Convention Special Issue – 2020

J. Indian Chem. Soc.,
Vol. 97, No. 12a, December 2020, pp. 2679-2688

Development of fire retardant eco-composite board from jute and bio based *Euphorbia latex*-polyvinyl alcohol (PVA) copolymer for structural purpose

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Manuscript received online 10 November 2020, revised and accepted 23 December 2020

Natural fast growing lingo-cellulosic fabrics are widely used for the preparation of composite board, as these are eco-friendly and biodegradable. The present article is based on the development of eco-friendly fire retardant composite board from jute as reinforcing agent and polyvinyl alcohol (PVA) as matrix and its partial substitution with natural *Euphorbia latex* coagulum in different ratio as adhesives. From the basic mechanical and fire retardant properties study (as per ASTM standard) of the composite board using *E. latex* with PVA adhesive shows appreciable increase in strength as well as fire retardant characteristics compare to the material made of jute and synthetic pure PVA adhesive. This technique of using bio based adhesive is cost-effective, eco-friendly and could be an alternative way to reduce the use of petroleum based non biodegradable adhesives.

Keywords: Jute fabric, adhesive, composite, fire-retardant, coagulum, PVA.

Introduction

Utilization of natural renewable resources[1–3] as reinforcing agent has been started from the beginning of our civilization. Increase in population associated with rapid demand of wood and wooden material for house hold and industrial purpose forced the people to develop wood substitute materials like metals and plastics. Due to high cost and heavy weight of metal, plastic and fabric reinforced composite materials became popular alternative resources. Also the fabric reinforced plastics have higher specific modular strength property which challenges the metal in different purposes. For composite preparation inorganic fabric (glass, asbestos) has major draw-back like huge cost, non-biodegradable, and have health hazard nature during preparation and handling which promoted natural resources[4–11] as reinforcing material. Natural fibers[12–23] are abundantly available, renewable, and cheaper compared to synthetic fibers and have a unique property to make strong bond with aqua base adhesive, as both fiber and adhesive have polar functionalities.

Many research works have been done on natural fibers like jute, sun hemp, banana, sisal coir etc. and their significant application in industry to prepare composite board because these boards are biodegradable in nature. Jute is one of the important bast fibres abundantly available in all over India, mainly in West Bengal. Scientists are attracted to jute instead of other fibers due to its easy availability, renewable (matured within three months), low price, low density leading and high specific strength. The present study emphasizes on utilization of *Euphorbia latex* coagulum with PVA adhesive in different proportion for the development of jute fabric reinforced composite board. Like other natural fabrics the scope of using jute fabric (specific modulus 1.29 and 40 GPa)[24] in place of traditional glass fiber (2.5 and 30 GPa) because of its much lower cost, low density, renewable nature, low abrasion, the ease with which it can be processed and handled with low energy (only 2% with respect to glass fiber)[1,2] requirement for production and processing of jute lift it up as an alternative fiber for use as reinforcing agent in preparation of composites.

Jute has some drawbacks like poor adhesion[25] with organic adhesives due to presence of some polar functionalities (-OH, -COOH, -OCH_3) which is not suitable for non-polar adhesives, fire hazard and damaged in humid condition but appreciable adhesion with aqua-base thermosetting adhesive like phenol formaldehyde, melamine formaldehyde, and urea formaldehyde resins. Modification of jute by suitable coupling agent or chemical treatment makes jute as a more useful reinforcing material for both aqua-base and organic adhesive as matrix. An improvement of strength of jute in dry and wet with the application of some interfacial agent have been reported elsewhere[25]. Pre-treatment on jute fabric with low molecular weight phenol formaldehyde and CSNL modified phenol formaldehyde shows better in reducing the moisture regain but strength of composite is found to be increased (Mitra *et al.*). *E. latex* is collected from Euphorbiaceae family such as *Euphorbia royleana*, *Euphorbia nerifolia* and *Euphorbia cadicifoliaalong* with *Hevea brasiliensis*[26]. The main constituents of Euphorbia latex[27,28] is adhesiveous mass (65–80%) containing tetracyclic triterpinoid, protein (10–20%), *cis*-1,4-polyisoprene (5–10%) and small amount of ash[29]. Partial replacement of PVA with bio-based *E. latex* is very much compatible with jute and an effective coupler for PVA and jute fabric improve the strength, fire retardant properties as well as water resistance, which may open the manufacture of jute based composite by using bio-based adhesive.

