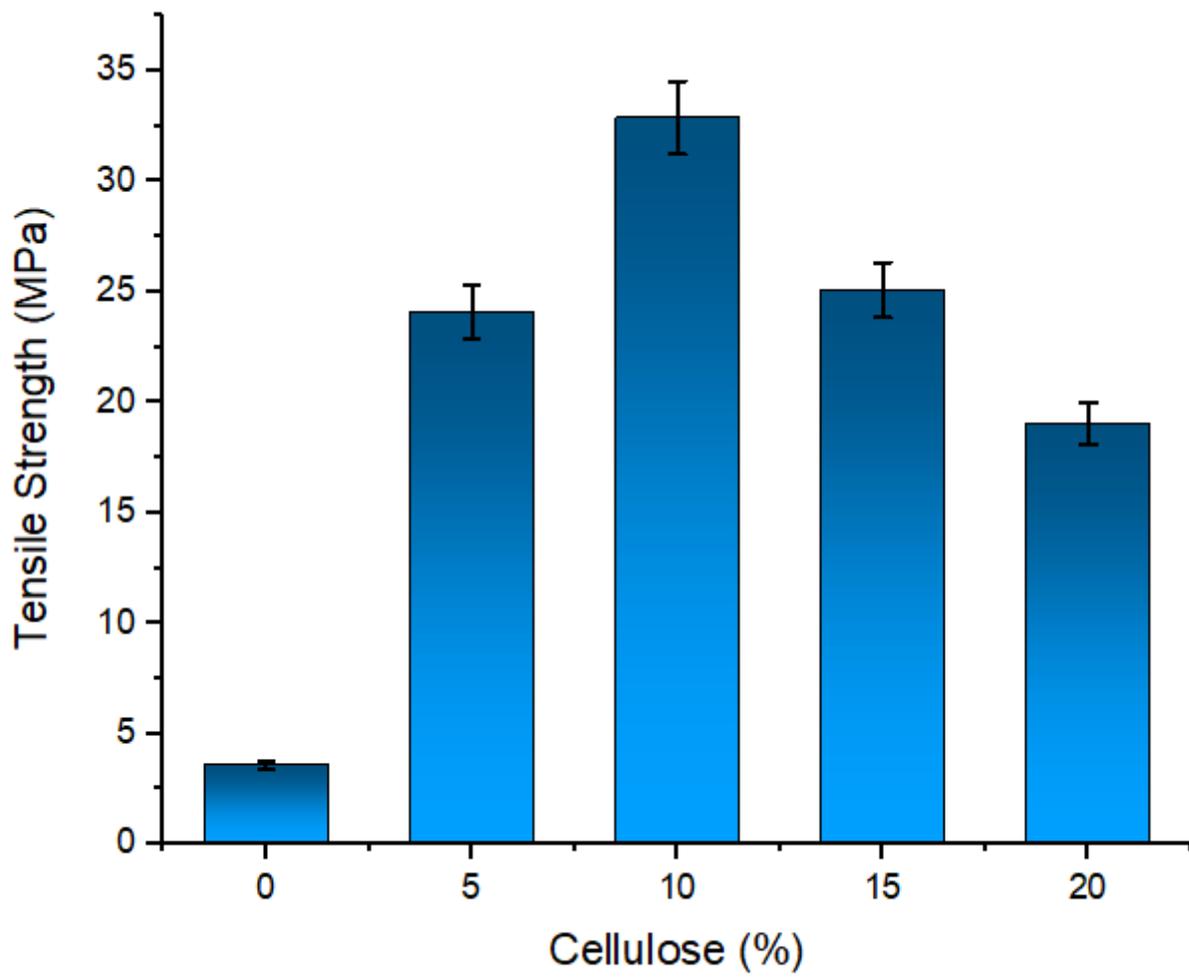


Figure 1

Variation in tensile strength values of all the composites tested

