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Investigation of TiO_2 Nano Filler in Mechanical, Thermal Behaviour of Sisal/Jute Fiber Reinforced Interpenetrating Polymer Network (IPN) Composites

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In this work, various proportionate (0%, 1%, 3% 5% & 7%) of titanium di-oxide nano-filler was utilized as particulate matter along with the sisal/jute fiber reinforcement, in the view to increase the physical properties of the composite materials. Beside, as matrix material the mixture of Epoxy (E) and Polyurethane (PE) was chosen with the proportionate of 70 and 30 wt.% respectively, in order to extract the specific qualities of both the matrices to achieve the interpenetrating polymer networks (IPNs). Moreover, to exactly find out the thermo-mechanical characteristics of the sisal/jute fiber reinforced IPN composites, tests like thermo-gravimetric analysis (TGA), tensile, flexural, Impact, short beam strength and water absorption tests were carried out as per standards. It was found that, incorporation of 5% of TiO_2, increases the mechanical properties such as tensile, flexural, impact and short beam strength. Similarly, incorporation of TiO_2 into IPN matrix enhances the thermal stability and water absorption resistance. All the obtained values of various filler weight % of TiO_2 were compared with each other against with and without particulate incorporated IPN laminate in the purpose of using the same in construction industries.

Keywords: *Epoxy, polyurethane, TiO_2, thermo-gravimetric analysis, mechanical properties.*

1. Introduction

Nowadays, natural fiber reinforced composites are currently becoming one of the focal topic due to their wide range of applications like low-cost, non-toxic, biodegradable, and easily recyclable. Because of its unique nature and good biocompatibility, it could be employed to substitute commonly used chemical fibers such as carbon and glass fibers[1-3]. Natural fiber reinforced with different matrices have been explored in recent studies moderately, in that about 15–20 percent is being employed in the vehicle industry and even more exclusively used in construction industries due to all applications[4-6]. In the researcher's point of view, natural fibers are generally divided into two categories: plant-based and animal-based. In the first category of plant fiber base, sisal, hemp, cotton, coir, pineapple, and kenaf are holding the main role in developing the natural fiber (plant fiber based) based composites[7]. Though animal fibers not having that much significant importance due to their unavailability as well as scarcity, still it passively holds as the second position in developing animal based fibers which includes silk, feathers, and wool etc., as the base material. Moreover, depending on the various applications, the fibers can be combined with thermoplastic elastomers and thermosetting polymers. Mostly the thermoplastic networks include polypropylene and polyethylene resins, while the thermosetting matrices are epoxy, vinyl ester, polyester, and polyurethane[8,9].

Among all sets of natural fibers available in the market, sisal and jute play the major role and are extensively employed in most of the industrial and commercial applications. In fact, the main reasons behind those fibers in the field of natural fiber reinforced composites are their cheapness and easy availability and processing[10]. Sisal is a fiber extracted from the Agave plant that is native to Tropical america. It is a tough shrub that thrives in hot conditions. Furthermore, it is capable of growing in dry places that are typically inappropriate for plants[11-13]. Similarly, Jute fibers are made from the plant called genus Corchorus of the Malvaceae family. Jute fibers are renewable and biodegradable and reusable, making them environmentally sustainable distinct processing methods are employed indeed. Likewise, Jute fibers have excellent insulation and strength properties, with moderate water absorption regain and no skin problems while having contact with the humans. On the other hand, the recent annual global production of jute fibers is estimated to be around 3 million tonnes as well which is used for a variety of applications all around the world[14]. Jute backpacks have

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gained in popularity as an eco-friendly alternative to both non - compostable poly bags derived from oil and garbage bags that require a significant amount of wood. Normally all kinds of natural fibers are extracted from the plants through the method called decortication, in which plant leaves or roots are crushed, brushed, beaten either manually or mechanically, and finally sent through rotating wheels in the intention to avail lengthy fibers[15,16].

In the due course, various abstraction methods and procedures are employed to extract the natural fibers to compete with the artificial fibers. Even though it become as the moderate substitute of synthetic fibers, still it is not completely believed as the suitable replacement of the synthetic fibers. Hence to improve the strength of the fibers various techniques were used to get the superior final product, in that sequence, techniques like utilizing the different sizes of the natural fibers as reinforcements as well properly choosing the appropriate alkali treatment methods, are nowadays widely used to get the required strength and compatibility, especially in the area of fiber strengthening aspects, dramatic research activity is happening in the past few decades. In line of the above, treatments like alkaline, acetylation, acrylation, bleaching, bezoylation, isocynate, silane are the universally and widely adopted methods to strengthen the fibers[17,18]. Additionally, matrix material also exclusively plays a wide spectrum of role in increasing the strength of the composite laminate. In that sequence, thermoset matrices like epoxy, vinyl ester and polyurethane generally play a vital role in structuring the composite material based on the user preference[19]. Though each material has had some unique characteristics while opting for fabrication purposes, manufacturers or users often complain of some service/service defects. To overcome the issues encountered in each matrix, the concept of mixing the two different sets of monomers has been proposed by chemists in the name of interpenetrating polymer networks (IPNs). Numerous research activities are presently happening in these particular IPNs, in order to exactly use the final matrix material to the various industrial applications[20,21]. In addition to that, to further strengthen the laminates to the discrete level, at the present time, particulate fillers have made remarkable contributions. Furthermore, numerous researchers have suggested and tried out different types of nano fillers in their own accord. Mostly, fillers are broadly classified as organic and inorganic fillers, in the family of series of inorganic nano fillers, widely accepted nano fillers are aluminium trihydrate, ammonium polyphosphate and Titanium dioxide etc., Among all nano fillers, TiO_2 have been considered as the special one as reinforcing agents because of its unique characteristics like high refractive index, hydrophobicity and non-toxicity. Though the separate mechanical analysis have been performed as substituting the TiO_2 as the particulate matter with the neat thermoset resins, the concept of incorporating the TiO_2 with IPNs along with the natural fibers are not completely exploited. In the view to meritoriously utilize the remarkable contributions of the IPNs and filler particulate (TiO_2) the concept of particulate filled natural fiber reinforced IPN laminate has been conceptualized[22,23].

