

A comparative study of pine rosin and glutaraldehyde cross linker on mechanical properties of jute corn starch based biocomposite

Karishma M. Sakhare*, Suraj R. Bamane^a and Shashikant P. Borkar^b

Textile Manufactures Department, Veermata Jijabai Technological Institute, Mumbai, 400019, India

(Received April 11, 2022, Revised January 12, 2024, Accepted January 24, 2024)

Abstract. Biocomposites made up of starch and jute fibres are biodegradable and environmentally friendly materials for sustainable development. In this study, corn starch has been separately modified with 15% pine rosin and 40% glutaraldehyde, and 30% glycerol is used as a plasticizer. The composites have been prepared for three different volume proportions of matrix and jute fibre such as 60:40, 70:30 and 80:20 by using a hot compression moulding machine. The effects of pine rosin and glutaraldehyde on mechanical properties have been studied. Pine rosin modified starch jute composites have shown higher tensile and flexural properties as compared with glutaraldehyde modified starch jute composite. The highest tensile strength and modulus are found at 60:40 matrix and jute fibre volume proportion of pine rosin modified starch jute composite which are 13.97 MPa and 782.94 MPa respectively. Similar trends were found in flexural strength and modulus for pine rosin modified starch jute composite having matrix to jute fibre proportion 60:40 which are 29.18 MPa and 1107.76 MPa respectively. But, in case of impact strength, glutaraldehyde modified starch jute composite having matrix to jute fibre proportion 80:20 have shown highest impact strength that is 59.05 KJ/m². Starch-jute composite with glutaraldehyde shows 33% more water absorbency as compared to composite having pine rosin as cross linker. Highest FTIR graph indicates that the number of -OH group is much lower in case of pine rosin modified starch than glutaraldehyde modified starch which indicates that bonds formed by pine rosin are much stronger than the bonds formed by glutaraldehyde. The surface morphology of the composite was influenced by pine rosin and glutaraldehyde which is shown in the SEM image.

Keywords: biocomposite; corn; glutaraldehyde; jute fibre; pine rosin; starch

1. Introduction

Biopolymer Composites are getting much more attention in recent years compared to synthetic polymer composites. The synthetic polymer composites are nothing but petroleum-based products. Some major problems with synthetic polymers are a generation of non-degradable plastic wastes, energy-intensive processes, difficulties in recycling and limited reserves of petroleum. Due to the above-mentioned reasons, now, the researchers are looking for polymers synthesized by sustainable and eco-friendly raw materials that are generally more abundant. These led to the use of natural

*Corresponding author, Research Scholar, E-mail: kmsakhare_p18@tx.vjti.ac.in

^aResearch Scholar, E-mail: srbamane_m19@tx.vjti.ac.in

^bProfessor, E-mail: spborkar@tx.vjti.ac.in

fibres and matrices for the fabrication of polymer composites (Vilaseca *et al.* 2007, Torres *et al.* 2007). The polymer composites made of natural fibres and biopolymers are named “biocomposite” (Dicker *et al.* 2014). Biocomposites are environmentally friendly, biodegradable, lightweight, flexible, renewable, better sound insulator, skin friendly, processing friendly and economical (Kandpal *et al.* 2015). Natural fibres such as sisal, coir, jute, hemp, kenaf, ramie, flax, bamboo, pineapple and banana can be utilized in the fabrication of biocomposite. The advantages of natural fibres include lightweight, high flexibility, renewability, non-abrasive, combustible, good thermal properties, non-toxicity, biodegradable etc. (Gupta *et al.* 2015). Natural fibre composites have been found applied in aerospace, sports, auto industry, building and construction industry, panels, windows frame, bicycle frame, decking, furniture, etc. (Shinoj *et al.* 2011). The major constituents of natural fibres are cellulose, hemicelluloses, lignin, pectin, and different waxy substances. Most of the natural fibres absorb moisture from the surrounding which causes weak bonding between matrix and fibre (Mohammed *et al.* 2015). Among many natural fibres, jute fibre is easily available as well as economical. Jute fibre having cellulose and lignin as main constituent part, it is a very much demanding material, especially in the clothing sector. Jute fibre structure consists of 60%-64% cellulose, 14%-16% hemicelluloses, 12%-14% lignin, and other components such as pectin, fats, water, ash content, etc. (Chand and Fahim 2008). Jute fibres have relatively high strength and stiffness with lower density which make it more suitable for the fabrication of reinforced composites. Along with good mechanical properties, jute fibres also have good thermal and acoustic insulation properties. It has moderate moisture regain but no skin irritation (Chand and Fahim 2008, Aly-Hassan 2015). Jute fibres had been found in various advanced composite application such as jute/coir composite boards, thermoplastic composite-based synthetic wood, jute glass composite components for railway coaches, etc (Alam *et al.* 2014). Therefore, jute fibre as reinforcement and biopolymer as matrix are used to develop cost-effective and environmentally green composite with better physical properties (Biswas *et al.* 2013, Gupta *et al.* 2015).

Based on origin, the biopolymer can be classified into two categories such as natural biopolymer and synthetic biopolymer. Synthetic biopolymers are developed by microbial fermentation and biotechnological production while natural biopolymers are developed from proteins, microbes and polysaccharides. Polysaccharides manufactured from monosaccharides have glycosidic linkages; Starch, alginate, cellulose, dextrin, hyaluronic acid (HA), chitin and chitosan and various gums are examples of polysaccharides. Soy and gelatin are protein-based, and PHB and PLA are microbes-based polymer (Aliyaa *et al.* 2021).

One of the main promising polymers for the production of biocomposite is starch. It is a carbohydrate-based renewable, natural, and inexpensive polymer gained from a variety of different plant sources such as corn, wheat, rice, and potato (Iman and Maji 2012). The corn plant is one of the main sources of commercial starch available, and the maize granule contains more than 70% of starch along with protein, oil, sugar, and ash. Corn starch is constructed with a semi-crystalline polymer which consists of a mixture of linear polysaccharide amylose and an extremely branched polysaccharide amylopectin. Among all varieties of starch, 80% is corn starch (Ibrahim *et al.* 2019). The native starch is a carbohydrate that includes a large number of separate and partially crystalline grains of glucose units linked together by covalent bonds that associate with the glucose molecule to other groups; this bond is named as glycosidic bond. Generally native starch has a crystallinity of 20-45% (Popescu *et al.* 2010). By doing thermal processing and addition of some amount of plasticizer, native starch can be converted into thermoplastic starch. The heating process in presence of a plasticizer is utilized to interrupt and convert the semi-crystalline structure of starch grains to a uniform and amorphous product known as thermoplastic starch (Vazquez and Alvarez 2009).