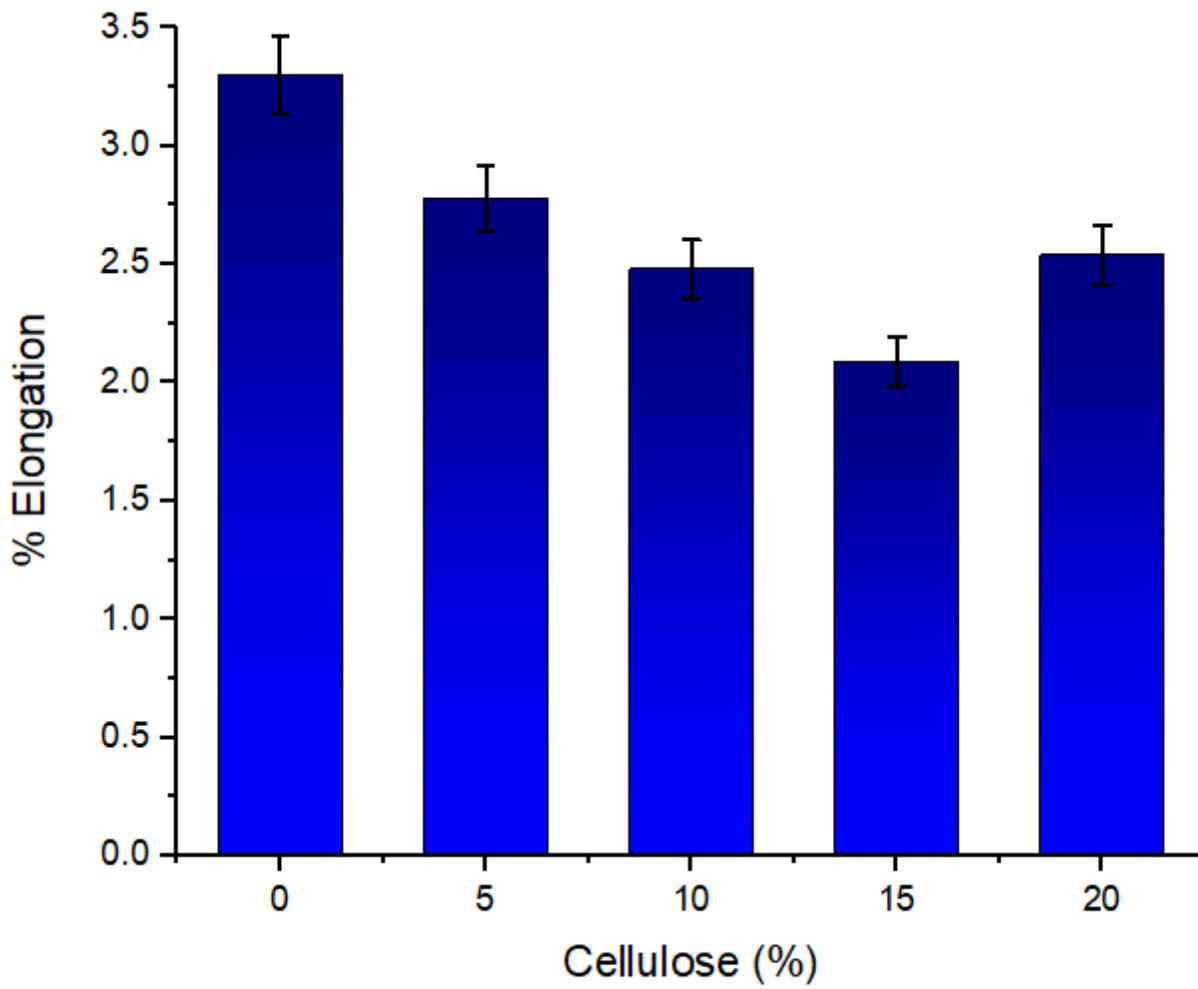


Figure 2

% elongation observed in different composites

