Continuous jute fibre reinforced laminated paper composite and reinforcement-fibre free paper laminate

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Abstract. Plastic bags create a serious environmental problem. The proposed jute fibre reinforced laminated paper composite and reinforcement-fibre free paper laminate may help to combat the war against this pollutant to certain extent. The paper laminate, without reinforcement fibre, exhibited a few fold superiority in tensile properties than single paper strip. The studies further show that an appreciable improvement in tensile properties can be achieved by introducing continuous jute fibre in paper laminates.

Keywords. Jute fibre; laminated paper composite; plastic bag pollution.

1. Introduction

Since the development of plastic in the earlier part of last century, it is being used widely in varieties of ways. Today plastic is extensively used to carry or wrap around many of the items that we buy or use and as an industrial packaging material. Plastics are used because they are easy to manufacture, cheap and they do not rot. Unfortunately, the same qualities make plastic a huge pollution hazard. The problem comes from indiscriminate use and subsequent disposal, particularly the throwaway plastic material used in wrapping and packaging (Williams 2004). India's plastic bag consumption is one of the highest in the world. Yet, little has been done to manage the waste and discarded bags. Plastic bags are difficult and costly to recycle and most of them end up on landfill sites or in seas and oceans. They take around 300 years to photo degrade and breakdown into tiny toxic particles that contaminate soils and water bodies and enter even the food chains. It may also be noted that a very small fraction of used plastic bags are recycled (World-jute.com). The distribution and abundance of large marine debris were investigated throughout the globe (Galgani *et al* 2000; Bugoni *et al* 2001; Derraik 2002; Ritter 2004). Plastic bags are rated amongst the top 12 items of debris, most often found along coastlines around the world. They are responsible for the choking of sewerage line, polluting water bodies and land, and indirectly encourage global warming. A large number of marine species is known to be harmed and/or killed by plastic debris, which could jeopardize their survival. Many cows

roaming on Indian streets die after chewing plastic bags containing scraps of food. Burning discarded plastic bags is not an uncommon sight in the streets and garbage yard of developing nations resulting in emission of carcinogenic pollutants. Environmentalists say that the hazards posed by plastic bags, wrapping and packaging materials can no longer be ignored.

Bleached and unbleached kraft papers and old newspapers (ONP) seem to be prosperous candidates for manufacturing cheap carry bags and wrapping materials. In fact, the ban on the use of plastic bags in different parts of the globe has encouraged the use of paper made bags. Recently, kraft paper and ONP are getting popularity to make grocery bags, kraft paper for packaging carryout food (Reid 2002; Anon 2003), and bleached and unbleached kraft papers as industrial and domestic wrapping and packaging materials (James and Chanson 2000; Miller 2002). National governments are implementing guidelines for the use of plastic bags and plastic packaging materials (Chen 2006). It may be recalled that, till mid 80s, ONP and kraft paper were most common means of packaging grocery and food in India. However, paper bags were practically phased out due to introduction of cheap and convenient plastic bags. ONP comprises a large volume of our solid waste, and therefore, its use as grocery sacks and carry-bags is expected to have two-fold advantages; firstly, it will reduce city garbage owing to effective recycling of ONP, and secondly, it will help in pollution control induced by throw-away plastic bags. It may also be noted that paper is accepted in most of the recycling programmes while recycling rate for plastic bags is very low. It is also important to mention that ONP are being recycled to generate cellulose, fibre and to make composite (Young 1990; Miller 2002; Grigoriou 2003; Samakovlis 2004).

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Environmentalists all over the world want to see the return of biodegradable packaging materials such as jute, cloth and paper. Biodegradable plastic is a potential material for packaging industries. However, it is not expected to become popular in near future, specially in third world countries. Unbleached kraft paper and ONP are viable solution to this problem. However, their low strength and poor tear resistance are major hurdles in their use as widely acceptable wrapping and packaging material.

The present study incorporates the development of continuous jute fibre (a natural, biodegradable and abundant fibre) reinforced laminated paper composite and reinforced fibre-free laminate and their characterization. The corn flour made glue was used to fabricate composite and laminate. It may also be noted that fine and white flour (*maida*) made glue is widely used in this country as cheap adhesive in paper industries. The traditional method of glue preparation was also followed to encourage its acceptability. Characterization of the fabricated laminates and composites was done by tensile test.

2. Experimental

2.1 Materials

In the present investigation, two different types of kraft papers and an ONP (old news paper) were fibre-reinforced using jute fibre of average diameter, 0.045 mm and tensile strength, 470 MPa. Thickness, tensile strength and weight of single paper strips are listed in table 1.