During the course of this study, sisal and jute fiber have been taken as the reinforcement agent whereas the combination of epoxy and polyurethane (IPNs) has been taken as the matrix material. Besides, various proportions of TiO_2 (0%, 1%, 3% 5% & 7%) is taken as the particulate matter along with the IPN (epoxy and polyurethane) mix. All through this study, the chosen reinforcement fibers are sisal and jute, singly each fibers (sisal and jute) have been selected as the reinforcing agent, also along with that combination of sisal/jute fibers separate laminate have been be prepared in the intention to fabricate the hybrid composite with various proportionate of particulate matter. As well, to test the formed laminate, tests like TGA, tensile, flexural, impact, short beam shear strength and water absorption tests are conducted on the fabricated laminate. The obtained results were compared to each other to evaluate the influence of particulate matter in the IPN matrix[24,25].

2. Materials and Method

2.1. Preparation of titanium dioxide nano powder

During the preparation of titanium dioxide, Nano powder as such received from the suppliers were used for the processing without any further purification. Likewise, titanium tetra iso-propoxide and heptane are also purchased from Chin Chin Chemicals for the chemical processing. Along with that, the required amount of heptane and propoxide was taken with a percentage of 29.3 ml and 100 ml respectively. Additionally, these solutions were physically stirred for the period of 2 hours. Above and beyond, the required amount of distilled water (8 ml) was slowly added to the mixture and it was kept at ambient temperature for the period of 80 hours. Finally, the obtained mixture was filtered with normal filter paper following which the calcination process was carried out for the period of 24 hours by maintaining the temperature of 80°C likewise again raising the temperature of 220°C for the period of 2 hours[26].

2.2. Preparation of ipns and titanium dioxide blend

To get the exact proportionate of the IPNs, the required amount of epoxy and polyurethane (70:30) wt. ratio was mixed and their viscosity also maintained, by placing the blend at the temperature of 80°C, for the period of 20 minutes as prescribed from the supplier data sheet as mentioned in the Table 1.

Following the mixing procedure, the various proportions (0%, 1%, 3% 5% & 7%) of TiO_2 was taken separately by weighing with accuracy of ±0.1 mg. While preparing the TiO_2 for the fabrication purpose, the particulate was dried completely by maintaining it at the temperature of 100°C. Most of the studies suggested that, it is the best method to disperse the particulate into the matrix was mechanical stirring, in this

Table 1. Property of raw materials[14].

Properties	Epoxy	Polyurethane
Density (g/cm^3)	1.15	1.45
Tensile strength (MPa)	74	46.43
Tensile modulus (GPa)	3.74	2.12
Poisson's ratio	0.32	0.24

work also high speed magnetic stirring was adopted followed by the sonication processes. The prime reason behind that of stirring was that, magnetic stirring might decrease the bubble formation and tend to reduce the void content significantly. The matrixes with TiO_2 of variant weight proportions were stirred for the period of one hour to completely disperse the nano particles into the matrix to avoid the agglomeration issues. Also, the required amount of hardeners like HY951 grade and MOCA was added for epoxy and polyurethane respectively. The IPN blend with titanium dioxide mixture was used for the final fabrication processes[27].

2.3. IPN laminate preparation

Initially to do the fabrication process, exact proportionate of particulates were taken along with the premixed matrix IPN Blend. The exact schematic processes representing the entire fabrication processes were represented in the Figure 1[28].

From the fiber preparation perspective, the chosen sisal and jute fiber was already dried at ambient temperature to completely remove the water absorption content and their corresponding physical properties were mentioned in Table 2.

Secondly the 60:40 wt.% ratio of (weighed) pre-cut sisal/jute fiber and matrix was taken for the laminate preparation, following which through the hand-layup technique the laminate were prepared by placing the fiber mat and matrix one over another. In the same way, the formed laminate was placed in the compression mould which was having the dimension of 300 x 300 x 3 mm, following this; required amount of pressure (12 MPa) was applied over the laminate, by maintaining the temperature of 100°C to form the hybrid IPN laminate. In fact, to thoroughly understand the various combinations of fiber/matrix and filler materials, a different set of combinations of natural fiber reinforced (particulate filled) IPN composite was fabricated as mentioned in the Table 3.