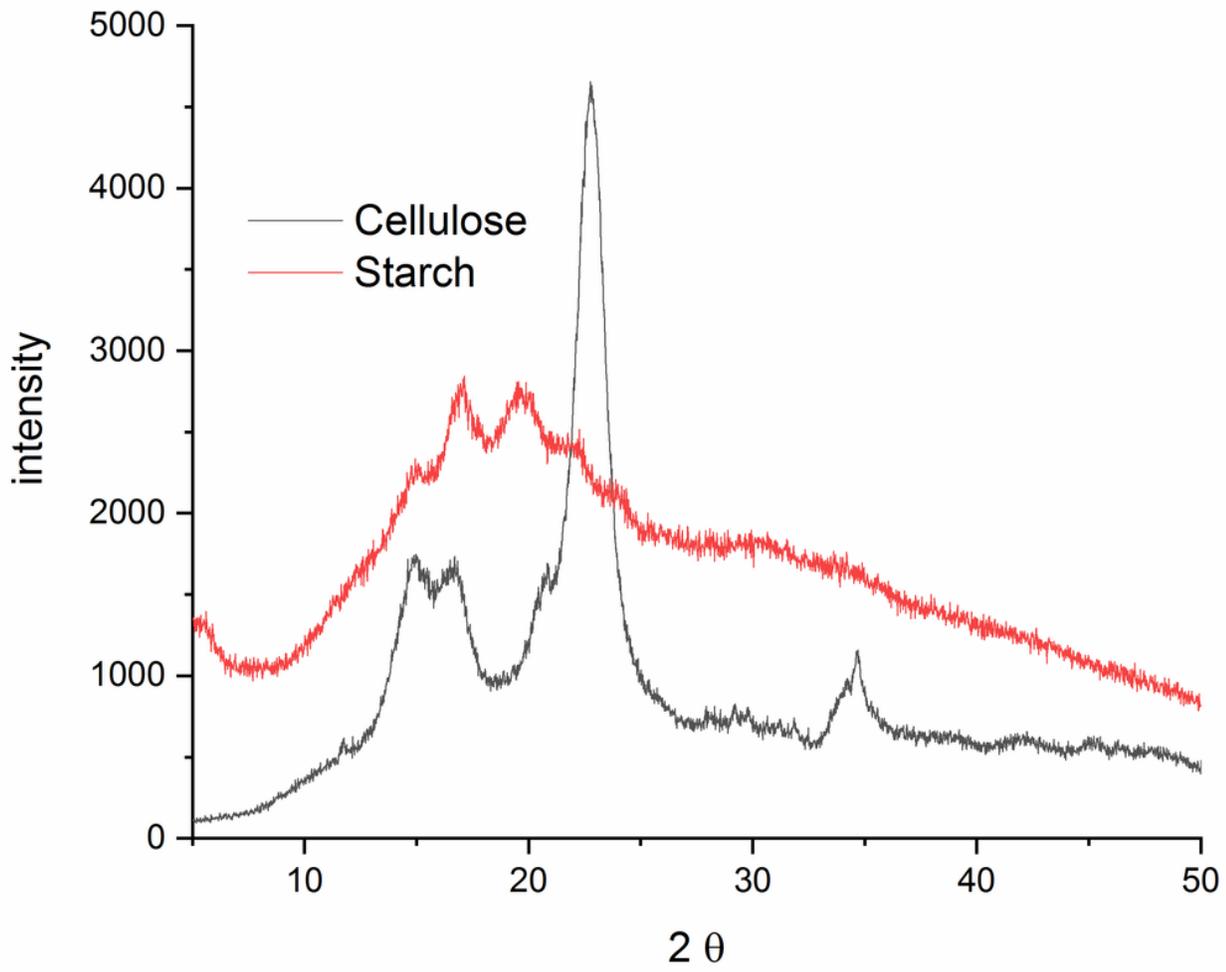


Figure 3

XRD spectra of the starch and cellulose

