In-Line Compression Moulding of Twisted Jute / Polypropylene: Study of Mechanical, Thermal and Moisture Properties

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Abstract

In-line compression moulding involves a single step process in which fibres are directly introduced into the molten thermoplastic in the extruder and the hot extrudate is instantaneously compression moulded. It eliminates additional stage of heating and processing and is suitable for mass production. However, open literature on in-line compression of twisted Jute/polypropylene is scarce. This paper reports properties of twisted Jute fibre/Polypropylene processed by in-line compression moulding by adopting vacuum degassing during compounding in twin screw extruder. The composite specimens were characterised for tensile, flexural and impact strengths, heat distortion temperature and moisture resistance in different media. Thermal study suggested that the processing temperature of Jute to be below 350˚C. Least moisture absorption but higher tensile and flexural degradations were observed in artificial seawater aging. Morphology of tensile fractured specimens revealed good fibre/matrix bonding.

Keywords: *In-line compression moulding, Screw extruder, Jute/ Poly-propylene*

1.0 Introduction

Stringent environmental legislation and consumer awareness force industries to support long term sustainable growth and develop new technologies based on renewable feedstock that are independent of fossil fuels [1]. Application of renewable resources, concern on reduction of greenhouse gases and CO_2 emissions drive plastic industries towards natural fibres [2]. This increases demand for natural fibre reinforced polymer composites (NFRP) and hence the need for commercially viable manufacturing processes [3]. Hygroscopic behaviour and low resistance to temperature above 200°C are the main concerns in the processing of

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natural fibres, which limit the choice of resins [4]. Processing methods of fibre reinforced polymers depends on the nature of the matrix [5]. Polypropylene (PP) is a widely used thermoplastic for industrial applications because it is inexpensive with relatively good performance in terms of strength and ease of processing [6]. NFRP based on PP is promising for several engineering applications [7, 8]. Jute is one of the most thermally stable natural fibres. It is a cost effective material with appreciable mechanical properties and it requires lower energy for processing. Considering its low density, the specific stiffness and strength are comparable to those of glass fibres [9]. It is also explored as a promising reinforcing fibre for thermoplastics with greater focus on PP. Jute as reinforcing fibre increases the crystallinity of PP due to the lignin content and hence melt viscosity increases [10].

Fabrication of NFRP is a challenging task due to the fibre handling issues and fibre intrinsic properties. General thermoplastic composites processing involves two stages, namely, manufacturing of fibre reinforced pellets (compounding) and processing of pellets by injection moulding or compression moulding (off-line process). It involves higher cost along with degradation of mechanical properties due to heating involved in both the stages. This can be overcome by a single step process in which the fibres are directly introduced into the molten polymer matrix and the extrudate is instantaneously processed (in-line process). It involves in-line compounding of matrix with fibres and additives followed by compression moulding of the hot extrudate as a continuous process. This single step process is called in-line compression moulding (ILCM) [11]. ILCM is extensively used for glass/PP and carbon fibre reinforced polymers. Its extension to NFRP is first time attempted in this research.

Temperature, screw configuration and screw speed of twin screw extruders influence quality of compounded Jute/ PP pellets [12]. In natural fibres, thermal degradation occurs above 180˚C which is around the melting temperature of polymers. To mitigate thermal degradation of Jute fibre during processing, control temperature has to be limited to 180˚C. Higher screw speed leads to greater shear force during compounding resulting in better fibre dispersion. Shear heating increases the temperature of extruded compounds by 10 to 20˚C. To minimize thermal degradation by shear heating, screw speed has to be maintained at around 150 rpm. In general, kneading elements can generate high shear stress during compounding and are often used to improve dispersion quality [13].