2.4. Thermo-gravimetric (TGA)

TGA was performed on all fifteen sets of sisal/jute fiber reinforced hybrid IPN laminates to determine their thermal stability and compare each other towards to predict the laminate behaviour. The testing was conducted on a Shimadzu DTG-50 instrument in a nitrogen atmosphere (40 ml/min). Hybrid IPN laminate specimens with a volume of 7-8 mg each were heated from 30°C to 530°C at a steady rate of 10°C/min[29].

2.5. Methodology

The fabricated natural hybrid laminate with various combinations of filler material was systematically investigated through the different mechanical tests to thoroughly understand the physical behaviourism of the IPN laminate. Nevertheless, to carry over the above processes different tests were performed on the IPN composite as shown in Table 4[30].

2.5.1. Tensile test

To do the tensile test analysis, specimen edges were firmly fixed on the universal testing machine chucks and the load was applied in longitudinal direction until the specimen fracture. Besides, during the testing procedure, cross head speed of 2 mm/min was maintained.

2.5.2. Flexural test

The specimen was laid horizontally over the two contact points and vertical load was applied on the top of the specimen till the sample specimen fractures. All through the course of study, cross head speed of 5 mm/min was maintained.

2.5.3. Impact

The impact test specimen was initially placed in the charpy test work piece holder and allowed the weight to fall

Table 2. Properties of sisal and jute fibers[10-12].

Properties	Units	Sisal fiber	Jute fiber
Density	g/m^3	1.5	1.3
Elongation at break	%	2.2-2.7	1.4-1.9
Tensile strength	MPa	514-720	382-774
Young's modulus	GPa	10.2-22.5	27.5
Cellulose	%	66	62-72
Lignin	%	10.2	12.5-13.5
Microfibrillar angle	-	23	9
Wax	%	2.2	0.6
Hemi-cellulose	%	23	15-21
Pectin	%	11	0.3
Ash	%	0.6-1.1	0.6-2.1

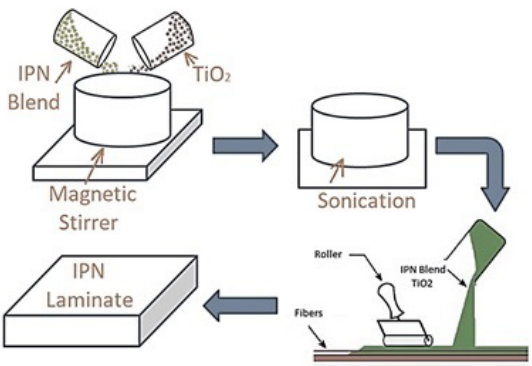

Figure 1. Schematic view of fabrication of Sisal, Jute, Sisal/Jute (TiO_2 loaded) IPN composite.

Table 3. Combination of sisal/jute hybrid IPN composites.

Fiber	IPN – Combinations (Epoxy/Polyurethane)	TiO_2				
		0%	1%	3%	5%	7%
Sisal (S)	70:30 (wt.%.)	SIP0	SIP1	SIP3	SIP5	SIP7
Jute (J)		JIP0	JIP1	JIP3	JIP5	JIP7
Hybrid (SJIP)		SJIP0	SJIP1	SJIP3	SJIP5	SJIP7

S – Sisal, J – Jute, E - Epoxy, PU – Polyurethane.

over the specimen and their corresponding energy absorption values were noted down. To do all sets of test procedures, five specimens from each proportionate were identified and their corresponding results were averaged.

2.5.4. Short beam shear strength

To do shear beam shear test, the specimens were placed on the horizontal shear test fixture by maintaining the fibers were parallel to the loading edge. As well, the beam strength was calculated with help of the formula mentioned in the Equation 1.

$$Short\ beam\ shear\ strength = \left(3 \times Peak\ load\right) / \left(4 \times breadth \times thickness\right) \quad (1)$$

Further to this, the entire procedure was done by keeping the cross head speed of 2mm/min.

2.5.5. Water absorption

To actually carry out the standard water absorption test on composites, during this course of study, specimens were thoroughly dried with (2 hours) help of the oven to make sure that specimens were completely water absorption free. After cleaning the specimens by manual process, their initial weight was measured with an electronic weighing instrument to the level of ±0.1. After weighing was done, the specimens' edges (sides) were completely coated with epoxy resin to avoid the water penetration through the fiber matrix bonding areas. At last the specimens were placed in the deionized water for the specific duration. In this particular study, the specimens were exposed to the humid environment for the period of 12 months. Moreover, to get to know the water absorption in a specific interval of time, specimens were taken from every interval and cleaned with linen free cloth and weighed. As well, the percentage of water absorption was calculated by the variation obtained from the initial weight and final weight. The following Formula 2 was mostly used in calculating the water absorption ratio of all composites[31].

Table 4. ASTM standards[30,31] and dimensions of the specimen.