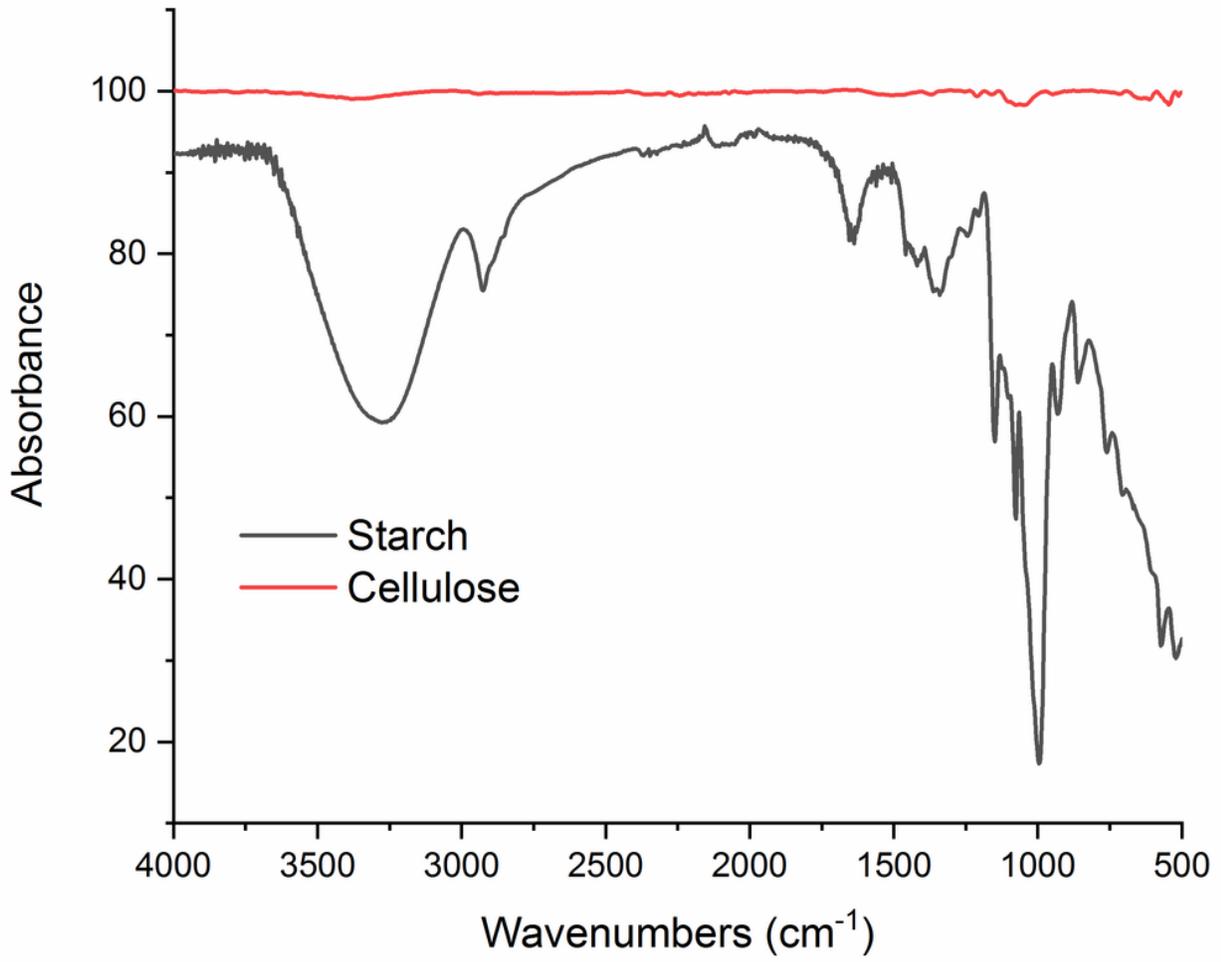


Figure 4

Fourier-transform infrared spectroscopy of Corn starch and Cellulose