Jute is hygroscopic and hence it quickly absorbs moisture (5 to 10 wt. %) in the range 1000 to 3000 ppm [14]. Presence of moisture during processing leads to dimensional variations, porosity and degradation. Moisture should be less than 3% for good quality moulding [1]. Moisture has to be removed during compounding and processing of Jute either by air-drying or by vacuum degassing during extrusion [15]. Disadvantages of air-drying include fibre handling, high time and cost along with fibre degradation. This work adopts vacuum degassing during twin screw extrusion.

Joseph et al. [16] studied mechanical properties of sisal/ PP with different orientations of fibres. They reported that specimens with longitudinally oriented fibres were superior to those of transverse or random orientations. Superior properties attributed in the longitudinal direction are due to effective stress transfer to the fibres. Hossain et al. [17] reported that greater tensile strength in longitudinal direction in Jute/ PP is due to its fracture morphology. Among the lignocellulosic fibres, Jute is the most thermally stable natural fibres [18]. Each lignocellulosic fibre has different chemical compositions and hence its thermal degradation behaviour must be known prior to processing. Higher heat distortion temperature allows faster moulding process and it is influenced by fibre content and fibre/matrix interface [19].

Moisture absorption is a concern in NFRP because it reduces interfacial adhesion between the fibre and the matrix [20]. The hydrophilic behaviour of fibres weakens fibre/matrix adhesion and hence deteriorates the performance of the composite [21]. Micro cracks in the polymer matrix enable water molecules to penetrate, while micro gaps between the polymer molecular chains may allow the inward diffusion of water molecules. Deterioration in mechanical properties due to moisture absorption depends on fibre content, fibre orientation, area of exposed surface, permeability of fibre, void content and hydrophilicity of the individual component [22]. George et al. [23] observed that moisture absorption rate in Jute/ PP increased with fibre content, decreased with fibre orientation (longitudinal to transverse) and reduced in chemically treated fibre composites. Rate of moisture absorption is rapid in the initial stage. High cellulose content in natural fibres contribute to greater amount of water penetrating into the interface through the micro-cracks induced by swelling of fibres, thus create swelling stresses leading to composite failure.

Jute is increasingly explored as a promising reinforcement fibre for PP for a wide range of applications. Compression moulding involves higher cost along with degradation of mechanical properties due to heating in two stages. This can be mitigated in an industrial process called ILCM which is a single step process. Reports on ILCM for NFRP are scarce. Vacuum degassing can effectively remove moisture during extrusion and its adoption is scarcely reported in the open literature. Properties of NFRP are controlled by fibre related parameters. Studies on the effect of twisted Jute fibres on the mechanical, thermal and moisture properties of Jute/PP involving ILCM is first of its kind. Co-rotating twin screw extrusion was adopted for compounding Jute fibres with PP. Screw configuration (kneading blocks for optimum shear), barrel temperature (180˚C) and screw speed (150 rpm) were adopted based on industry practice. The specimens were characterized for tensile, flexural and impact properties based on ASTM. Thermal stability was analyzed using Thermal Gravimetric Analyzer (TGA), Derivative Thermogravimetric (DTG) and Heat Distortion Temperature (HDT). Interfacial adhesion and morphology were studied using Scanning Electron microscopy (SEM). Moisture absorption behaviour of the composites in different media such as distilled water, tap water and artificial seawater was studied.

2.0 Materials, Process and Characterization

2.1 Materials

Polypropylene homopolymer Repol (PP AM650N) pellets was used as polymer matrix having melting point of 180 °C, melt flow rate of 65 g/10 min at 190˚C / 2.16 kg (MFI 65). Double twisted continuous Jute fibres are used as the reinforcement in PP matrix. These fibres are NaOH treated to eliminate the wax layer.