Table 1 Different composition of composites

Codes for Sample	Matrix Fibre Volume Proportion	Cross Linker
SJ60	60:40	No cross linker
SJ70	70:30	
SJ80	80:20	
SJP60	60:40	Pine Rosin
SJP70	70:30	
SJP80	80:20	
SJG60	60:40	Glutaraldehyde
SJG70	70:30	
SJG80	80:20	

Plasticizer also improves mechanical properties and processing capability (Singha and Kapoor 2014). Starch-based films can be alternatives of different polyethene in packaging well as they can be widely used in medical delivery systems and devices. But, because of the hydrophilic nature and lower mechanical strength, its application is limited (Kaur *et al.* 2007).

Cross-linking is a low cost and effective method to improve the physicochemical properties of composites. Sun *et al.* (2018) prepared cross-linked starch/PHA composites by various cross-linking agents which were citric acid, adipic acid, borax, and boric acid. Murshid Iman *et al.* used glutaraldehyde, as a cross-linking agent to prepare starch and jute fabric-based composite (Iman and Maji 2012). Baishya and Maji (2014) cross-linked starch-based wood composite with three different cross-linker such as glutaraldehyde (GA), dimethylol dihydroxy ethylene urea (DMDHEU), and N-methylol acrylamide (NMA). These authors suggested that improvement in properties such as mechanical, thermal, water resistance, and chemical properties could be possible with the addition of cross-linking agents. Pine rosin is natural material which is extracted from pine tree can be used as crosslinking agents in the formation of biocomposites. It is brittle at room temperature and softens during heating. Pine rosin is a mixture of volatiles and non-volatiles components secreted from pine trees. When the volatile resin components have been distilled off, the remaining solid material is pine rosin. Pine rosin is semi-transparent mass and readily fusible with a glossy appearance, which varies in colour from yellow to black. It is not soluble in water but soluble in chloroform, ether, alcohol, carbon disulphide, glacial acetic acid, many fixed and volatile oils, and light petroleum (Sousa *et al.* 2019). Pine rosin is composed of 90% resin acids and 10 % neutral compounds such as terpenic, esters, hydrocarbons, aldehydes, and alcohols. In resin acids, 90% are isomeric abietic acids and, the remaining 10% are dehydroabietic acid (Aldas *et al.* 2020). Pine rosin has several advantages like low cost, good electrical insulation, ease in converting to high-performance molecular materials, good water resistance and antimicrobial property (Mahendra 2015). It is used in paper sizing, printing inks, surface coatings, pharmaceuticals, adhesives, and rubber additives along with advanced applications in the biomedical and construction industry (Ribeiro *et al.* 2018).

In this study, jute fibre has been used as reinforcement and modified corn starch as a matrix. Starch has been separately modified with 15% pine rosin and 40% glutaraldehyde, and 30% glycerol is used as a plasticizer. The composites have been prepared for three different volume proportions of matrix and jute fibre such as 60:40, 70:30 and 80:20. The effect of cross-linkers (pine rosin and glutaraldehyde) on the physicochemical properties of different proportions of composites has been studied.

2. Experimental

2.1 Material

Corn Starch Soluble Extra Pure was obtained from Loba Chemie Pvt. Ltd. Jute fibre strands were purchased from the Central Research Institute for Jute & Allied Fibres, Kolkata, West Bengal, India. Glutaraldehyde of concentration 25% (w/v) was provided by Merck Private Limited (Mumbai, India), Glycerol was purchased from Qualigens Fine Chemicals, (Mumbai). Pine Rosin in solid crystal form was obtained from M/s. Seba Associates Bhubaneswar Odisha. Codes for samples of a different matrix to fibre volume proportions have been shown in Table 1.

3. Methods

3.1 Preparation of jute fibre and pine rosin

At first, received jute strands had been cut of predetermined length 20 mm by scissor, afterwards these cut fibres were opened in the opening machine to remove its trash and obtained separated fibres so that these fibres can be uniformly mixed in the resin, then fibre was conditioned at room temperature for 24 hours. Since the received pine rosin was solid and brittle so it was converted into powder form by using a regular grinder.

3.2 Preparation of the starch paste

In the current research three types of composites were fabricated. For first type of composite, starch was mixed with 3 times of water calculated based on the weight of starch and fibre. Temperature for the mixture was kept between 60-70 °C and continues stirring with a magnetic stirrer for 20 min to avoid lumps formation in the mixture. 30% of glycerol on the weight basis of a matrix was added to the mixture and stirred again for 10 min immediately after that; the solution was transferred into a heated water bath where the temperature of the water bath was kept at 75-80 °C while stirring continuously till the gelatinization of the starch obtained and the solution became more viscous. Further, the starch paste was allowed to cool and this paste was used as resin for fabricating the composite. For preparing the second type of sample similar process was used to make starch paste and 15% pine rosin powder on the weight basis of starch was added to the prepared paste and mixed uniformly by using a glass rod. And, for producing third type of samples starch paste was got ready in similar manner as first type then 40% glutaraldehyde on the weight basis of starch was added to the mixture of starch paste.

3.3 Preparation of composite

After preparation of resin, measured weights of chopped and opened fibres were slowly added manually to the resin and then the mixture was stirred manually to maintain uniform distribution of fibres in the matrix. This mixture was kept for 24 hr at room temperature to improve its viscosity. Then resin/fibre mixture was transferred into mould (200 mm×200 mm×5 mm) to make a uniform sheet of composite. The mould was kept in a compression moulding machine and the pressure was gradually applied to avoid flowing out resin/fibre mixture. The mould is kept at 80 °C for 30 min for