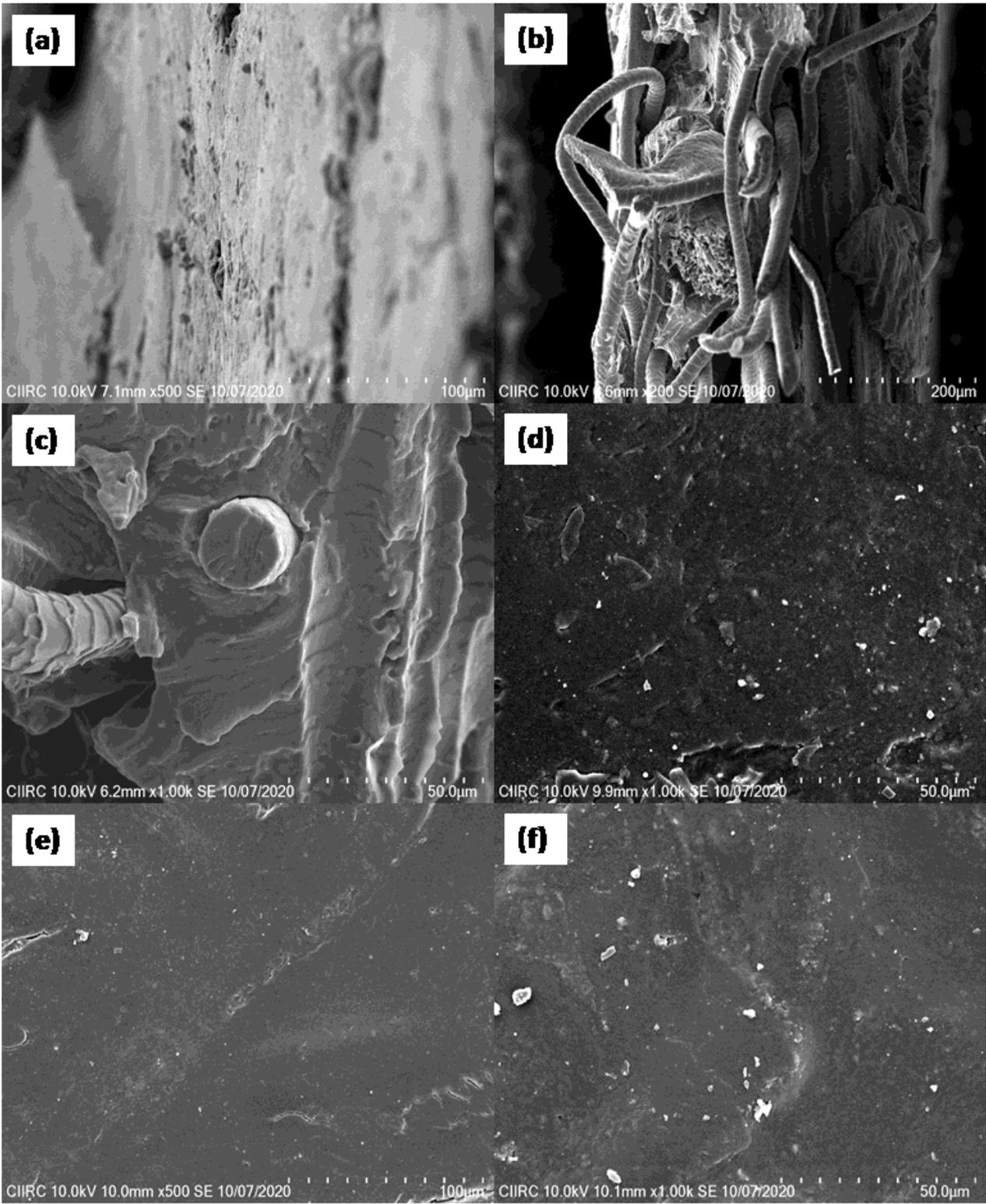


Figure 5

Fractured surfaces of CS Film and VCF bio-composites