The paper sheets were cut into 120 mm long and 25 mm wide strips maintaining their length parallel to the machine direction of sheets. The cut pieces of paper were oven dried at 60°C for 15 min to partially remove moisture. The oven dried paper strips were then weighed and subsequently used to prepare laminates and composites.

The procured jute was carefully brushed with the help of a brass brush to separate it into fine fibres. The separated fibres were subsequently cut into 120 mm long pieces and divided into several batches of 0.02–0.08 g. Composites were not fabricated using more than 0.08 g of fibres to avoid large fibre overlapping. Freshly prepared glue was always used for laminate/composite preparation.

2.2 Glue making

White corn flour made glue was used as an adhesive for fabricating laminates and composites. A method popular

in paper bag cottage industry was adopted in the present investigation. To prepare glue, 5 g of flour was taken and thoroughly mixed with 75 ml of water at room temperature. The mixture was then boiled and continuously stirred in a controlled manner by placing the beaker on a hot plate for nearly 45 min to turn it into a thick and translucent sol. It was subsequently allowed to cool down at room temperature for 45 min to stabilize the moisture contained in it. It was used for laminate and composite preparation after thorough mixing.

2.3 Composite fabrication

To fabricate a fibre reinforced laminated composite, a pair of paper strips were taken. A thin layer of glue was applied uniformly on the rough surface of one of the paper strips. The weighed bunch of jute fibre was uniformly spread on the glued surface parallel to the length of paper strip. The other piece of paper strip was glued and carefully placed on the first one. Fibre distribution on paper surface can be visualized in figure 1(a). The jute-paper sandwich was subsequently subjected to light rolling to remove excess glue, air pockets and enhance adhesion between paper and fibre. The green laminate and sandwich were placed between two glass slabs for an hour to avoid its bending. This was followed by oven drying at 60°C.

2.4 Tensile test

Characterization of fabricated laminate and composite were done by conducting tensile test. They were cut into tumbler shaped tensile specimen to avoid their fracture from gripped region during test. A tumbler shaped specimen with its dimensional details is illustrated in figure 1(b). The tests were done using Instron 1195 with a 500 kN load cell. All tensile tests were conducted at a cross head speed of 10 mm/min.

3. Results and discussion

3.1 Stress-strain plot

Typical stress-strain plots representing the behaviour of a single paper strip (bleached kraft paper, *P1*), its fibre free laminate and laminated fibre reinforced composite (containing 0.03 g fibre) under tension are presented in figure 2.

Table 1. Thickness, weight and tensile strength of single paper strips.

Type of paper	Thickness (mm)	Weight (120 × 25 mm), g	Tensile strength (MPa)
Thin bleached kraft paper, <i>P1</i>	0.070	0.155	4.06
Thick unbleached kraft paper, <i>P2</i>	0.173	0.368	14.41
ONP, <i>P3</i>	0.073	0.183	11.88

The figure shows that single paper strip is more stretchable. A nonlinear and gradual increase in stress, up to ~ 6.00 MPa (~ 2.00 MPa above the tensile strength of paper, may be due to some stress transfer to interface/fibre) is followed by linear and steep increase with straining of both fibre free laminate and composite. The similar nature of curves in both the laminate and composite indicates the presence of identical fracture mechanism. That is, the early damage of constituent paper is followed by load transfer to stiffer and stronger boundary between the strips. This indicates that the strengthening comes from the interface which is further enhanced due to the presence of high strength fibre.

3.2 Fibre free laminate

In tables 2a–c maximum load ($P_{\max}$), tensile strength (σ_{ts}) and energy to fracture (E_f) are presented, respectively for single strips (ss), reinforcing fibre free laminates (rfl) and 0.05 g fibre composites (05 fc) from all three papers.

Figure 1. (a) Fibre distribution on paper surface and (b) dimensional details (in mm) of tensile test specimen.

Figure 2. Stress-strain plot of single P1 paper strip, laminated paper and 0.03 g composite.

The superiority of fibre free laminate over single paper strip can be visualized from the above tables. Several fold increase in tensile strength and fracture energy in laminates may be due to introduction of interfaces between the strips. The fracture surfaces of these laminates were observed under SEM and presented in figure 3. It is important to note that P1 (bleached thin kraft paper) made laminate has an open boundary between strips, which may have restricted the growth of crack resulting in better improvement in properties. On the other hand, P2 and P3 made laminates exhibited nearly similar improvement in load carrying capacity, tensile strength and modulus. It is worth to mention that in these laminates boundaries between strips were not visualized and they appeared as single paper strip under the present condition. However, the masking of the thin boundary by the protruding fibres cannot be ruled out.