2.2 Fabrication of In-Line Compression Moulded Twisted Jute/ PP

Jute/ PP strands were prepared using twin screw co-rotating extruder. The extruder has eight temperature controlled barrels and was operated in co-rotating inter meshing self-wiping mode. As procured PP pellets were fed through the main volumetric feeder to the intake barrel which is provided with water cooling jackets. It moves through the conveying elements to the kneading block in which polymer melting takes place. Twisted Jute fibre fed through the side volumetric feeder mixes with the molten PP. Both mixing and distribution of fibres take place in this section. The compounded material is transferred to the vacuum section from which all the volatiles are evacuated. The compounded Jute/ PP melt strands were compression moulded with 100bar pressure for 30 seconds to produce the laminates. This single stage method of fabrication of Jute/ PP was shown in Fig.1.

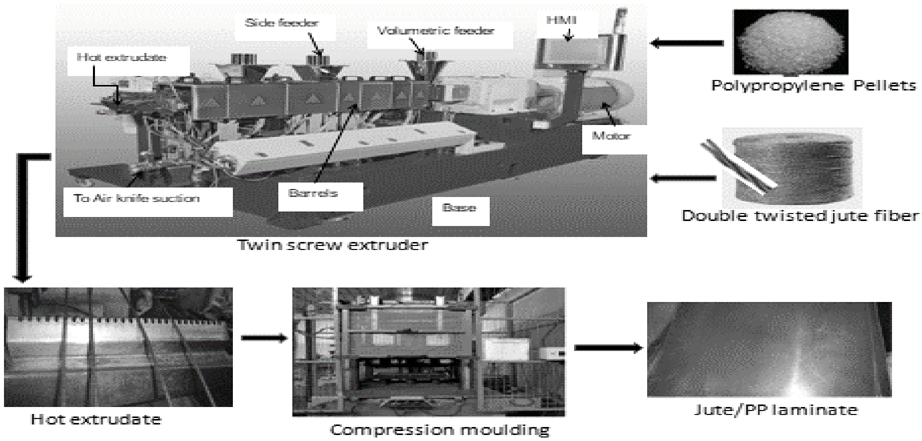

Fig. 1. In-line compression moulding method of twisted Jute/PP

Experiments were conducted on longitudinal and transverse direction specimens by considering PP (70 wt. %) reinforced with 30 wt. % twisted Jute fibre (70/30). Sample size of five was adopted for each experimen.

2.3 Mechanical Characterization

The laminates were cut to the size of test specimens in longitudinal and transverse direction. Tensile tests were performed on 165 x 19 x 3.2 mm^3specimens as per ASTM D638. Flexural tests were performed on 127 x 12.7 x 3.2 mm^3 specimens according with ASTM D790A. Instron Universal Testing Machine, Model–5569A was used for these tests with constant strain rate of 5 mm/min. Un-notched impact strengths were measured from Pendulum Impact Tester Model - Impactor 2, CEAST, as per ASTM D256.

2.4 Thermal Characterization

Heat Distortion Temperature (HDT) tests were performed as per ASTM D648 using Universal Testing Machine (Model – 6921, 6 stations, CEAST, Italy) on 127 x 3.2 x 13mm^3 specimens. The specifications of instrument are temperature range: 20 to 300˚C, thermal distribution: ±0.2 to ± 0.3 ˚C (100 to 250 ˚C), thermal stability: ±0.5 ˚C (300 ˚C) and Heat transfer medium is Silicon oil.

Thermal gravimetric analyzer (model-TGA4000, Perkin Elmer using ASTM E1131) was used to characterize TGA and DTG properties of Jute/PP. The experiments were conducted in the temperature range 30˚C to 500˚C at heating rate of 10˚C/min in Nitrogen atmosphere with a flow rate of 30 ml / min.TGA and DTG curves were plotted for the specimens.

2.5 Moisture Absorption Study

Jute/PP specimens were exposed to moisture in distilled water, tap water and artificial seawater chamber as per testing requirements. The specimens were initially dehumidified in oven and then exposed to different mediums at room temperature for 90 days and moisture absorption studies were carried out. The composition of artificial sea water was according to ASTM D1141.