ASTM Standards	Size of the specimen (Length x breadth x thickness in "mm")
ASTM D 3039 - Tensile	250 x 25 x 3.2
ASTM D 790- Flexural	127 x 12.7 x 3.2
ASTM D 256 – Impact	63 x 12.8 x 3.2
ASTM D 2344 – Short beam shear	18 × 6 × 3
ASTM D 570 – Water absorption	⌀50.4 x 3.2

$$\Delta M\left(t\right) = \left(\left(m_1 - m_2\right) / m_2\right) \times 100 \quad (2)$$

Where, ΔM (t) – Water uptake at time "t", m_1 – wet weight, m_2 – dry weight.

2.6. Fractrography

The JEOL JSM-6480LV instrument was often used to examine the Fractrographic characteristics of sisal/jute fiber reinforced hybrid composites. Across most application fields, samples are gathered over a specific area of the material's surface, and a two-dimensional image displaying spatial heterogeneity characteristics was generated. The morphological study can also be used to analyse specific locations on the sample; this method was particularly useful in evaluating the surface chemistry, crystal structures, and crystallographic structure in a descriptive or moderate manner. Specimens were also fixed with base coat before going through graded alcohol dehydration series to prepare them for the analysis. The samples are fully prepared to be interpreted on the SEM once the gold coating was already completed[32].

3. Result and Discussion

3.1. Thermo-gravimetric

The thermo-gravimetric analysis of the sisal, jute, sisal/jute (hybrid) fiber reinforced (TiO_2) particulate loaded IPN composite were shown in the Figure 2 (a) (b) (c). It was observed that, irrespective of fiber reinforcement, the significant weight loss was observed up to the level of 5%, during the initial time, this effect or weight loss was purely because of the removal of moisture content from the IPN composite. Subsequently major weight loss was witnessed from all the graphs up to the level of 75%, at higher temperatures due to the effect of matrix degradation as well volatilization of IPNs. After the test specimens were subjected to higher temperatures, residue formation had been visualized such a way that maximum weight loss would be accomplished; this was called the final weight loss. The initial and major weight loss of the sisal, jute, sisal/jute fiber (particulate loaded) reinforced composite temperatures were given in the Table 5.

It was clearly found that, irrespective of fiber reinforcement, all kind fiber reinforced IPN composite specimens had shown the same set of TG values. However, the particulate addition into the IPN composite significantly shows a higher TG value than the neat IPN resin (without particulate loading) composite. To prove this, a particulate (sisal fiber reinforced) loaded specimen gives the TG value as 310°C, 340°C, 380°C and 410°C for 1%, 3%, 5% and 7% respectively.

Table 5. TGA – analysis of sisal, jute and sisal/jute (particulate loaded) hybrid IPN composite.

Fiber /Particulate	0%	1%	3%	5%	7%
	WL (°C)	WL (°C)	WL (°C)	WL (°C)	WL (°C)
SISAL	480	490	495	510	515
JUTE	445	485	490	512	513
SISAL/JUTE	465	492	492	514	516

WL – Weight Loss at 75%.

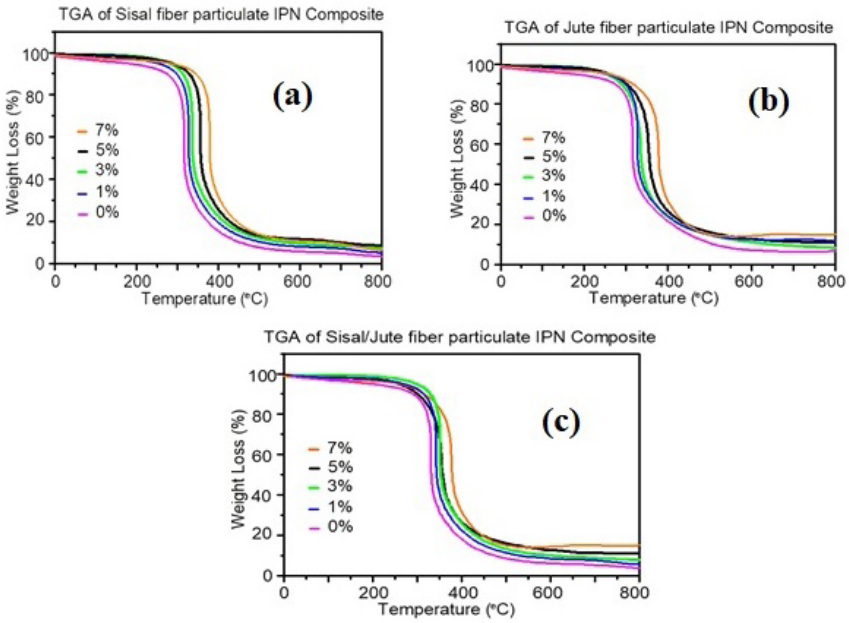

Figure 2. Thermogravimetric analysis of (a) sisal (b) jute (c) sisal/jute fiber reinforced (TiO_2) particulate loaded IPN composite.