Materials

Woven jute fabric:

For the work woven jute fabric of weight 400–600 GSM and 1.5 mm thickness was used for this project as shown in Fig. 1. Jute fabric is available in the market (collected from Gloster Ltd., 21, Strand Road, Kolkata) in roll form and this roll was cut to size as per the dimension required for the study. The strength properties are given in Table 1.

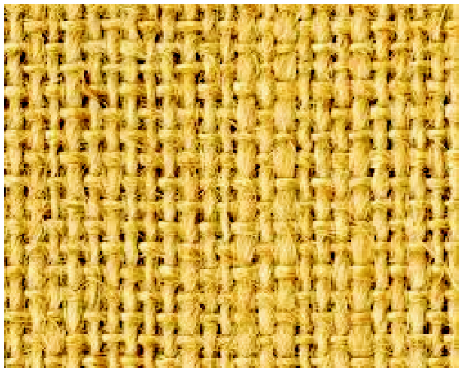

Fig. 1. Woven jute fabric.

Euphorbia latex:

Euphorbia latex was collected from foothills of Himalayas around the region of Sahastradhara, Dehradun, India. The milky latex was coagulated by adding 5% aqueous solution of tannic acid. The coagulated latex was washed several times with pure water to remove tannic acid till the pH of the material become 7 (checked with E. Merck pH paper). Ultimately the material is dried in a hot air oven at a temperature 60°C[16].

PVA adhesive and solvent:

PVA (hot) small beads of industrial grade and amyl acetate as solvent were collected from Agarwal Chemicals, 40, Strand Road, Kolkata, West Bengal.

Table 1. Mechanical properties of jute fabric

Type of fabric	Physical properties		Mechanical properties						
	Density (kg/m^3)	Diameter (μm)	Tensile strength (MPa)		Young's modulus (GPa)		Specific strength (MPa/g^{-3})	Specific modulus (GPa/g^{-3})	Failure strain
			Ave.	M.I.[a]	Ave.	M.I.[a]	Ave.	Ave.	(%)
Jute	1290[b]	29–52	380	152	25.2	7.1	293.2	18.1	1.38

M.I. (Minimum Individual).

[a]The observations number was about 50.

[b]Density of jute is calculated from the specific gravity referred to Ref. [7].

[Tensile tests were conducted using a small-capacity testing machine (ASG-H/Ez Test-00: Shimadzu) at 1 mm/min of crosshead speed].

Experimental

Modification/pre-treatment of fabric:

Pre-treatment on jute fabric make the surface clean and open, so that adhesive can react more easily introduce proper bonding between fabric and adhesive making composite more stable, low moisture regain and improved strength. Several pre-treatment have been attempted on jute like alkali treatment for scouring[30], pulping[31] and bleaching etc. as given in Table 2.

Table 2. Different pre-treatment on jute fabric

Nature of jute	Chemical for pre-treatment	pH	Bundle strength (g/tex)
Woven jute fabric	Control	7	25.60
Woven jute fabric	2% NaOH (2 h)	7	22.49
Woven jute fabric	1% NaOH + 1% Na_2S	7	24.46
Woven jute fabric	Bleached (H_2O_2 in NH_3 buffer)	7	23.03

Mixing of E. latex, PVA adhesive for glue preparation:

PVA, latex coagulum, fire retardant chemicals and marble dust (filler) were mixed thoroughly in different ratio with 1000 ml of amyl acetate and the glue viscosities were are also checked as given in Table 3.

Table 3. Preparation of glue from PVA, *E. latex* and respective viscosity values

Sl. No.	Adhesives	Ratio	Total (part)	Viscosity (cp)
1.	PVA and *E. latex*	100:00	100	620
2.		90:10	100	710
3.		80:20	100	780
4.		70:30	100	795
5.		60:40	100	820

Viscosity of the glue was determined using Brookfield viscometer of spindle-1@ speed 10.