The specimens were periodically withdrawn from the mediums, dried with tissue paper to remove the adsorbed moisture and weighed using a sensitive balance (± 0.001 g). Moisture absorption was determined using equation (1).

$$M_t = \frac{(\mathrm{m_s} - \mathrm{m}_d)}{\mathrm{m}_d} \times 100 \quad (1)$$

where, M_t = Moisture absorbed at time t (sec), m_s = Mass of aged specimens (gm), m_d = Mass of dry specimens (gm) [24].

2.6 Scanning Electron Microscopy

The tensile fractured surfaces were examined using Scanning Electron Microscopy (SEM) (Hitachi SU 1500), at accelerating voltage of 5000 volt and Emission current of 86000 mA to study the interfacial strength in the specimens.

3.0 Results and Discussion

3.1 Influence on Mechanical Properties: Tensile, Flexural and Impact Strength

In general, mechanical properties in the longitudinal direction were superior to those of transverse direction (Fig. 2). Superior properties in the longitudinal direction are due to effective stress transfer to the fibres. In the present study the improvement in strength is attributed to twin screw extrusion. The load is effectively transferred from the matrix to the fibre.

In 30 wt. % continous double twisted Jute/PP specimens, highest tensile strength of 29.15 MPa was yielded in longitudinal direction. The greater tensile strength in longitudinal direction is because of the Jute fibre strength. The resultant tensile strength in the present work is in agreement and superior to the elsewhere reports [25].

Flexural behaviour of the specimens were similar to that of tensile behaviour (Fig. 2). Improvement in flexural strength is due to homogenous distribution of fibres in the matrix. Flexural strength increased by 6.04 MPa in longitudinal direction. Flexural modulus was superior in longitudinal direction. Flexural modulus increased by 2.54 GPa in longitudinal direction and the improvement was 1.32 GPa in transverse direction with that of PP (1.35 GPa). The composite specimens of the present study showed greater impact energy. Improvement in un-notched impact strength in transverse direction was 24.6 % and was 13.5 % in longitudinal direction.

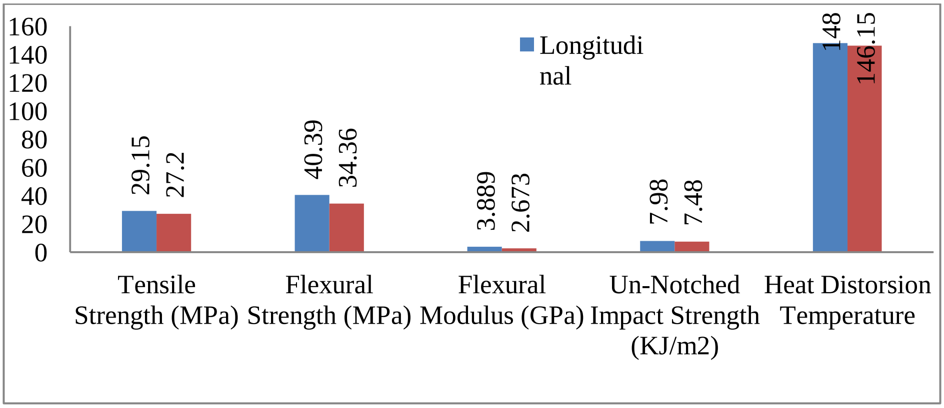

Fig. 2. Influence of fibre direction on in-line compression moulded twisted Jute/PP

3.2 Morphology of twisted Jute/PP

Tensile fractured surfaces were examined under SEM and the micrographs revealed superior fibre/matrix bonding, good fibre wetting and uniform fibre distribution in all the specimens. Less micropores or voids are observed due to vacuum degassing during the fabrication (Fig. 3a). Specimens cut in transverse direction shown in Fig. 3b clearly evidenced that load is transferred through the fibres in transverse direction also. It directly indicates better fibre/matrix bonding.