During the study, it was observed that, the 1% of loading of particulates had substantially raised the TG value as 4.8%, this uptrend was kept seen upon loading of particulates into the IPN composite. Likewise, 3%, 5% and 7% addition of particulates had shown the rise in value of nearly 9.5%, 11.5%, and 7.8% respectively. In the same way, jute fiber reinforced particulate loaded composite as well shows the same trend as such positive trend line shown in the sisal fiber, even the hybrid fiber as well showed the same hike in trend line as seen in the sisal and jute fibers. Mostly all the fiber reinforced composites had shown the equal rise in trend line upon loading of particulate matters. From the study, it was clearly understood that, the fiber reinforcement had not shown any remarkable impact on the TG values, in-turn particulate loading into the IPNs shown the significant rise in the TG value. The additions of particulate matter into the IPN composite greatly rise the TG value to distinguished manners. Actually, the particulate fillers and IPN blend inevitably makes a better mutual entanglement mechanism between them and vice versa enhances the improved heat resistance upon the fiber and resins. Since particulate matter improvises adhesion property between the fiber and resin amongst the IPN laminate, this effect directly enriches the TG value to notable level.

3.1. Tensile strength

The tensile strength and modulus of various proportions of particulate loading of TiO_2 were shown in the Figure 3.

During the initial investigation on the specimens with 0% (pure resin loaded) particulate loading had shown the tensile strength value of 38.46, 22.41, 30.53 MPa for SIP, JIP and SJIP respectively. However, it was found that the sisal fiber reinforced specimens had shown the higher strength value as compared with the jute fiber reinforced IPN composites. This effect predominately shows that sisal fiber transfers

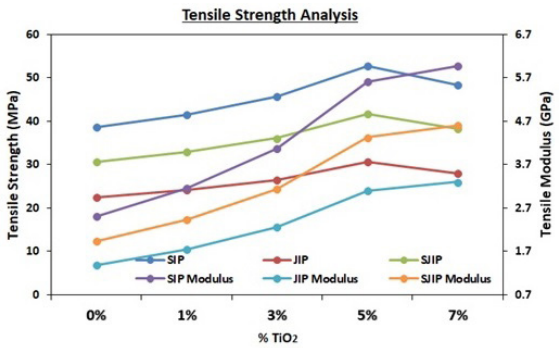

Figure 3. Tensile strength analysis of (a) sisal (b) jute (c) sisal/jute fiber reinforced (TiO_2) particulate loaded IPN composite.

higher levels of stresses as compared with their counterpart jute fiber. As well, the common failure procedure starts from cracks on the matrix, follows with the detorting the interfacial stresses between the fiber and matrix and at last ends up with the fiber scissoring, pull out and fiber breaking. All the above fiber breakages as well were observed on all the specimens irrespective of the particulate loading and fiber reinforcement. Whereas, particulate loading significantly improves the laminate strength to a considerable level. This was evident on all the specimens upon loading of the particulate matter into the IPN laminate. Moreover to prove this, the 1% particulate addition into the IPNs had shown the stress value as 41.53, 24.09 and 32.81 MPa for SIP, JIP and SJIP respectively. Yet evidence upon the particulate addition, IPN laminate shows the substantial level of strength advancement in all levels of incorporation of titanium dioxide. Upon loading the 1% of particulate, the difference of climb was seen as 6.95%, 6.54% and 7.12% for corresponding

SIP, JIP and SJIP. Similarly the 3% addition of particulate into the IPNs showed the significant rise in stress value, the observed values as 45.62, 26.46 and 36.04 MPa for SIP, JIP and SJIP individually. In turn their respective level of climb was observed as 8.96%, 9.12% and 9.01% for all SIP, JIP and SJIP. Again the same trend of increased strength value was observed upon adding the 5% of titanium dioxide. But the observed values were quite high in all levels of fiber reinforcement; the obtained numerals were 52.72, 30.58 and 41.65 MPa for SIP, JIP and SJIP respectively. In this above connection of increasing trend in all sets of strength values were shown that, the titanium dioxide extensively plays the major role in stiffening the laminate, the added particulate matter completely diffuses into the matrix material in large, and evenly distributes into the IPN blend as much as possible. Besides, the incorporated particulates were filled deep into the voids present in the fibers and creates the better interfacial adhesion between the fiber and matrix, due to this reason, the stress transfer was happening in the linear manner along the direction of fiber and laminate bears the higher strength value, all this effect were evident in the graphs upon addition of particulate into the matrix material. As such, an increasing trend of values observed in the tensile stress graphs, their corresponding modulus value also indeed shows the same level of increasing effect by loading the particulate matter into the IPN matrix. To evidence this, the neat IPN laminate (without particulate loading) showed the modulus value as 2.51 GPa for SIP, whereas the increase upon loading the particulate matter, it shows the value of 3.15 GPa as modulus value. However, this trend of increase in modulus value was continuously observed in all sets of IPN laminate's with due respect of addition of particulate matter into the matrix. The actual reason behind that increase in modulus was directly correlated with the stiffening effect of IPN laminate. The addition of the particulate matter into the matrix inversely increases the modulus level and directly increases the tensile strength level irrespective of all fiber reinforcement and matrix material[33,34].