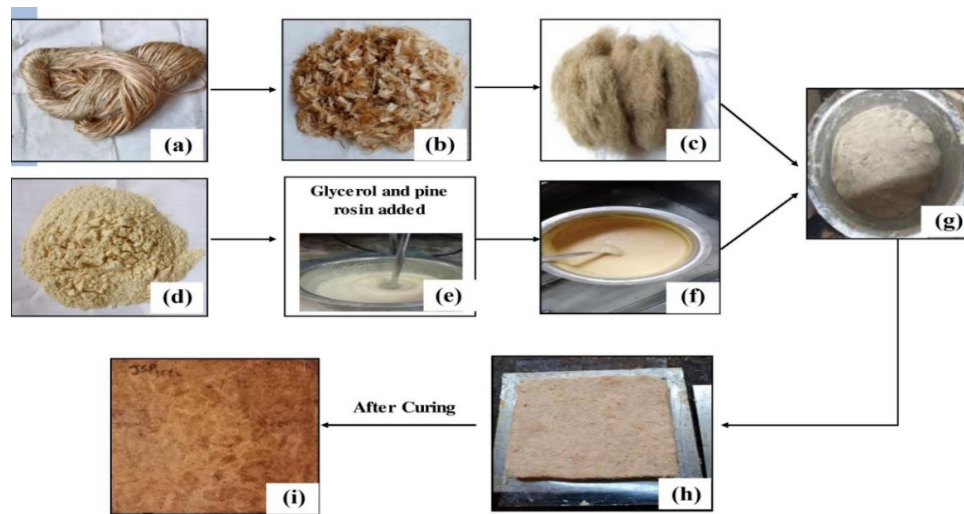


Fig. 1 Fabrication route of biocomposite (a) Strand of jute fibre, (b) Chopped jute fibre (20 mm), (c) Opened jute fibre, (d) Starch Powder, (e) Preparation of Resin, (f) Resin ready for use, (g) Resin/fibre mixture, (h) Pre-cured biocomposite sheet on mould and (i) Biocomposite after Curing

drying resin/fibre sheet. Curing was done at 120 °C for 3 hr under a pressure of 50 kg/cm². Route for fabrication of biocomposite, shown in Fig. 1.

4. Characterizations

4.1 Tensile testing

For tensile testing, a specimen of size 115 mm×19 mm×4 mm was cut from composite according to ASTM D638-03. A cross-head speed of 2 mm/min and a gauge length of 65mm were used for carrying out the test. Five replicate specimens, each of all variants were tested at 23 °C temperature and 55% RH, and mean values were reported. Tensile properties of composites were determined using Universal Testing Machine (UTM).

4.2 Flexural testing

For flexural testing, specimens were prepared according to ASTM D790-07. The 3-point bending method was adopted as specified in the standard. Specimen with the nominal dimensions of 120 mm×12.7 mm×4 mm having a span length of 80 mm and a crosshead speed of 2mm/min were used. Five replicate specimens, each of all variants were conditioned and tested at 23 °C temperature and 55% RH. Tests were performed on the same Universal testing machine (UTM) as used in tensile testing.

4.3 Impact testing

This test was performed on the Izod Charpy Impact Tester of making the Avery-Denison Impact

meter; a 2.7-joule striker was used. The specimen dimensions were 63.5 mm×12.7 mm×4 mm and 'v' notch of depth 2.54 mm and notch angle 45° as per ASTM D 256-6a was made by notch cutter at the middle of the face having dimension 63.5 mm×4 mm. Five replicate specimens, each of all variants were conditioned and tested at 23 °C temperature and 55% RH. The impact strength in KJ/m² was calculated by dividing the recorded absorbed impact energy by the cross-sectional area of the specimen.

4.4 Water absorption test

Water absorption tests were carried out according to ASTM Designation: D570-98 (Reapproved 2005), Standard Test Method for Water Absorption of Plastics. Specimens of each formulation were selected and dried in an oven for 24 hours at 50 °C. The dried specimens were later weighed to a precision of 0.001 gm and were placed in distilled water and kept at room temperature for 24 hours. After 24 hours, they were removed from the water one at a time, all surface water was wiped off using a dry cloth and the equilibrium weight value was determined after 24 hrs soaking in water at room temperature. Results were presented as percent water absorption in relation with the dry weight of the specimens. Percentage increase in weights during immersion were calculated to the nearest 0.01 %.

4.5 FTIR

The FTIR spectra were recorded using a Spectrum 100 Spectrophotometer infrared spectrometer equipped with a Smart Endurance single bounce diamond ATR accessory (Perkin Elmer, USA). Using this system, small pieces of polymer film were brought directly into contact with the ATR objective crystal using slight pressure in order to collect the spectrum. Spectra were collected in the spectral range of 4000 to 525 cm⁻¹, using 64 scans at a resolution of 4 cm⁻¹. The absorption spectra of the samples obtained were corrected against a background spectral scan that was first performed and subtracted from the FT-IR spectrum. Spectra were manipulated and plotted with the use of the Galactic 187 Industries Corporation GRAMS/32 software package (Salem, NH, USA). The final FTIR spectra were normalised with respect to the distinct peak located at 1150 cm⁻¹ since this peak was the least changed among the FTIR spectra for the composites.

4.6 SEM

The surface morphology and fracture surface of the tensile specimen were studied by using a scanning electron microscope (Philips XL-30 SEM, The Netherlands). Samples were recorded with the microscope operating at 10 kV; samples were coated with a thin layer of gold before recording microphotographs.

5. Results and discussion

5.1 Tensile testing

The effects of different cross-linker and matrix to fibre proportion on tensile properties are shown in Fig. 2. It is observed that the addition of pine rosin and glutaraldehyde show significant