These five sets of adhesives mixed properly just before impregnation of the glue mixture in jute fabric.

Impregnation of glue into jute fabric:

Three piece of woven jute fabric (approximate weight 0.15 kg) was taken as reinforcing agent of specific dimension with an allowance of 1 inch both in the length and width for shrinkage and soaked in glue by hand lay-up technique using a brush and dried in open air and assembled for hot pressing.

Manufacture of composite board:

Three layer glue impregnated jute fabric were assembled in between two stainless steel plates coated with a thin layer of mould releaser (a mixture of wax). Then assembled glued fabrics are hot pressed at specific pressure of 14 kg/cm^2 at the curing temperature at 100°C[22] for 8 min as required time of the glue to form gel at 100°C is 5 min.

Results and discussion

Properties E. latex (FTIR analysis):

The rubber from *E. latex* shows the FTIR bands at 1664 cm^{-1}, 835 cm^{-1} and 3342 cm^{-1} which are due to C=C stretching and C-H bending and N-H stretching respectively shown in Fig. 2.

Properties E. latex and PVA adhesive mixture (FTIR analysis):

The iodine values of the latex coagulum as well as mixed adhesive (with catalyst, accelerator) was determined (before gel time in mixed adhesive) as per ASTM D-1959. The iodine values for only coagulum, only PVA adhesive and mixed glue of PVA and coagulum at a ratio 70:30 are 155, 134 and 122 respectively. The high value for coagulum indicates the presence of higher degree of unsaturation where as in case of glue the comparative less value strongly indicate about some cross linking of unsaturated bonds by MEKP (Methylethylketone peroxide) catalyst. The FTIR spectra also indicate that latex contain unsaturated carbon, amide group (protein part in latex). The ester C=O and unsaturated C=C stretching vibration as shown in Fig. 3 reduced may be due to resonance with coagulum unsaturated chain indicating their interaction.

SEM analysis:

Morphological study of the composite board was done by field emission scanning electron microscope(JEOL). Magnification at X35 reveals that porosity and pore size distribution in SEM images were used to characterize all the samples and allowed to identify their microstructure. Large air voids, with a diameter of about 40–60 μm, were found in the PVA based composite sample, as shown in Fig. 4, while, small cavities were observed in the samples containing PVA with latex coagulum as shown in Fig. 5.