3.2. Flexural strength

The flexural strength and modulus with various proportions of particulate loading of TiO_2 were shown in the Figure 4.

While doing the flexural strength analysis, as observed from tensile strength results, the SIP specimens were shown a higher level of flexural load bearing capacity as compared with the remaining all set of IPN composites such as JIP and SJIP. To evidence this, the neat IPN (without loading of particulate) had shown the flexural strength value as 42.64 MPa, whereas their counterpart JIP shows flexural strength value as 31.94 MPa. Similarly their combination (hybrid) had shown the flexural strength value as 37.29 MPa. Equally, upon loading of the particulate matter into the IPN matrix significantly increases their flexural strength value to marginal level. To mark this, IPN laminate's with various reinforcement shows the strength value as 44.52, 33.35 and 38.93 MPa for 1% TiO_2 loading into the matrix for the fiber reinforcement of SIP, JIP and SJIP respectively. Nevertheless, the 3% particulate loading again shows the same trend line of strength growth. The observed values were 49.12, 36.79 and 42.96 MPa SIP, JIP and SJIP respectively. Beside the 5% particulate addition also shows the hike in the flexural strength value, the obtained value was 53.82, 40.31 and 47.07 MPa for SIP, JIP and SJIP respectively. All through the investigation of the flexural analysis, it was observed that the load was evenly distributed between fibers and the IPN matrix when the specimen was subjected to gradual loading before fracture occurs. As well, crack propagates longitudinally along the IPN laminate cross section and simultaneously disturbs the inter-laminar shear strength between the fiber and matrix. However, the loading of 7% particulate loading into the IPN matrix slightly shows the decreasing scenario of flexural strength value, the obtained values were 47.14, 35.31 and 41.22 for SIP, JIP and SJIP respectively. The prime reason for absorbing the negative trend was that, the particulate could not evenly distribute into the matrix and thus the problem of agglomeration (clustering) happened in the laminate, hence it loses the stress transfer mechanism between the fiber and matrix. Due to this concern, the energy transfer was not properly initiated between the fiber and matrix instead it forms the localized zone of stress concentration along the line of fiber and initiates the premature crack following with the fiber breakage and pull-out, the same was again leads to the earlier failure due to the continuous prorogation of the cracks along direction of stress concentration zones. Flexural modulus also increased up to the level of 2.01, 1.62 and 1.82 GPa for SIP, JIP and SJIP respectively. As such interpretation observed in the tensile test analysis, in flexural modulus also the same kind of stiffening of the laminate happens due to this stress value increasing marginally, and directly proportional to the modulus. Again, it was clearly understood that, as much as addition of particulate matter into the matrix, it significantly induces the stiffness factor and thus increases the modulus with due respect to the loading of particulate matter[35,36].

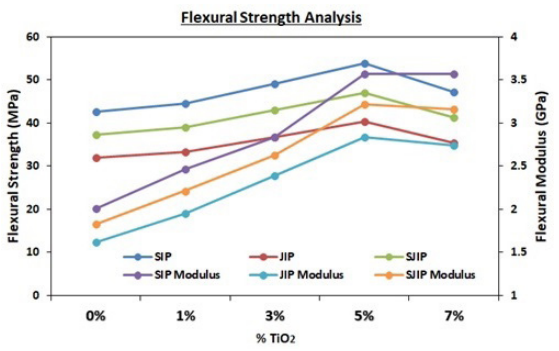

Figure 4. Flexural strength analysis of (a) sisal (b) jute (c) sisal/jute fiber reinforced (TiO_2) particulate loaded IPN composite.

3.3. Impact strength

The impact strength analyses of various proportions of particulate loading of TiO_2 were shown in the Figure 5. Upon loading the particulate matter into the IPN blend with the reinforcement, the impact strength parameters of the IPN laminate shows phenomenal growth like trend, up to the level of 5% of addition of particulate matter.

To prove this, the neat IPN laminate shows the impact strength value as 27.13, 24.12 and 25.625 kJ/m^2 for SIP,

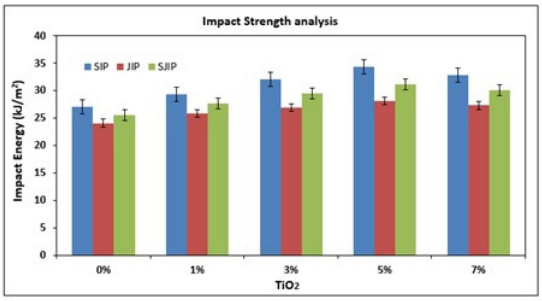

Figure 5. Impact strength analysis of (a) sisal (b) jute (c) sisal/jute fiber reinforced (TiO_2) particulate loaded IPN composite.