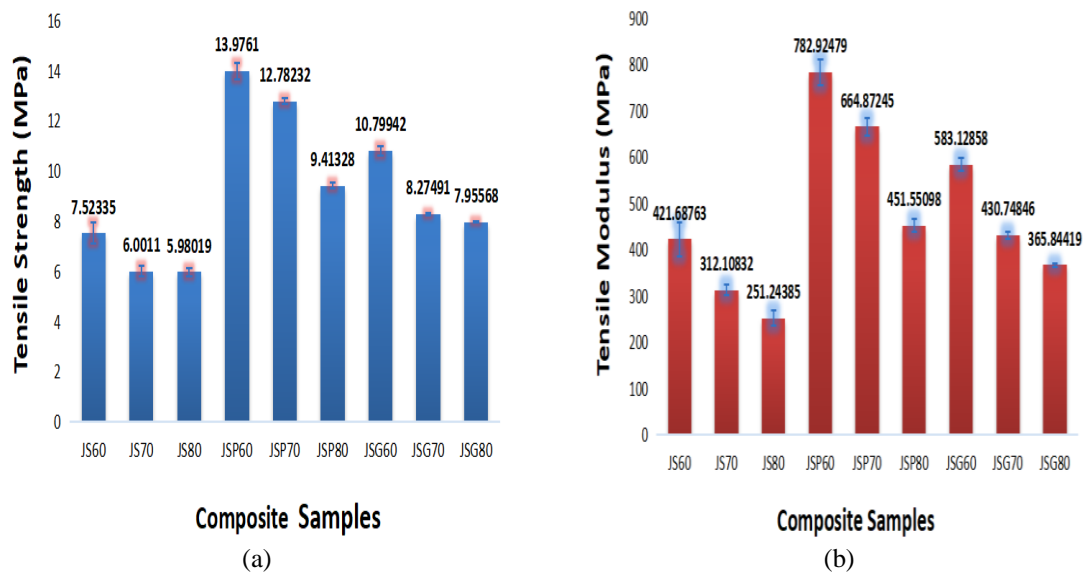


Fig. 2 (a) Tensile strength of composites and (b) Tensile modulus of composites

improvement in the tensile properties of a composite. In the case of glutaraldehyde modified starch jute composite, crosslinking between glutaraldehyde and starch or between glutaraldehyde and jute fibre may be responsible for enhancing tensile properties. The Aldehydic group of glutaraldehyde may be the main component for the formation of crosslinking between glutaraldehyde and starch or between glutaraldehyde and jute fibre (Iman and Maji 2012 and Baishya and Maji 2014).

Pine rosin has major content of rosin acids which consist of abietic acid. Abietic acid comprises the carboxylic group (Aldas *et al.*2020). In the case of pine rosin modified starch jute composite, improvement in tensile strength and tensile modulus may be due to the formation of new hydrogen bonds between starch and pine rosin. As compared to glutaraldehyde modified starch jute composite, pine rosin modified starch jute composite showed 35-45% higher tensile strength and modulus; it may be due to more density of crosslinking between pine rosin and starch. In addition, a higher molecular weight (302.44 g/mol) of pine rosin may also be responsible for increased tensile strength and modulus (Sharma and Singh 2016). It was also noticed that the tensile strength and modulus increased with an increase in fibre volume fraction in composites. It may be due to an improvement in the interfacial bond between fibre and resin.

5.2 Flexural testing

The flexural strength and modulus values of composites are presented in Fig. 3. The flexural strength and modulus of pine rosin modified starch jute composites are showing 84%, 73% and 42% for composites having fibre volume fraction 40%, 30% and 20% respectively while glutaraldehyde modified starch jute composites are showing 41%, 24% and 13% improvement in flexural strength and modulus for composites having fibre volume fraction 40%, 30% and 20% respectively when compared to the composite without crosslinking agent. When trends for flexural strength and modulus are compared with previous trends of tensile strength and modulus these appear similar. Pine rosin modified starch jute composite composed of fibre volume fraction 40% having highest

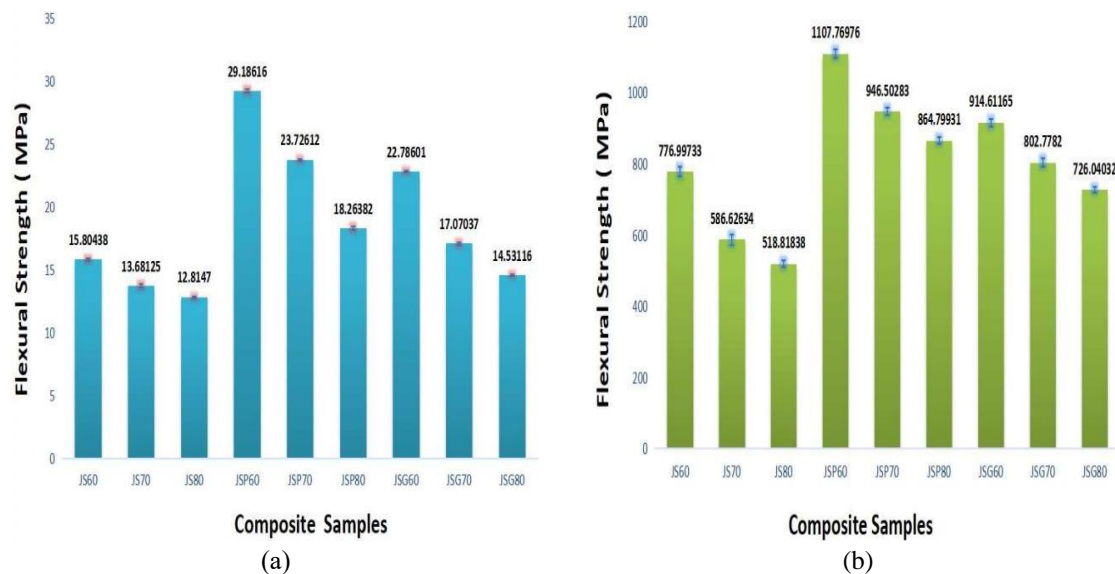


Fig. 3 (a) Flexural strength of composites and (b) Flexural modulus of composites

flexural strength and flexural modulus i.e., 29.18 MPa and 1107.76 MPa respectively while glutaraldehyde modified starch jute composite having same fibre volume fraction showing flexural strength 22.78 MPa and modulus 914.61 MPa respectively. The increased flexural strength and modulus may have been due to increased fibre-matrix adhesion. Better fibre matrix adhesion provides increased stress transfer between them (Rahman *et al.* 2008). Adhesion of fibre-matrix increases due to good interfacial bond between them. As compared to glutaraldehyde modified starch jute composite, pinoresin modified starch jute composite showed higher flexural strength and modulus it may be due to the formation of more number of hydrogen bonds between pine rosin and starch. In addition, a higher molecular weight of pine rosin may also be responsible for increased flexural strength and modulus.