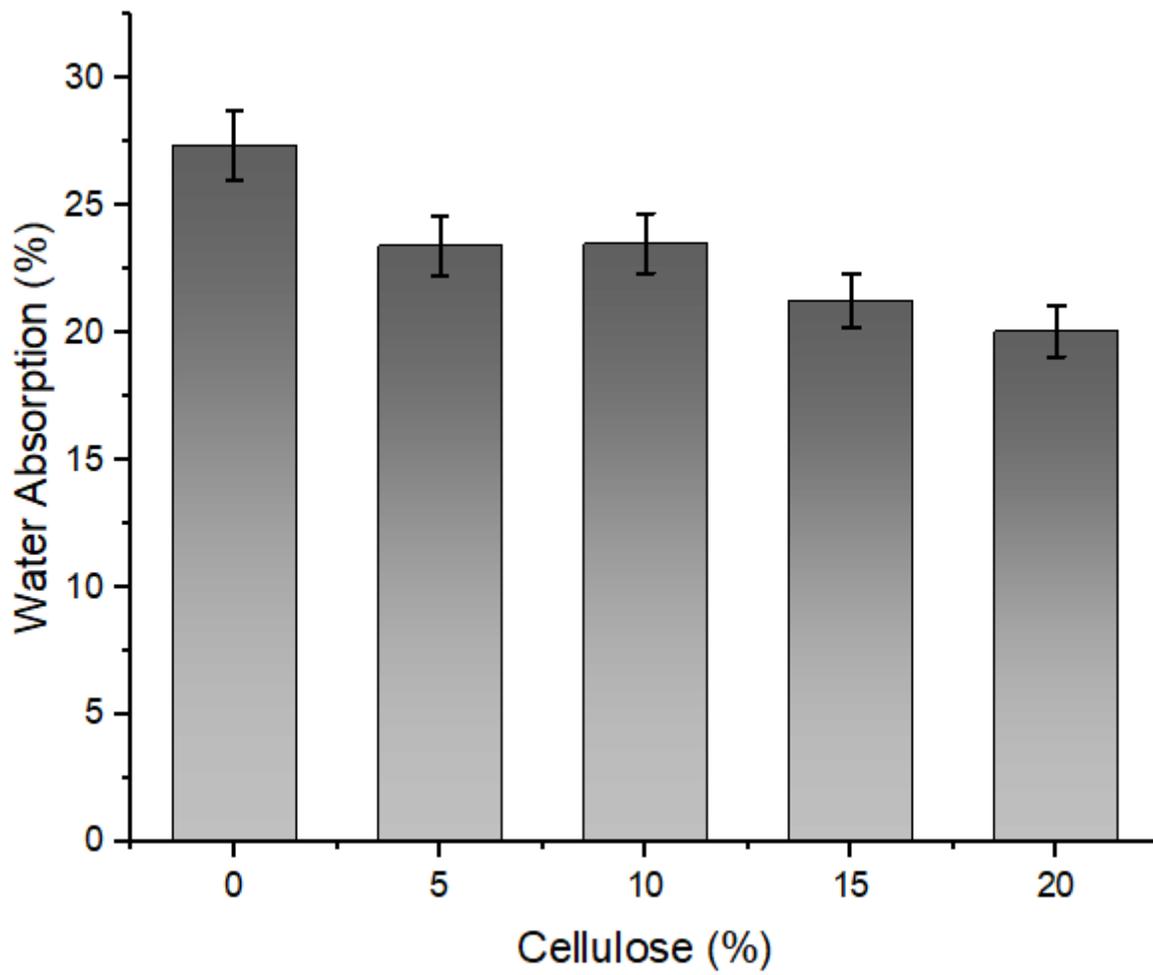


Figure 6

Moisture absorption percentage of all the biocomposites tested