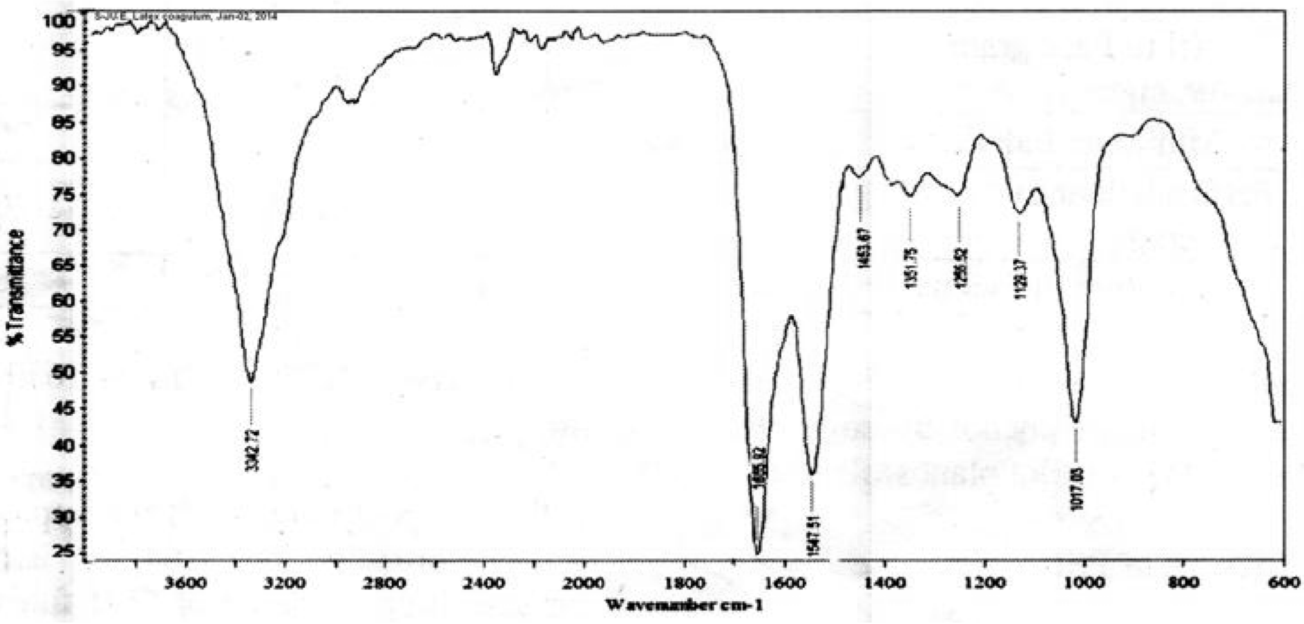

Fig. 2. IR spectrum of *E. latex* coagulum.

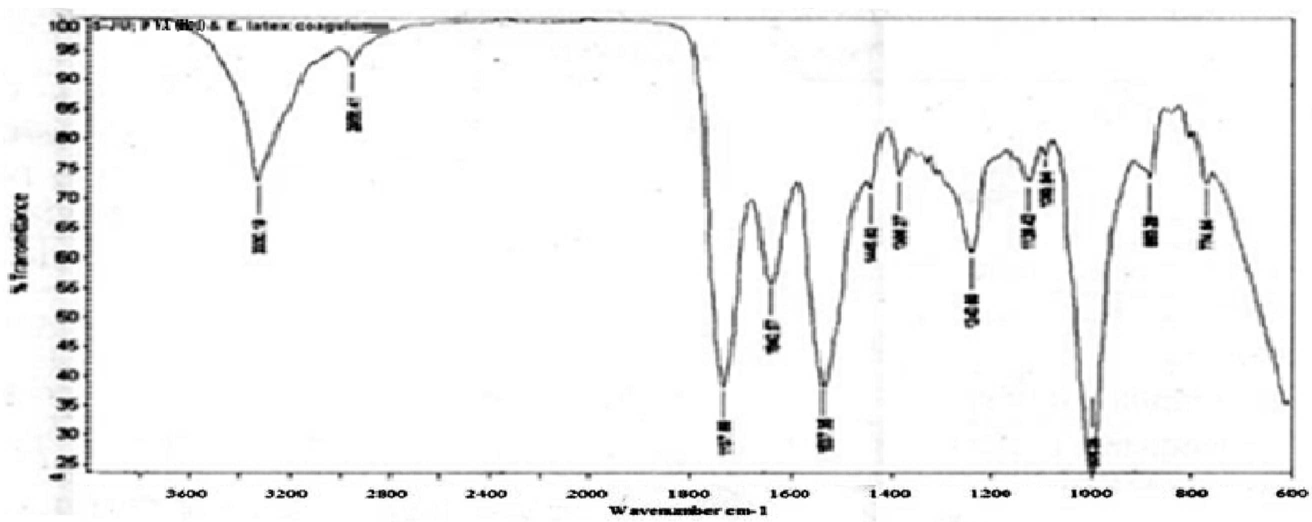

Fig. 3. IR spectrum of PVA and *E. latex* coagulum (70:30) glue.

Furthermore, it was interesting to observe that 30% inclusion of *E. latex* into PVA has reduced air voids size smaller as well as better surface finish may be due to compatibility and cross linking by catalyst MEKP between PVA and *E. latex*.

Water absorption test:

The water absorption test was carried out as per of ASTM D-570. The test results indicate that the composite from treated jute fabric shows comparable water absorption property with the control jute but slight improvement in case of 2% NaOH scouring for two hour. Also the water absorption property increases with increase in the percentage of *E. latex* may be due to better bonding between partial removed polar hydrophilic functionalities as well as opening of fabric and hydrophobic adhesive mixture as shown in Table 4 and Fig. 6.