3.3 Fibre reinforced composite

Figure 4 shows the variation of tensile strength of continuous jute fibre-reinforced thick kraft paper (P2) composite, fibre aligned parallel to the length of the specimen and distributed on 120 mm long and 25 mm wide pair of paper strips, as a function of weight of fibre. Tensile strength data of fibre free laminates are also included in this figure to present an overall behaviour. It is interesting to note

Figure 3. Fracture surfaces of (a) P1 and (b) P2 paper laminates.

that even an introduction of 0.02 g of continuous fibre (equivalent to weight of fibre per unit weight of pair of paper strips = 0.0272 and weight of fibre per unit area of pair of paper strips = $6.67 \times 10^{-6} \text{ g/mm}^2$) resulted in an increase in strength. The introduction of 0.05 g fibre ($1.67 \times 10^{-5} \text{ g/mm}^2$) on an average enhanced the strength

Figure 4. Tensile strength vs wt of jute in jute/P2 composite.

Figure 5. Weight of fibre vs tensile strength and fracture energy in P1 paper-fibre composite.

Figure 6. Weight of fibre vs tensile strength and fracture energy in P2 paper-composite.

from 31.17 MPa (fibre free laminate) to 43.02 MPa, i.e. an increase of 38%. It may be noted that data points are scattered, specially in composites comprising large amount of fibre. The non-uniform distribution of fibre and glue, lack of uniform adhesion between paper and fibre, and use of different batches of glue for fabrication may be responsible for this behaviour.

The effect of weight of reinforcing fibre on average values of tensile strength and total energy absorbed in fracturing P1, P2 and P3-paper made composites are presented in figures 5, 6 and 7, respectively. The data show appreciable increase in the properties on introduction of fibre. The increase in tensile strength and fracture energy of the laminated composite are due to the introduction of high strength continuous jute fibres. In this case the load transfer took place from paper to the fibre through glue. The paper is reinforced by continuous and unidirectional jute fibre with the help of starch glue and can be considered as a discrete phase (Jang 1994). The high energy absorption in fracturing the composite may be due to increased constraint imparted by interface discontinuities and strong and stiff fibre restricting the deformation of

Figure 7. Weight of jute fibre vs tensile strength and fracture energy in P3 paper-composite.

Figure 8. Normalized modulus as a function of fibre per unit area of P1, P2 and P3 based composites.

Table 2a. Maximum load, $P_{\max}$ (N) of single strips, fibre free laminates and 0.05 g fibre composites.

	Single strip ($P_{\max}^{ss}$)	Fibre free laminate ($P_{\max}^{rfl}$)	$P_{\max}^{rfl}/P_{\max}^{ss}$	Composite with 0.05 g fibre ($P_{\max}^{05fc}$)	$P_{\max}^{05fc}/P_{\max}^{rfl}$
P1 paper	3.15	40.7	12.92	105.6	2.59
P2 paper	29.90	136.9	4.58	234.6	1.71
P3 paper	10.3	58.5	5.68	132.8	2.27

Table 2b. Tensile strength, σ_s (MPa) of single strips, fibre free laminates and 0.05 g fibre composites.

	Single strip (σ_{ts}^{ss})	Fibre free laminate (σ_{ts}^{rfl})	$\sigma_{ts}^{rfl}/\sigma_{ts}^{ss}$	Composite with 0.05 g fibre (σ_{ts}^{05fc})	$\sigma_{ts}^{05fc}/\sigma_{ts}^{rfl}$
P1 paper	4.06	22.83	5.62	32.22	1.41
P2 paper	14.41	33.09	2.29	43.02	1.30
P3 paper	11.88	30.50	2.57	36.76	1.21

Table 2c. Fracture energy, E_f (J) of single strips, fibre free laminates and 0.05 g fibre composites.

	Single strip (E_f^{ss})	Fibre free laminate (E_f^{rfl})	E_f^{rfl}/E_f^{ss}	Composite with 0.05 g fibre (E_f^{05fc})	E_f^{05fc}/E_f^{rfl}
P1 paper	0.0022	0.0392	17.82	0.0907	2.31
P2 paper	0.0256	0.1767	6.92	0.2802	1.59
P3 paper	0.0083	0.0602	7.25	0.1247	2.07

Figure 9. Transverse optical view of (a) 0.05 g fibre-P3 and (b) 0.05 g fibre-P2 composites.

paper and glue. This may be partly a result of load transfer (Jang 1994; Hull and Clyne 1996). Jute fibre is not completely brittle and undergoes some plastic deformation. The deformation followed by fracture of reinforcing lignocellulose fibre may have contributed significantly to the fracture energy of composite (Hull and Clyne 1996; Kakisawa and Kakawa 1999). The interfacial debonding and interfacial frictional sliding are also expected to play key role in these composites. It may also be noted that interfacial frictional sliding is potentially most significant source of fracture work. In all three paper based composites a gradual increase in properties are followed by steep

increase. The gradual increase in low fibre containing composites may be due to domination of matrix whereas fibre contributed significantly in large fibre containing composites. In P2 based composites increase in properties are followed by a saturation and drop in properties. This may be due to glue cracking instead of fibre pull-out and fibre breaking (Argon 2000). However, P1 and P3 based composites did not show such an effect. The characteristics of constituent papers may be responsible for their difference in behaviour.