5.3 Impact testing

The impact strength values of composites are presented in Fig. 4. The impact strength of the fibre reinforced polymeric composites depends on the nature of the matrix, fibre and fibre-matrix interface. The impact strength increased with a decrease in fibre volume fraction it may be due to better orientation of fibre in a longitudinal direction as compared to composite with higher fibre volume fraction. As shown in Fig. 4, the impact strength improved with the addition of glutaraldehyde and pine rosin. Glutaraldehyde gives the highest impact strength in every fibre proportion of fibre matrix composite it might be due to the liquid nature of glutaraldehyde which helps in the higher orientation of fibre as compared to pine rosin. Brittle materials absorb very little energy and brittle material shows more growth of crack that result in lower impact strength. Starch- jute based composite without any additives will be having more moisture content in the mixture of matrix and fibre due to more -OH group in their structure which may lead to more void in composite. Higher void will result in lower impact strength of composite. As previous study by Pradeep (2018) it was revealed that jute fibre reinforced epoxy composite gives better results that is 76 KJ/m² in comparison with

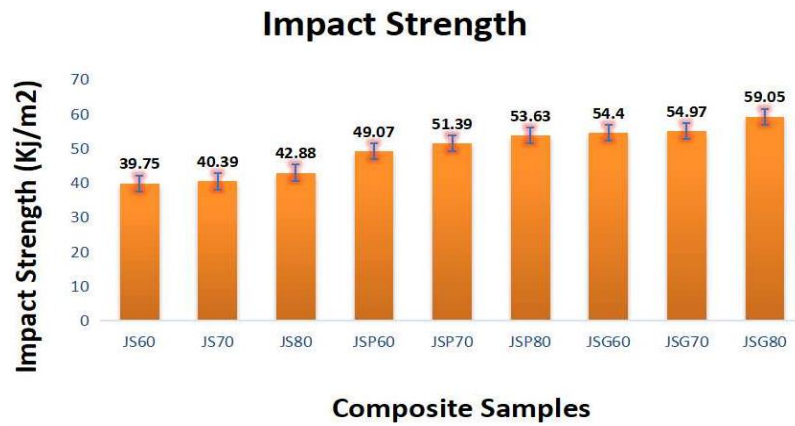


Fig. 4 Impact strength of composites

Table 2 Water absorbency of composites

Composite Samples	Matrix Fibre Proportion	Cross Linker	Conditioned Weight (gms)	Wet Weight (gms)	Absorbency %
SJ60	60:40	No cross linker	2.098	3.101	47.81
SJ70	70:30		1.843	2.705	46.77
SJ80	80:20		2.012	2.887	43.49
SJP70	60:40	Pine Rosin	1.898	2.487	31.03
SJP70	70:30		2.036	2.654	30.33
SJP0	80:20		2.00	2.604	30.18
SJG60	60:40	Glutaraldehyde	1.964	2.757	40.35
SJG70	70:30		2.046	2.846	39.10
SJG80	80:20		1.659	2.317	39.62

polyester resin it is 53 KJ/m². In another work, it was found that jute fibre reinforced soy protein isolate resin showed impact strength of 38.34KJ/m² while jute fibre reinforced composite with pine rosin modified soy protein isolate have shown better results that is 47.78 KJ/m² when 15% pine rosin was used as cross-linking agent (Sakhare and Borkar 2022). Vilaseca *et al.* worked on starch and jute strand based biodegradable composite and found that at 20% fibre loading sodium hydroxide treated jute fibre gives better results that is 13.2 KJ/m² as compared to untreated jute fibres that is 11.4 KJ/m². In case of present research, it has been exhibited that the composite having jute fibre as reinforcement and glutaraldehyde modified starch as matrix having impact strength 59.05 KJ/m² at 40% proportion of glutaraldehyde which is quite acceptable if compared with previous researches.

5.4 Water absorption test

Water absorption property of composite samples (Table 2) exhibits that the starch-jute composites with pine rosin as cross linker resist the water most while starch -jute composite without any additives has worst to resist the water. Pine rosin cross linked starch-jute composite comprising of 20% fibre volume fraction has lowest water absorbency that i.e., 30.18 % which is 30 % lower

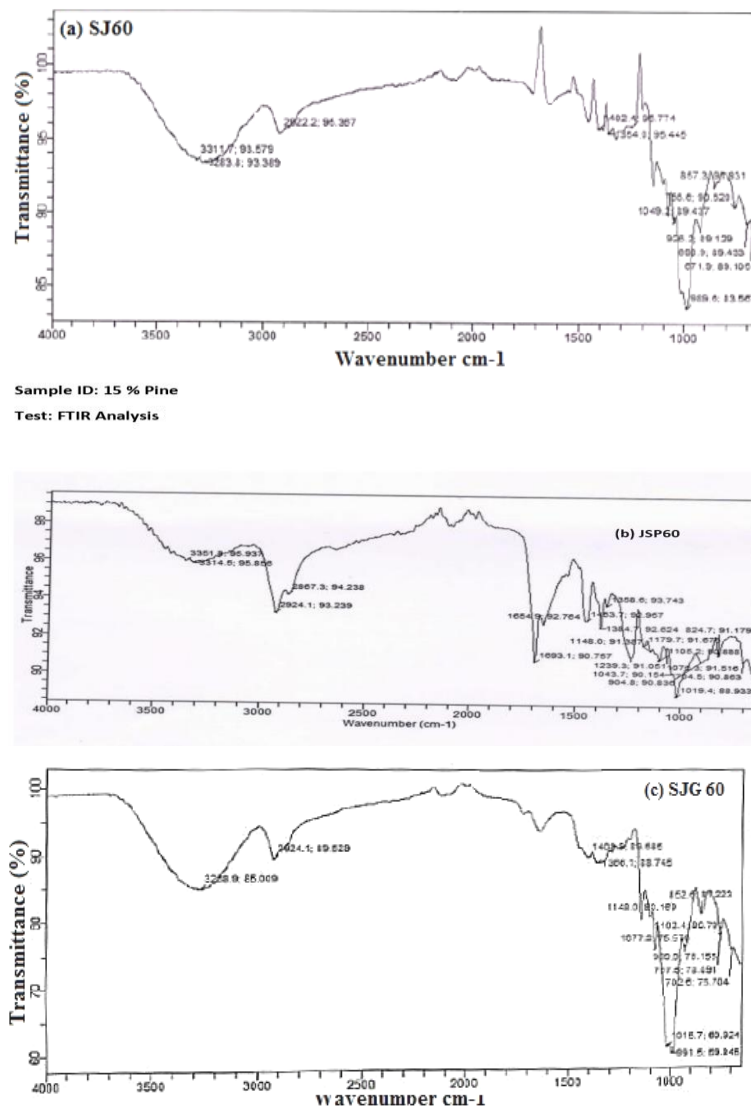


Fig. 5 FTIR of Composite Samples

than starch-jute composite without a cross linker at same fibre volume fraction. On the other hand, starch-jute composite with glutaraldehyde shows low water resistant properties as compared to composite having pine rosin as cross linker which reveals that pine rosin has better hydrophobic nature as compared to glutaraldehyde. From results, it was also found that as the fibre proportion in the composite increases, water absorbency also increases. It might be due to hydrophilic nature of jute fibre and also might be due to more voids in the composite. At high fibre content, matrix content becomes less which is not enough to wet and cover all the jute fibres. It was also observed that the absorption of water adversely affects the mechanical properties of composite materials due to differences in water uptake of fibers and matrix that leads to dimensional changes and weak adhesion between matrix and fibers.