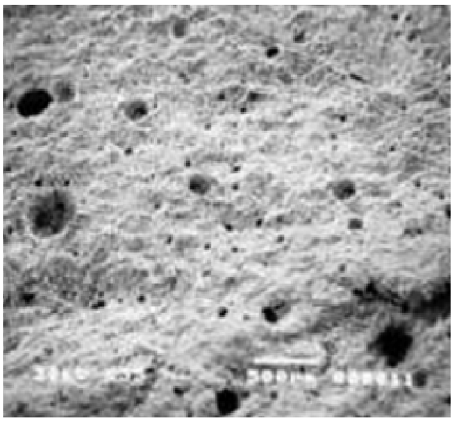

Fig. 4. SEM micrographs of composite made from jute (2% NaOH scouring for two hour) and polyvinyl alcohol with 3% additive composition.

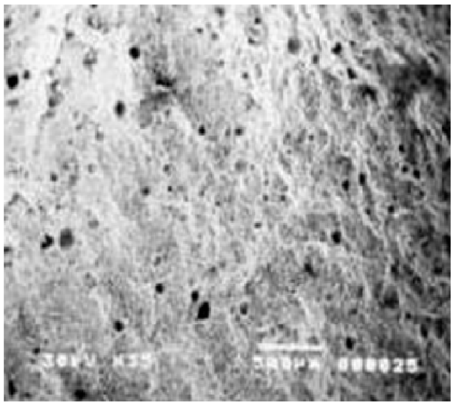

Fig. 5. SEM micrographs of composite made from jute (2% NaOH scouring for two hour) and polyvinyl alcohol:*E. latex* mixed glue (70:30) with 3% additive composition.

Flexural test:

For flexural test the specimens were cut from molded sheet. The dimension of the test specimens was 127 mm× 12.7 mm×3.0 mm. Flexural strength of coagulum modified PVA jute fabric composites specimens were tested on universal tensile machine (Instron, 4302 model, UK) at the cross-head speed of 5 mm/min and at a span length of 96 mm. Three-point bend test were performed to measure flexural strength. Temperature was maintained at 23±2°C as per the guidelines of ASTM D-790. Flexural strength property increase though strength decreases with the increase in percentage of latex coagulum as shown in Table 5 and Fig. 7,

Table 4. Water absorption test of composite of size 2 inches diameter

Nature of jute	PVA adhesive:*E. latex*	After 2 h (%)	After 24 h (%)
Control jute fabric	100:00	7.80	9.80
	90:10	7.65	9.75
	80:20	7.50	9.36
	70:30	7.30	9.15
	60:40	7.15	9.12
2% NaOH scouring for 2 h	100:00	6.65	7.78
	90:10	6.42	7.67
	80:20	6.40	7.63
	70:30	6.35	7.52
	60:40	6.27	7.14
1% NaOH + 1% Na_2S scouring for 2 h	100:00	6.64	7.77
	90:10	6.40	7.72
	80:20	6.40	7.70
	70:30	6.38	7.71
	60:40	6.37	7.68
Bleaching by H_2O_2 in NH_3 buffer solution	100:00	6.92	7.87
	90:10	6.88	7.86
	80:20	6.86	7.82
	70:30	6.77	7.82
	60:40	6.78	7.80

The specimens are dried in an oven then placed in desiccators to cool and weight immediately.