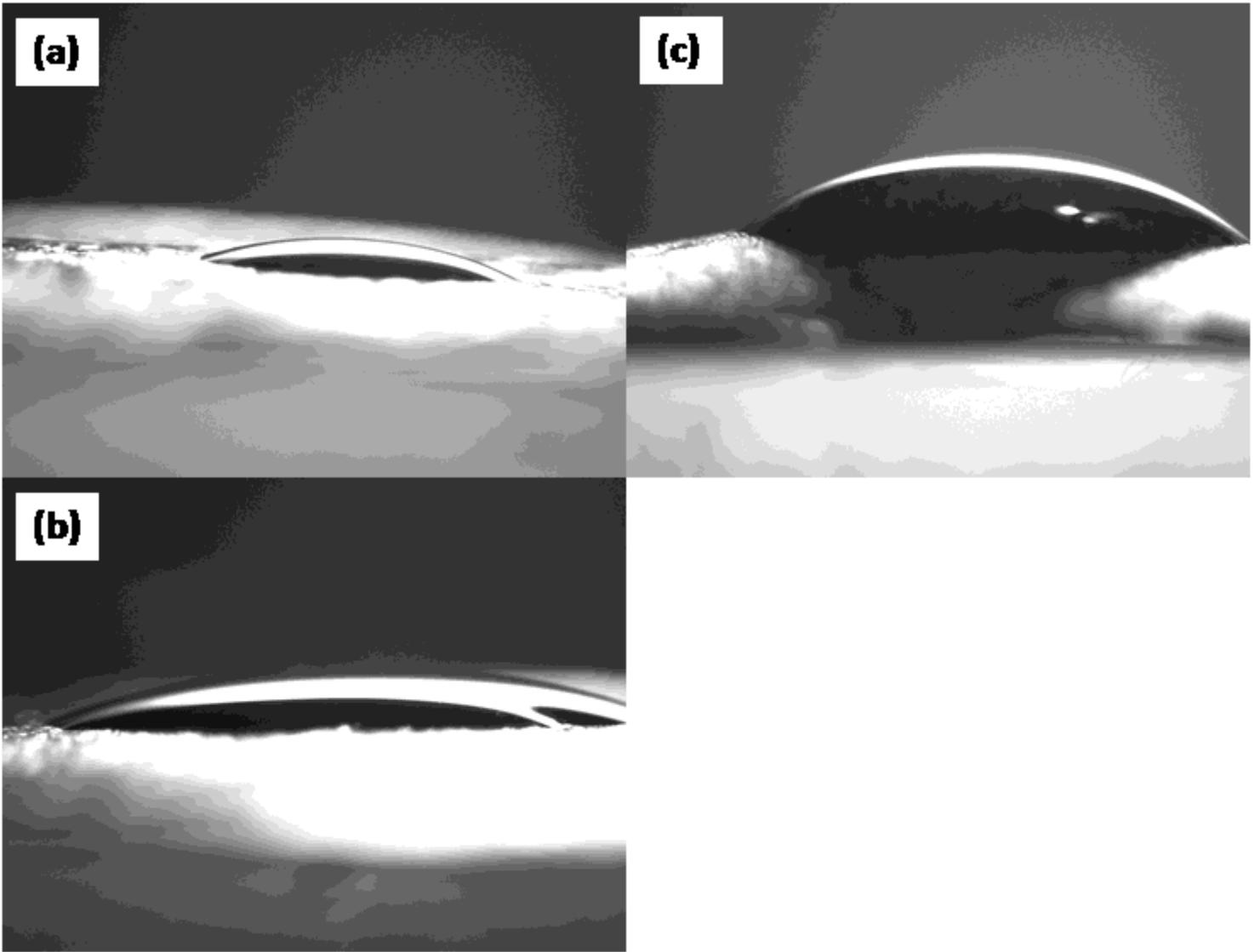


Figure 7

Images of contact angle CS film and VCF reinforced composites