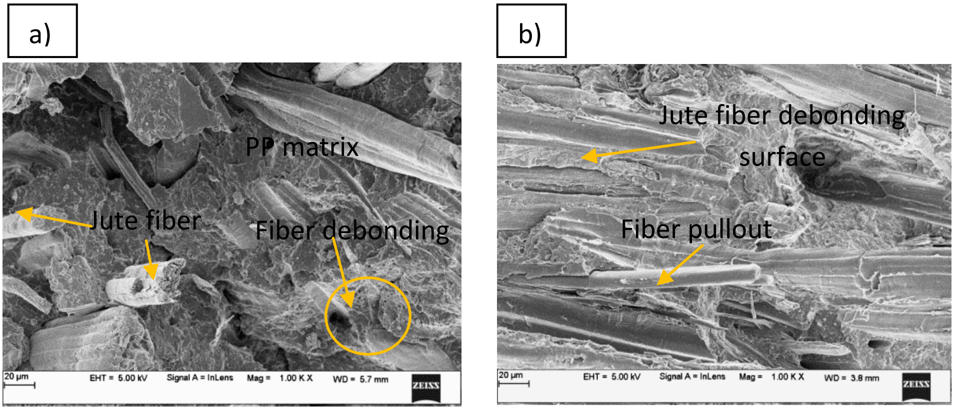

Fig. 3. SEM micrographs of tensile fractured specimens (a) 30 wt.% longitudinal direction twisted Jute/PP and b) 30 wt.% transverse direction twisted Jute/PP

3.3 Thermal Properties

3.3.1 Heat distortion temperature of twisted Jute/PP

Heat distortion temperature of PP is 105˚C, it increased with fibre content and was greater in longitudinal direction 30 wt. % twisted Jute/ PP. Highest heat deflection indicates higher load bearing capacity. Heat distortion temperature was greater by 29.1% in longitudinal direction and 28.7 %in transverse direction when compared with that of PP. The influence of fibre direction on Heat distortion temperature is shown in Fig. 4.

3.3.2 DSC of twisted Jute/PP

Melting temperature and crystallization temperature varied nominally with the addition of fibre to PP. From TGA, it is clearly observed that the residual increased with fibre content. DTG thermogram of Jute/PP showed decomposition in four stages. The first peak corresponds to evaporation of adsorbed water from the fibres. The second peak corresponds to decomposition of hemicellulose, decomposition of α-cellulose as strong third and the fourth peak indicates thermal degradation of PP. DSC thermogram of 30 wt.% twisted Jute/PP is shown in Fig. 4.

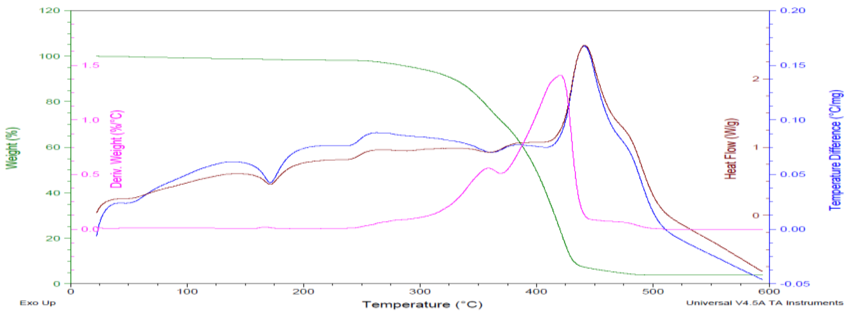

Fig. 4. TGA and DTG curves of twisted 30 wt. % Jute/PP under nitrogen atmosphere

3.4 Moisture Diffusion Properties

The specimens were initially dried in hot oven at 70 ± 2 °C [20] and the weight loss was measured after each hour. The procedure was repeated till the weight of the specimen remained constant, which indicates the specimens are completely dried. The specimens were completely dried in 4 hours. Based on this observation, all the specimens were dried for 4 hours in hot oven at 70 ± 2 °C before immersing in the medium for moisture diffusion study. Sample size of three was adopted for all the tests.