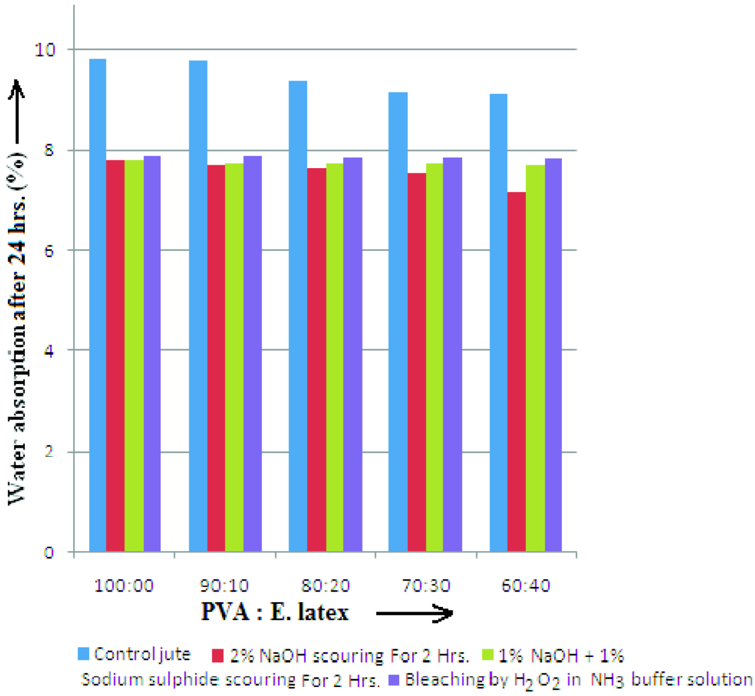

Fig. 6. Water absorption of composite board made from jute and mixed glue at different ratio.

Table 5. Tensile and flexural strength of composite board

Nature of jute	PVA:*E. latex* adhesive	Av. tensile strength (MPa)	Elongation at break (mm)	Flexural strength (MPa)
Control jute fabric	100:00	50	1.85	22.2
	90:10	52	1.94	24.5
	80:20	55	2.00	24.9
	70:30	54	1.98	25.1
	60:40	52	2.02	25.6
2% NaOH scouring for 2 h	100:00	60	1.92	23.6
	90:10	63	1.92	24.9
	80:20	67	1.96	28.2
	70:30	63	1.99	32.4
	60:40	59	2.00	32.6
1% NaOH + 1% Na_2S scouring for 2 h	100:00	55	2.03	23.9
	90:10	55	2.24	24.5
	80:20	57	2.08	27.2
	70:30	57	2.44	30.6
	60:40	53	2.48	32.0
Bleaching by H_2O_2 in NH_3 buffer solution	100:00	53	4.03	24.2
	90:10	54	4.42	26.5
	80:20	55	4.48	29.3
	70:30	53	4.69	33.2
	60:40	53	4.92	33.8

Av. tensile strength is obtained from three samples tests were done by universal tensile machine (Instron, 4302 model, UK).

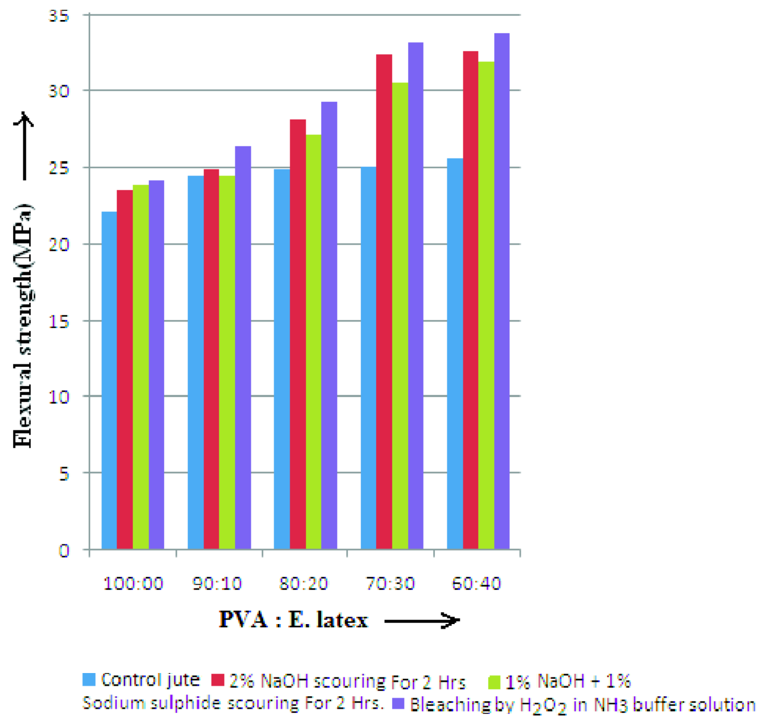

Fig. 7. Average flexural strength of composite board made from jute and mixed glue at different ratio.

indicate utilization of such type of mixed glue incorporate with latex coagulum increase the flexural strength of composite.