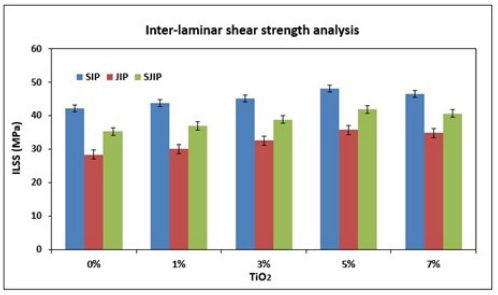

Figure 6. Inter-laminar shear strength analysis of (a) sisal (b) jute (c) sisal/jute fiber reinforced (TiO_2) particulate loaded IPN composite.

JIP and SJIP respectively. The addition of 1% of particulate into the matrix increases the impact strength value to the level of 29.42, 25.91 and 27.665 kJ/m^2 for SIP, JIP and SJIP respectively. The value obtained by the addition of even 1% of particulate into the matrix had significantly increased to 8.44, 7.43 and 7.89% for SIP, JIP and SJIP respectively. In this context, upon loading of particulate into the matrix massively increases the impact strength value with due respect to particulate addition. All sets of IPN laminates had shown the same kind of trend line throughout the entire study. In the same way, the 3% addition of particulates into the matrix showed the values of 32.12, 26.99 and 29.55 kJ/m^2 for SIP, JIP and SJIP respectively. Along the way, 5% particulate loading as well showed the values of 34.32, 28.13 and 31.225 kJ/m^2 for SIP, JIP and SJIP correspondingly. But it was interesting to note that, to the contrary, the 7% loading of particulates showed the decrease in impact strength value. The obtained value of 7% of particulate loading was 32.91, 27.32 and 30.115 kJ/m^2 SIP, JIP and SJIP respectively. Despite the TiO_2 loading these specimens had shown the negative trend of impact strength values. The understandable reason was that, the same problem of agglomeration into the matrix, the same kind of negative trend or fall of strength values were also absorbed in tensile and flexural strength upon loading of 7% of particulate. The particulates were not properly distributed throughout the entire matrix due to which, the energy absorption of the laminate had fallen to a marginal level. From the study, again it was clearly understood that the 5% of particulate loading proves as the optimistic level of incorporation into the matrix material beyond the limit the laminates show the negative level of energy absorption and stress transfer[37].

3.4. Short beam shear strength

The inter-laminar shear strength analyses of various proportions of particulate loading of TiO_2 were shown in the Figure 6. The main objective of this study was that to thoroughly analysis the composite resistance against the delamination damage.

During this entire course of study, the SIP specimens always held and exhibited higher levels of short beam shear strength as compared with other sets of laminate's. The neat SIP showed the value of 42.14 MPa, upon loading of various proportionate of particulate into the matrix subsequently increases their shear strength value up to the level of 5%, their corresponding obtained values were 43.92, 45.13, 48.11 and 46.52 MPa for 1%, 3%, 5% and 7% respectively. All the specimens showed the gradual propagation of cracks and delamination upon subjecting with vertical force, in the end, losses its stiffness and structural strength before failure occurs. As such value noted in all sets of previous analysis, the 7% particulate loading showed the early fracture as compared with 5% addition of TiO_2 addition. Likewise, the JIP also showed the positive trend up to the level of 5% and their subsequent 7% loading shows the negative trend of energy absorption, as a proof for this, the obtained values were 28.42, 30.13, 32.62, 35.74 and 34.91 MPa for 0%, 1%, 3%, 5% and 7% separately. Whereas the SJIP showed the mixture response of SIP and JIP, it shows that the sisal and jute fibers evenly absorbs the stresses and impact energy, depending on their individual strengths and contributes the same energy transfer when hybrid formation happens. Eventually, it was proved that the 5% of particulate loading demonstrates the higher level of stress transfer and 7% particulate addition shows the marginal level of negative trend due respect to their individual tests[38].

3.5. Water absorption

Water absorption behaviour of the sisal, Jute and sisal/jute (hybrid) fiber reinforced TiO_2 particulate loaded IPN laminate were thoroughly investigated with respect to various intervals. The corresponding graphs were plotted against water absorption versus square root of no of days as shown in the Figures 7, 8, 9.

During the initial days of study, from the Figure 7 it was seen that, the laminate absorbed the water absorption content very rapidly. Moreover, it was clearly seen from the graph that in between the days of √o to √4 days (during initial days of immersion) the IPN laminate started to vigorously absorb the water absorption content for all sets of particulate loaded IPN laminate irrespective of fiber reinforcement. However, it was realized that the 7% particulate loaded IPN laminate had absorbed nearly 0.8% to 0.85% of total water absorption of total weight, as compared with the remaining set of particulate loaded specimens during the early days.

To evident this, From the Figure 8, it was noticed that, for the same period of immersion time, the 1%, 3%, 5% and 7% particulate loaded specimens had shown the water absorption rate as 1.3%, 1.25%, 1.1% and 0.9% respectively, whereas the 7% particulate loaded IPN laminate conversely

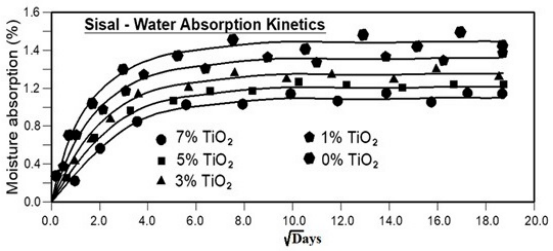

Figure 7. Water absorption absorption analysis of sisal fiber reinforced (TiO_2) particulate loaded IPN composite.