Table 1. Influence of fibre direction on saturation moisture and time to saturation in the three diffusion media

Fibre content (%)	laminate direction	Saturation moisture, M_∞ (%)			Time to saturation, t (hours)		
		distilled water	tap water	artificial seawater	distilled water	tap water	artificial seawater
30	Lg	8.32	7.97	6.97	1800	1950	2200
	Tv	8.43	8.09	7.09			

Moisture absorption was gravimetrically determined using equation (1) in each medium. Fig. 5 shows influence of diffusion media in 30 wt. % Jute/PP in both longitudinal and transverse direction specimens. Moisture absorption is greater in distilled water than that of tap water and artificial seawater. Moisture absorption is directly related to the size of molecules (least of the three in distilled water) of the diffusion medium. In 30 wt. % Jute/PP, moisture saturation occurred at 1800 hours in distilled water, 1950 hours in tap water and 2200 hours in artificial seawater. Moisture absorption was nominally greater in transverse direction than that of longitudinal direction in all the three media. The saturation moisture absorption and time to moisture saturation in three media with fibre direction were presented in Table 1.

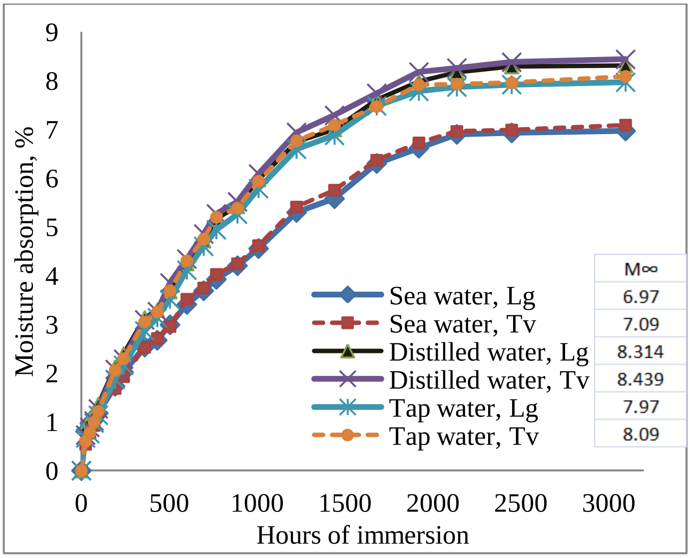

Fig. 5. Moisture absorption in three media - 30 wt. % Jute/PP in longitudinal and transverse directions

3.4.1 Retention of tensile and flexural properties

Tensile strength (Fig. 6a) decreased drastically due to moisture absorption in all the three media and the decrease was highest in seawater. Decrease in tensile modulus was highest in seawater and lowest in distilled water (Fig. 6b).

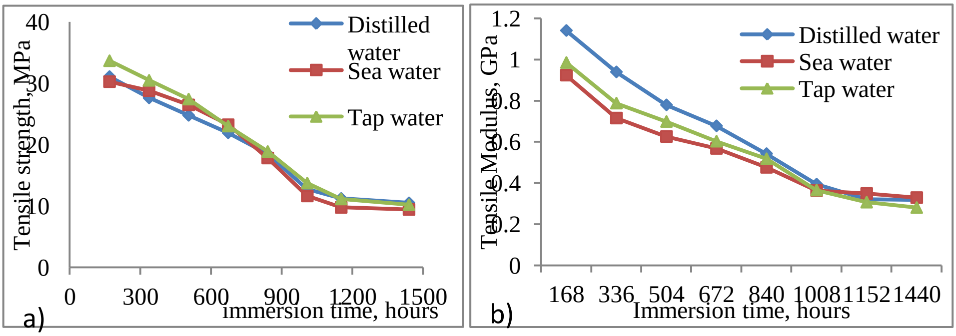

Fig. 6. Influence of diffusing media on a) tensile strength and b) tensile modulus

Flexural strength of the specimens drastically decreased in all the three media (Fig. 7a). The extents of decrease were 47.9 MPa in distilled water, 46.43 MPa by sea water and 40.4 MPa in tap water. Flexural modulus decreased drastically till 37 days of exposure and the decrease was nominal for further immersion. The decrease was highest in seawater and lowest in distilled water (Fig. 7b).