In figure 8 normalized modulus (= modulus of composite/modulus of fibre free laminate) of fibre reinforced

Figure 10. SEM fractographs of (a), (b) 0.05 g fibre-ONP (*P3*) composite, (c) 0.02 g fibre-thin kraft (*P1*) composite, (d) 0.08 g fibre-thin kraft (*P1*) composite and (e) 0.05 g fibre-thick kraft (*P2*) composite.

paper composites, fabricated from *P1*, *P2* and *P3* papers are presented as function of weight of fibre per unit area of pair of paper strips. Increase in modulus are observed with increasing weight of fibre in all three cases. The fractured part of laminated composites are examined

under optical microscope at 50X and transverse view of a few are presented in figure 9. Thin kraft paper (*P1*) and ONP (*P3*) exhibited a significant high energy fibre pull-out mechanism of fracture. This may be due to weak interface adhesion promoted debonding (Ray 2005). The

weak interface may have developed between fibre-glue and/or paper-glue surfaces. On the other hand, fibre breaking dominated fracture in thick kraft paper (P2) indicating relatively strong bonding in P2–paper composite.

The SEM fractographs of composites fabricated from all three different papers have been presented in figures 10(a) to (e). Figure 10(a) shows the fractograph of 0.05 g fibre–P3 composite. Glue starved pockets and fibres of fractured constituent paper can be seen clearly. In addition, there is extensive fibre pull-out indicating weak interface bonding between fibre and P3 paper. The SEM fractograph of same specimen at high magnification (shown in figure 10(b)), shows that the surface of the pull-out fibre is bare, i.e. glue does not adhere on the fibre surface as well as there is indication of some fibre damage. The fibre pull-out and fibre damage reveal effective toughening of composite due to introduction of fibre. Figures 10(c) and (d) show fractographs of 0.02 g and 0.08 g fibre–P1 paper composites, respectively. Fibre pullout along with some fibre breakage are seen in 0.02 g composite. However, 0.08 fibre–P1 composite (figure 10(d)) reveals lack of strong fibre–paper adhesion and high energy fibre pull-out fracture. The fracture surface of 0.05 g fibre–P2 composite exhibits relatively better bonding between fibre and paper (see figure 10(e)). There is no indication of fibre pull-out in the examined region of fracture surface and fracture is dominated by low energy absorbing fibre breakage mechanism. This difference in the mode of fracture is clearly due to different properties of the papers and their constituent fibres. SEM fractographs (see figures 3(a) and (b)) indicate that the constituent fibres in thick kraft paper are mostly coarser than thin kraft paper and ONP. Fine fibres of thin kraft paper and ONP may have enhanced capillary action induced penetration of glue deep into the paper leaving behind less glue to develop strong fibre–glue and paper–glue bonding. A glue absorption test conducted on pairs of thick kraft (P2) and ONP (P3) paper strips shows 0.1598 g and 0.2175 g glue absorption, respectively beneath the exposed surface, under similar conditions. It is worth to mention that moisture can penetrate into these materials by capillary action (Roy *et al* 2001; Ray 2006). Weak fibre–glue and paper–glue interface in the thin kraft paper and ONP perhaps resulted in greater enhancement in tensile strength and energy absorption. However, the role of other physical and chemical characteristics of papers in this matter cannot be ruled out. It is reasonable to mention that the fibre/matrix adhesion is strongly influenced by the status of interfaces (Ray 2005).

4. Conclusions

(I) The fibre free laminates exhibited a few fold increase in load carrying capacity, tensile strength and fracture

energy relative to single paper strip. Improvement in properties are more in P1 paper laminate which may be due to the presence of more open boundary between the paper strips.

(II) Significant enhancement in load carrying capacity, tensile strength and fracture energy have been noticed on introduction of continuous jute fibre when compared to the reinforcing fibre free laminate.

(III) Appreciable improvement in normalized modulus has also been observed due to the presence of fibres.

(IV) Thin kraft paper and ONP made laminates and composites exhibited superior property enhancement than thick kraft paper laminates and composites. The different characteristics of papers and hence different modes of fracture (fibre pull-out and fibre breakage) appear to be responsible for such behaviour of developed composites.

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