Tensile strength of compressed impregnated jute fabric:

The tensile test was performed according to ASTM D-638. The samples were 150 mm long, 25 mm wide with thickness of 3 mm. three specimens were tested on universal tensile machine (Instron, 4302 model, UK) at cross head speed of 0.5 mm /min. Load displacement curves were generated for each sample. For measuring elongation, two marks along the central load axis were made at a distance of 15 mm, on either side from the centre of the sample. The test results showed that better bonding with in case of 2% NaOH treatment. This may due to removal of polar functionalities and opening of fabric as a result there is better interaction which leads to better bonding between jute and glue as the glue is hydrophobic in nature. The results also revealed that interaction is effective in between PVA and *E. latex* at a ratio 80:20 may be due to optimize compatibility among adhesives and effective bonding with jute fabric. The results are given in Table 5 and Fig. 8.

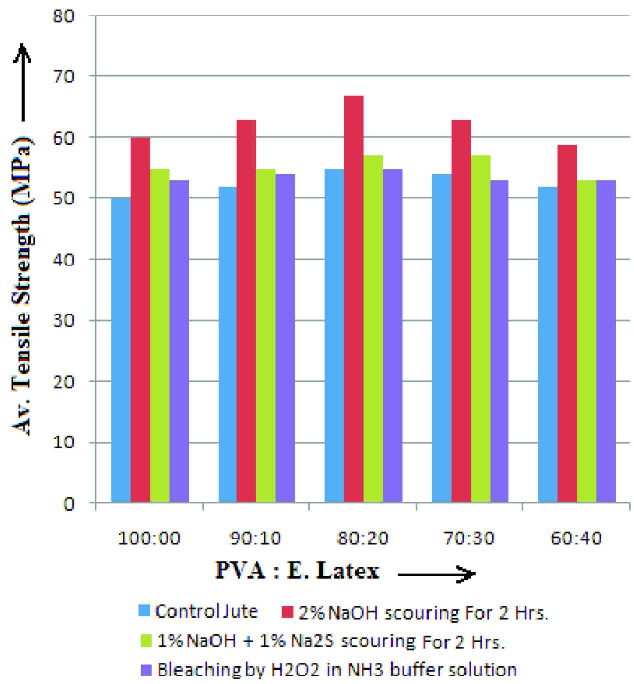

Fig. 8. Average tensile strength of composite board made from jute and mixed glue at different ratio.

Fire retardant test:

Limiting oxygen index (LOI):

The fire retardant test were done as per the limiting oxygen index (LOI) method. LOI was determined for each specimen of the size 6 mm×25 mm as per ASTM D-2863. The values of LOI in different composites are given in Table 6. It is observed that with the increase percentage of the latex coagulum replacing PVA in the composites increase the LOI value. Further, increase LOI values are found in every case of treated jute fabric may be due to removal of lignin[32] from jute fabric showed that only jute-PVA composite is fire hazard where as LOI property is improved by incorporation of *E. latex*, may be due to generation of nitrogen and different oxides of nitrogen during burning, liberated from protein[29] in coagulum (a eco-friendly material).

Smoke density:

Smoke density[33] was tested at NIRJAFT (under Indian council of Agricultural research, New Delhi) by smoke density tester (Supplied by Asian Test Equipments, F-49, Massori Gulawthi Road U.P.S.I.D.C. Industrial area, Ghaziabad, Uttar Pradesh, India) as per ASTM D-2843. During the burning process of reinforced polymeric materials, smoke generation is a cause of concern, as it will affect not only visibility during rescue operation but also pollute surrounding environment. Incorporation of coagulum in PVA shows varying degrees of smoke generation. A remarkable decrease in smoke densities strongly indicate the formation of N_2 and NO_X during combustion of proteinous mass of coagulum, as reflected in Fig. 9. Here loss in % of light transmission w. r. to smoke density have been studied in standardized conditions.