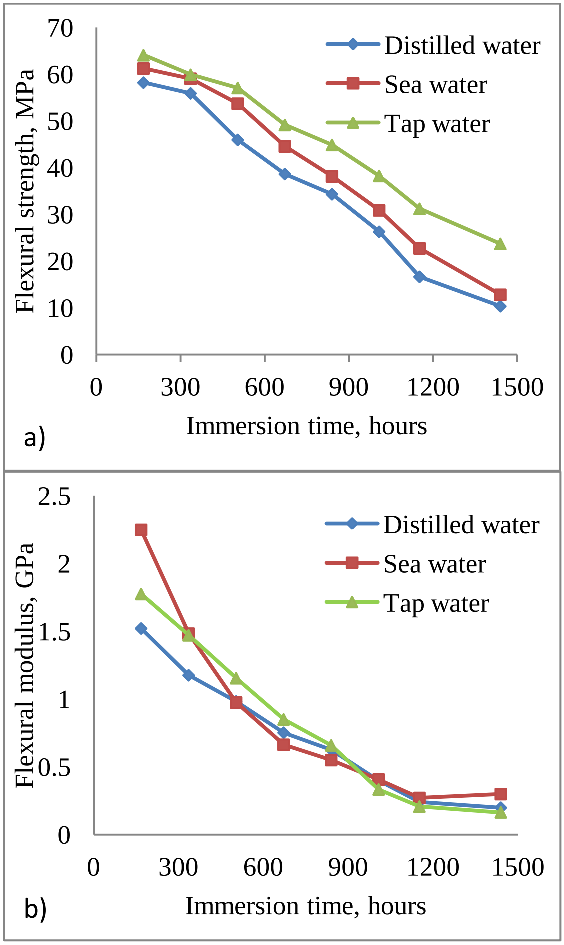

Fig. 7. Influence of diffusing media on a) Flexural strength and b) Flexural modulus

3.4.2 Scanning electron microscopy

Scanning electron micrographs of tensile fractured specimens before and after 60 days of aging were studied. The micrographs revealed breakage of Jute fibres (Fig. 8a) indicating that the load is transferred through the fibre. Influence of diffusing media was clearly observed at the fibre surface ends. Uneven cracking was observed in the fibres in case of distilled water with no influence of PP matrix (Fig. 8b). Artificial seawater diffused through the fibre/matrix interface creating gap between the fibre and the matrix (Fig. 8c) with clear influence of seawater on PP matrix. Influence of tap water was less than that of seawater on PP (Fig. 8d).