5.5 FTIR

The FTIR spectrum of Starch-jute composite without pine rosin shows (Fig. 5) the typical absorption bands corresponding to the functional groups of starch, jute and glycerol. The band

corresponding to C-O stretching was found at 1019 cm^{-1} . The O-H groups are found at 3314 cm^{-1} and C-H stretching is located at 2867 cm^{-1} . The representative bands of the glycerol plasticizer were displayed at 2924 cm^{-1} (C-H), associated with the hydroxyl groups, as well as at 1453 cm^{-1} and at 1239 cm^{-1} (C-O stretching), associated with carbon-oxygen (C-O) absorption peaks characteristic of primary and secondary alcohols. The spectrum of starch jute composite with pine rosin shows reduction of the -OH group band at 3043 cm^{-1} from 3314 cm^{-1} . There are significant shifts in the characteristic peaks of starch jute composite with pine rosin, which indicates that the interaction among pine rosin, starch or jute is strong. Reduction in the peak indicates that the -OH groups of composite have more affinity with resins than with water. Thus, it can be stated that -OH groups are involved in positive interaction with pine rosin such as hydrogen bonding (Aldas *et al.* 2020).

The spectrum of starch jute composite with glutaraldehyde band corresponding to -OH stretching appeared at 3228 cm^{-1} . It was observed that the intensity of the hydroxyl peak decreased and shifted to 3228 cm^{-1} from 3311 cm^{-1} . The decrease in intensity and shifting of the -OH stretching peak were the more for the starch-jute composite with pine rosin followed by glutaraldehyde. The decrease in peak intensity and shifting to a lower wave number in the composite might be due to the enhancement of interaction between hydroxyl groups of jute with starch and glutaraldehyde. A similar result was found by Prasanta Baishya and Tarun K. Maji (2014) while studying the effect of cross linker on starch based wood composite (Singha and Kapoor 2014). On comparing pine rosin with glutaraldehyde, it was found that the number of -OH group is much lower in case of pine rosin modified starch than glutaraldehyde modified starch which indicates that bonds formed by pine rosin are much stronger than the bonds formed by glutaraldehyde.

5.6 SEM

To observe surface morphology, SEM was performed for Jute-starch composite without a crosslinking agent, pine rosin modified starch jute composite and glutaraldehyde modified starch jute composite. Images for surface morphology of the longitudinal surface of the fractured sample have been shown in Figs. 6(a), (c) and (e) respectively, in Fig. 6(a), it is seen that the surface of the Jute-starch composite without cross-linker is smoother than the surface of the composite with cross-linkers (pine rosin and glutaraldehyde). From Figs. 6(c) and (e), it is revealed that because of crosslinking agent, the surface of the composite becomes rougher and irregular as compared to the composite without a crosslinker. It leads to better adhesion between jute fibre and matrix along with the betterment of strength and rigidness. Similarly, SEM images of cross-sections of fractured samples of the jute- starch composite without a crosslinking agent, pine rosin modified starch jute composite and glutaraldehyde modified starch jute composite has been shown in Figs. 6(b), (d) and (f) respectively. Among these figures Fig. 6(d) shows a comparatively denser surface, it may be due to better adhesion of fibre with pine rosin modified starch-based matrix. It results in enhanced tensile and flexural properties of pine rosin modified starch jute composites as compared to glutaraldehyde modified starch jute composite.

Morphological structure studied by Karishma *et al.* (2022) exhibits that Jute-SPI composite without pine rosin have smoother surface as compared with Jute-SPI composite having 15% pine