Table 6. Limiting oxidation index (LOI) values

Nature of jute	PVA:*E. latex* adhesive	LOI
Control jute fabric	100:00	18.5
	90:10	21.1
	80:20	22.9
	70:30	23.2
	60:40	23.6
2% NaOH scouring for 2 h	100:00	19.3
	90:10	21.6
	80:20	22.8
	70:30	23.8
	60:40	24.6
1% NaOH + 1% Na_2S scouring for 2 h	100:00	19.5
	90:10	22.6
	80:20	23.9
	70:30	24.5
	60:40	24.5
Bleaching by H_2O_2 in NH_3 buffer solution	100:00	19.9
	90:10	21.9
	80:20	23.2
	70:30	25.8
	60:40	26.8

LOI was done by the Instrument and Research Equipment Ltd.,Unit - 7, Grays Farm Production, Vill. Grays farm Rd. Orpington Kent (PRS)3, DD. UK, Telephone No. +44(020)83006939, Model type; LOI Test apparatus, Supply 230 V : Current; 5 amp, Serial No. -200000-020.

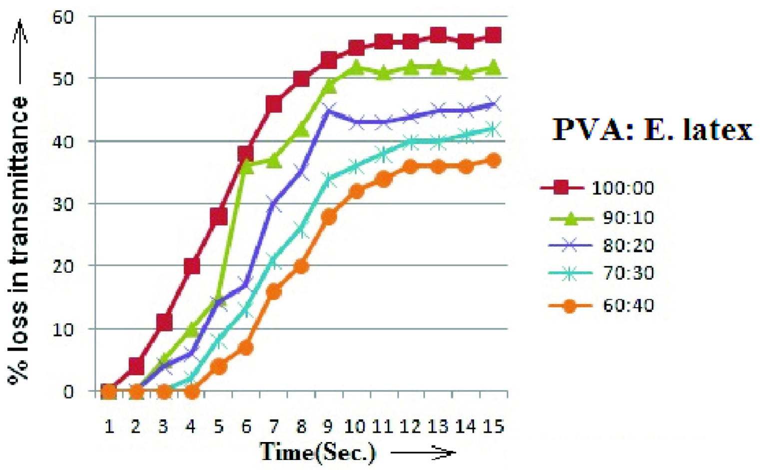

Fig. 9. Smoke density results of composite board made from jute and mixed glue at different proportion.

Finishing of composite board:

To make the product appealing in to the market as demand for different purpose the product requires proper finishing on the surface. To break the monotony of the same colour and lack of uniformity of its own colour on the surface, overlay of any colour is possible as given in Fig. 10 and Fig. 11.

Fig. 10. Composite board from jute 2% NaOH scouring for 2 hours and mixed glue of ratio 70:30.

Fig. 11. Composite board from jute 2% NaOH scouring for 2 hours and mixed glue of ratio 70:30.

Summery and Conclusion

Jute PVA adhesive composite is known to all; here an attempt has taken to replace PVA adhesive with compatible natural bio-base latex from euphorbia plant which is abundantly available in all over India.

(i) Comparable water absorption property with the control jute and slight improvement in case of 2% NaOH scouring for two hour indicate better bonding due to opening of jute fabric.

(ii) The flexural strength indicates that the latex has great effect on it, and it becomes maximum 30% replacement of PVA adhesive.

(iii) The tensile strength results of the composites indicate that partly (20%) incorporation of natural bio based adhesive is highly effective in bonding.

(iv) From the result it can be concluded that due to presence of proteinous (containing nitrogen) material the latex is itself a fire retardant.

(v) The study finally concluded that wood substituted fire retardant composites from jute and partly substituted PVA adhesive can be used as false ceiling, partition wall, and panel and as an alternative solution with beneficiary for hardboard industries.

Acknowledgement

Financial assistance for this project from Indian Council for Agricultural Research (ICAR), New Delhi, India is gratefully acknowledged.

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