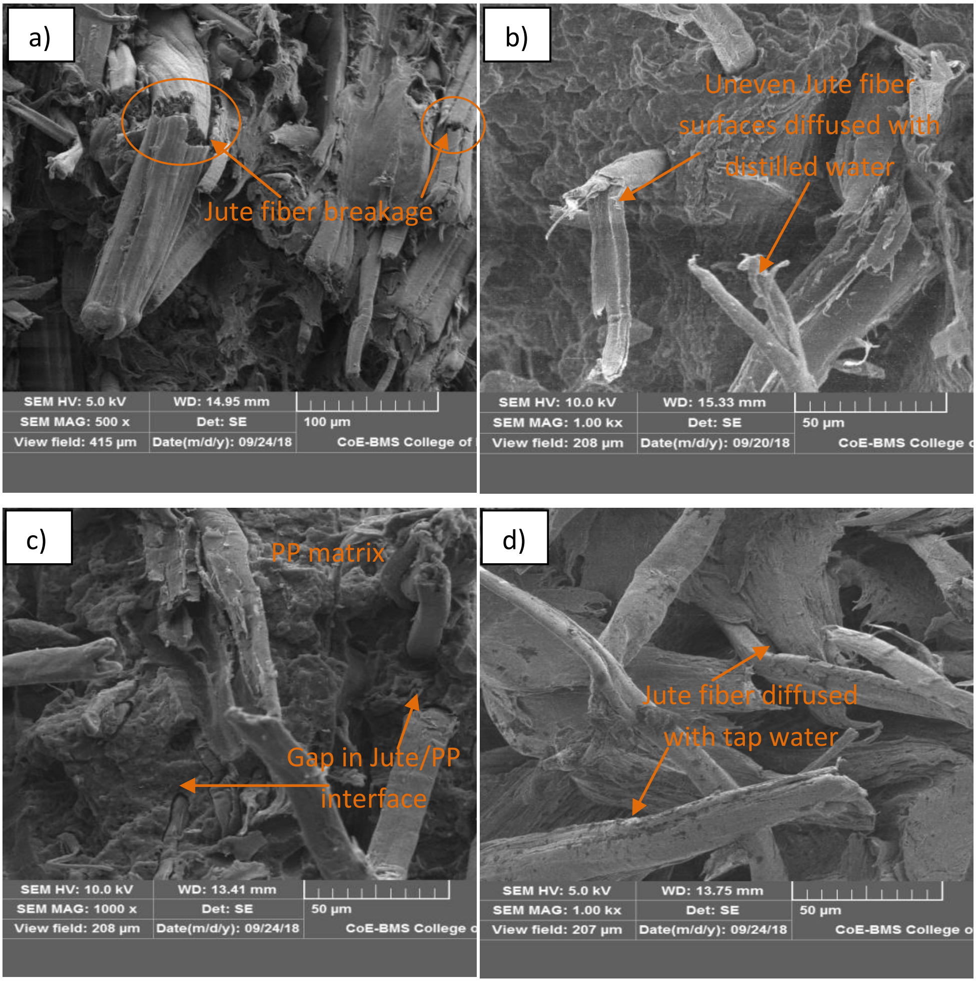

Fig. 8. Tensile fractured 30 wt.% Jute/PP SEM micrographs a) before aging, b) after 60 days distilled water diffusion, c) after 60 days' immersion in seawater and d) after 60 days' immersion in tap water

4.0 Conclusions

Jute/PP composites were fabricated by In-line compression moulding method. Vacuum degassing was adopted in twin-screw extruder during compounding instead of fibre drying and chemical treatment. Mechanical, thermal and moisture properties were studied in 30 wt. % double twisted Jute/PP composites in longitudinal and transverse direction. Based on the experimental results, the following conclusions were arrived at:

- Tensile strength in the longitudinal direction was greater than those in transverse direction. Flexural strength was greater in the longitudinal direction and the improvement was around 6.2 MPa when compared with that of PP. Flexural modulus was superior in the longitudinal direction specimens and the improvement was around 2.54 GPa when compared with that of PP. Un-notched impact strength improvement was 24.6 % in transverse direction specimens when compared with that of PP.
- Heat distortion temperature was greater by 29.1 % in longitudinal direction specimens when compared with that of PP. Thermal resistance of PP decreased with fibre content suggesting the processing temperature of Jute to be below 350˚C. DTG curves evidence improved Jute/PP adhesion resulting in improved thermal properties.
- Moisture absorption was least in artificial seawater and highest in distilled water. Effect of fibre direction on moisture diffusion was nominal. Degradation in tensile and flexural strength was highest in seawater aging and lowest in tap water aging. But, degradation in tensile and flexural moduli was highest in seawater and lowest in distilled water.

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