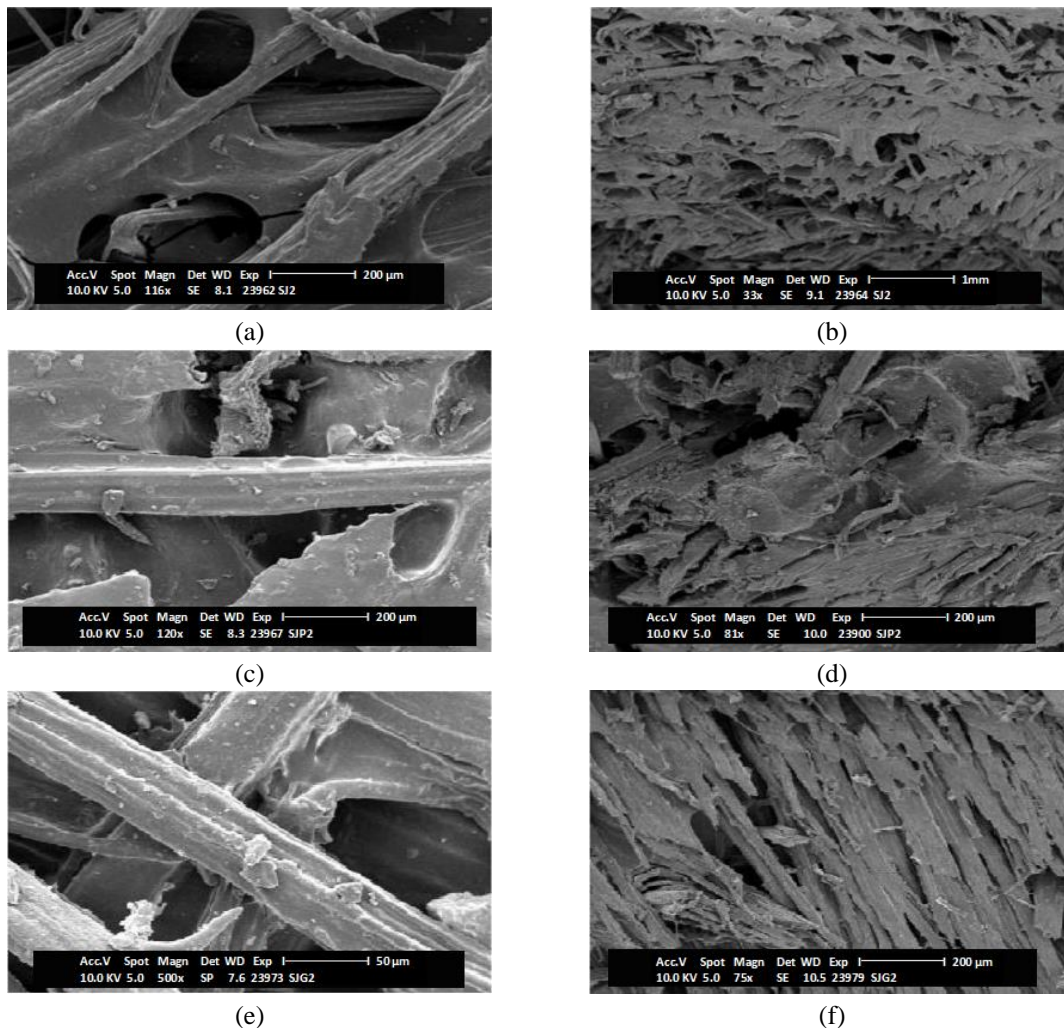


Fig. 6 SEM Photographs of (a) Longitudinal view of fractured composite sample (SJ60), (b) Cross-sectional view of fractured composite sample (SJ60), (c) Longitudinal view of fractured composite sample (SJP60), (d) Cross-sectional view of fractured composite sample (SJP60), (e) Longitudinal view of fractured composite sample (SJG60) and (f) Cross-sectional view of fractured composite sample (SJG60)

rosin. Torres *et al.* (2007) worked on the composite of Sisal Fibre and Potato Starch. Fractured surface of the composite shows the fibre pullout stuck with brittle starch. Large pullout lengths indicate poor adhesion between fibre and starch. In another research produced by Karishma *et al.* (2023) reveals that the addition of pine rosin with starch makes fibre surface more rougher in comparison to Jute-Starch composite without pine rosin which indicates the enhancement of mechanical bonding between matrix mix and jute fibre. SEM images of composite produced by using thermoplastic starch and recycled paper cellulose fibres indicate that there is no fibre pullout during fracture development which reflects the better interfacial bond between recycled fibre and starch matrix (Amnuay *et al.* 2011). In the current research work morphological views indicate that in case of plain jute/starch composite, surface of jute fibre of fractured composite became rougher

when pine rosin and glutaraldehyde were used as crosslinking agents. Therefore, both pine rosin and glutaraldehyde contribute in adhesion between fibre and matrix. But, the pine rosin contributes more than the Glutaraldehyde.

6. Conclusions

Pine rosin modified starch jute composites have shown higher tensile strength and modulus as compared with glutaraldehyde modified starch jute composite. Tensile strength and modulus of pine rosin modified starch jute composite with 60:40 matrix to jute fibre volume proportion are found to be 13.79 MPa and 782.94 MPa which are highest among the other proportion. Similar trends were found in flexural strength and modulus of pine rosin modified starch jute composite with 60:40 matrix to jute fibre volume proportion which are 29.18 MPa and 1107.76 MPa respectively; that are also maximum but in case of impact strength, glutaraldehyde modified starch jute composite with 80:20 matrix to jute fibre volume proportion was found to be highest that is 59.07 KJ/m². With improved fibre volume fraction, tensile and flexural properties have improved. However, impact strength got reduced. Starch-jute composite with glutaraldehyde shows lower water resistant properties as compared to composite having pine rosin as cross linker. FTIR graph indicates that the number of -OH group is much lower in case of pine rosin modified starch than glutaraldehyde modified starch which indicates that bonds formed by pine rosin are much stronger than the bonds formed by glutaraldehyde. SEM image shows that surface morphology has been changed due to the addition of pine rosin and glutaraldehyde when compared with composite without a crosslinking agent.

Acknowledgments

Authors specially acknowledge the supports of Department of Fibre Science and Textile Processing of Institute of Chemical Technology Mumbai and would also like to acknowledge the Department of Production Engineering, Veermata Jijabai Technological Institute Mumbai for providing their testing facilities.

References

- Alam, M., Maniruzzaman, M. and Morshed, M. (2014), "Application and advances in microprocessing of natural fiber (jute)-based composites", *Comprehens. Mater. Pr.*, **2014**, 243-260. <https://doi.org/10.1016/B978-0-08-096532-1.00714-7>.
- Aldas, M., Pavon, C., López-Martínez, J. and Arrieta, M. (2020), "Pine resin derivatives as sustainable additives to improve the mechanical and thermal properties of injected moulded thermoplastic starch", *Appl. Sci.*, **10**(7), 2561. <https://doi.org/10.3390/app10072561>.
- Aliyaa, B., Sunooj, K. and Lackner, M. (2021), "Biopolymer composites: A review", *Int. J. Biobased Plast.*, **3**(1), 40-84. <https://doi.org/10.1080/24759651.2021.1881214>.
- Aly-Hassan, M. (2015), *Multifunctionality of Polymer Composites*, Elsevier, Amsterdam, Netherlands.
- Amnuav, W., Katavut, P., Supranee, K. and Pichan, S. (2011), "Green composite of thermoplastic corn starch and recycled paper cellulose fibres", *Songklanakarin J. Sci. Technol.*, **33**, 461-467.
- ASTM Standard D 256-10 (2010), Standard Test Method for Determining the Izod Pendulum Impact

- Resistance of Plastics, ASTM International, West Conshohocken, PA, USA.
- ASTM Standard D 570-98 (2010), Standard Test Method for Water Absorption of Plastic, ASTM International, West Conshohocken, PA, USA.
- ASTM Standard D 638-03 (2003), Standard Test Method for Tensile Properties of Plastic, ASTM International, West Conshohocken, PA, USA.
- ASTM Standard D 7390-07 (2007), Standard Test Method for Flexural Properties of Unreinforced and Reinforced Plastic and Electrical Insulating Materials, ASTM International, West Conshohocken, PA, USA.
- Baishya, P. and Maji, T. (2014), "Studies on effects of different cross-linkers on the properties of starch-based wood composites", *ACS Sustainab. Chem. Eng.*, **2**(7), 1760-1768. <https://doi.org/10.1021/sc5002325>.
- Biswas, S., Ahsan, Q., Cenna, A., Hasan, M. and Hassan, A. (2013), "Physical and mechanical properties of jute, bamboo and coir natural fiber", *Fib. Polym.*, **14**(10), 1762-1767. <https://doi.org/10.1007/s12221-013-1762-3>.
- Chand, N. and Fahim, M. (2008), *Tribology of Natural Fiber Polymer Composites*, Woodhead Publishing, Cambridge, UK.
- Dicker, M., Duckworth, P., Baker, A., Francois, G., Hazzard, M. and Weaver, P. (2014), "Green composites: A review of material attributes and complementary applications", *Compos. Part A: Appl. Sci. Manuf.*, **56**, 280-289. <https://doi.org/10.1016/j.compositesa.2013.10.014>.
- Gupta, M., Srivastava, R. and Bisaria, H. (2015), "Potential of jute fibre reinforced polymer composites: A review", *Int. J. Fiber Textile Res.*, **5**, 30-38.
- Ibrahim, M., Sapuan, S., Zainudin, E., Zuhri, M. and Edhirej, A. (2019), "Corn (maize) - its fibers, polymers, composites, and applications", *Biodegrad. Compos. Mater. Manuf. Eng.*, **10**, 14-35. <https://doi.org/10.1515/9783110603699-002>.
- Iman, M. and Maji, T. (2012), "Effect of crosslinker and nanoclay on starch and jute fabric based green nanocomposites", *Carbohydr. Polym.*, **89**(1), 290-297. <https://doi.org/10.1016/j.carbpol.2012.03.012>.
- Kandpal, B., Chaurasia, R. and Khurana V. (2015), "Recent advances in green composites - A review", *Int. J. Technol. Res. Eng. (IJTRE)*, **2**(7), 742-747.
- Kaur, L., Singh, J. and Liu, Q. (2007), "Starch - A potential biomaterial for biomedical applications", *Nanomaterials and Nanosystems for Biomedical Applications*, Springer Science+Business Media, Dordrecht, Netherlands.
- Mahendra, V. (2015), "Fabrication of biocompatible hydrogels from pine resin", *Winter Solstice*, **5**, 1-5.
- Mohammed, L., Ansari, M., Pua, G., Jawaid, M. and Islam, M. (2015), "A review on natural fiber reinforced polymer composite and its applications", *Int. J. Polym. Sci.*, **2015**(1), 1-15. <https://doi.org/10.1155/2015/243947>.
- Popescu, E., Stoica, A. and Elena, B. (2010), "Morphological and thermal properties of maize starch", *Annal. Food Sci. Technol.*, **11**(2), 35-39.
- Pradeep, Y. (2018), "Synthesis, fabrication and characterization of jute fibre reinforced laminar composites", *Int. J. Mech. Eng. Technol. (IJMET)*, **9**, 722-731.
- Rahman, M., Huque, M. and Islam, M. (2008), "Improvement of physico-mechanical properties of jute fiber reinforced polypropylene composites by post-treatment. Composites part A", *Appl. Sci. Manuf.*, **39**, 1739-1747. <https://doi.org/10.1016/j.compositesa.2008.08.002>.
- Ribeiro, R., Marques, A. and Alves, J. (2018), "Sustainable composites based on pine resin and flax fibre", *Advances in Natural Fibre Composites: Raw Materials, Processing and Analysis*, Springer International Publishing, Cham, Switzerland.
- Sakhare, K. and Borkar, S. (2022), "Characterization of jute fibre reinforced pine rosin modified soy protein isolate green composites", *Adv. Mater. Res.*, **11**(3), 191-209. <https://doi.org/10.12989/amr.2022.11.3.191>.
- Sakhare, K., Borkar, S. and Kale, R. (2023), "Fabrication and characterization of Bio composite based on jute fibre and pine rosin modified potato starch", *J. Chem. Health Risk*, **13**, 258-271.
- Sharma, L. and Singh, C. (2016), "Composite film developed from the blends of sesame protein isolate and gum rosin and their properties", *Polym. Compos.*, **39**(5), 1480-1487. <https://doi.org/10.1002/pc.24088>.
- Shinoj, S., Visvanathan, R., Panigrahi, S. and Kochubabu, M. (2011), "Oil palm fiber (OPF) and its composites: A review", *Indust. Crops Prod.*, **33**(1), 7-22. <https://doi.org/10.1016/j.indcrop.2010.09.009>.

- Singha, A. and Kapoor, H. (2014), "Effects of plasticizer/cross-linker on the mechanical and thermal properties of starch/PVA blends", *Iran. Polym. J.*, **23**(8), 655-662. <https://doi.org/10.1007/s13726-014-0260-9>.
- Sousa, D., Biscaia, S., Viana, T., Gaspar, M., Mahendra, V., Mohan, S. and Mitchell, G. (2019), "Rosin based composites for additive manufacturing" *Appl. Mech. Mater.*, **890**, 70-76. <https://doi.org/10.4028/www.scientific.net/AMM.890.70>.
- Sun, S., Liu, P., Ji, N., Hou, H. and Dong, H. (2018), "Effects of various cross-linking agents on the physicochemical properties of starch/PHA composite films produced by extrusion blowing", *Food Hydrocoll.*, **77**, 964-975. <https://doi.org/10.1016/j.foodhyd.2017.11.046>.
- Torres, F., Arroyo, O. and Gomez, C. (2007), "Processing and mechanical properties of natural fiber reinforced thermoplastic starch biocomposites", *J. Thermoplast. Compos. Mater.*, **20**(2), 207-223. <https://doi.org/10.1177/0892705707073945>.
- Vázquez, A. and Alvarez, V. (2009), "Starch-cellulose fiber composites", *Biodegradable Polymer Blends and Composites from Renewable Resources*, John Wiley & Sons Inc., Hoboken, NJ, USA.
- Vilaseca, F., Mendez, J., Pèlach, A., Llop, M., Cañigüeral, N., Gironès, J. and Mutjé, E. (2007), "Composite materials derived from biodegradable starch polymer and jute strands", *Pr. Biochem.*, **42**(3), 329-334. <https://doi.org/10.1016/j.procbio.2006.09.004>.