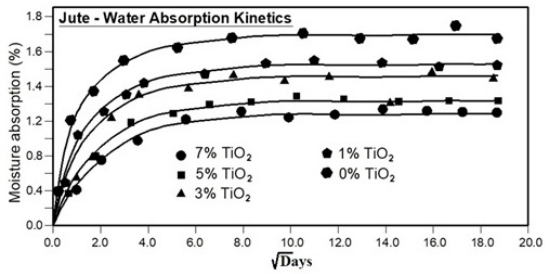

Figure 8. Water absorption analysis of jute fiber reinforced (TiO_2) particulate loaded IPN composite.

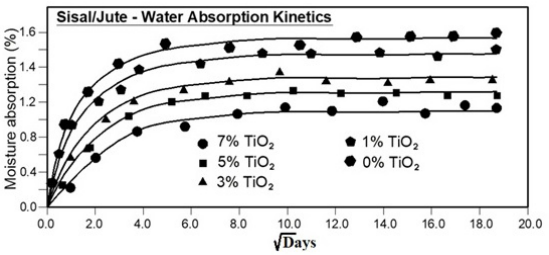

Figure 9. Water absorption analysis of sisal/jute fiber reinforced (TiO_2) particulate loaded IPN composite.

shown the absorption rate as 0.8% which was very less amongst all combinations of IPN laminate. Since the IPN blend was incorporated with the combinations of VER and PU, the PU often shows good water resistance as compared with other types of resins (same class). Though PU resin was hydrophobic in nature, when it was mixed with other sets of resin material their counterpart blend resins habitually showed smaller variation towards the hydrophobic nature, this was the reason that the absorption rate of IPN polymer network possibly changes the absorption rate. Along with that, as the IPN was blended with the PU, their soft segment presence generally repels the nonpolar molecules and maintains the hydrophobic nature upon the blend. Another finding from all three sets (a), (b) and (c) laminate was that the absorption rate was drastically slowed down after √4 days. In fact, the sisal, a jute and sisal/jute fiber absorbs the water absorption content very slowly once their initial rapid absorption period is over. Almost, after remaining √4 days of study, the absorption rate was conversely negligible and reaches to the level of almost saturation in √19 days.

From the Figure 9, it was seen that, as much as loading of particulate matter into the IPN blend, the laminate's level of water absorption significantly decreases. This was evident in all sets of IPN laminate; the 1% particulate into the IPN laminate decreases its value to the level of 1.21%, 1.31% and 1.26% for SIP, JIP and SIP/JIP respectively. Similarly, the 3% addition of particulate decreases the value of 1.29%, 1.45% and 1.37% for the corresponding SIP, JIP and SIP/JIP, this trend has kept advances in all sets of particulate inclusion into the matrix material. Eventually, the 7% predominantly takes the minimum water uptake to the level of 1.45%, 1.71% and 1.58% for the same. Actually, the particulate arrests the partial voids in between the fiber and matrix materials. Apart from that, most of the natural fibers had the nature of hydrophilic network, in that initially the water penetrates deep into the cellulose network of the fibers and through the action of capillary rise water absorption fills the space between the fibrils. As well, the rigidity of the cellulose molecule and structure was partially or completely plasticized because of the water absorption uptake in turn induces the cellulose molecules to freely move; due to this most of the cellulose system was substantially softened and changes the structure of the fiber easily. From the entire study, it was observed that as much as addition of the particulate reduces the hydroxyl group in turn reduces the water absorption uptake to the notable level and saturates after some quiet interval of time, irrespective of fiber reinforcement[39].

3.6. Fractrography

The sisal, jute and sisal/jute fiber reinforced impact fractured specimens were subjected to the scanning electron microscope investigations in order to understand the various micro and macro failure mechanisms that happened during the fractured test. During the Fractrographic analysis, it was clearly understood from the Figure 10 that 3% particulate loaded specimens had shown better fiber matrix adhesion also lessens the void presence on account of adding the particulate into the matrix material. Common issue of matrix cracking was observed on all sets of fractured specimens; this effect predominantly shows the strengthening mechanism of the matrix due to the addition of particulates into the IPN composite. Another breakage of fiber scissoring was commonly noticed in all jute fiber reinforced particulate loaded specimens. Furthermore, shear cups were also noticed upon the addition of particulate into the IPN matrix, this shear cups normally enrichens the strengthening factor between the fiber and matrix in significant way and also it was the proof for better interfacial adhesion between the fiber and matrix material[40].

The Figure 11 illustrates the fracture mechanism of the jute fiber reinforced with various amounts of particulate loaded fractured specimens. Common finding was that, irrespective of fiber reinforcement all sets of IPN laminates showed the fracture on account of fiber pull out. Actually, the particulate were better circulated over the void presence of the fiber as well on the circumference of the fiber and creates the substantial adhesion between the fiber and matrix, thus the way, it correspondingly increases the better strength

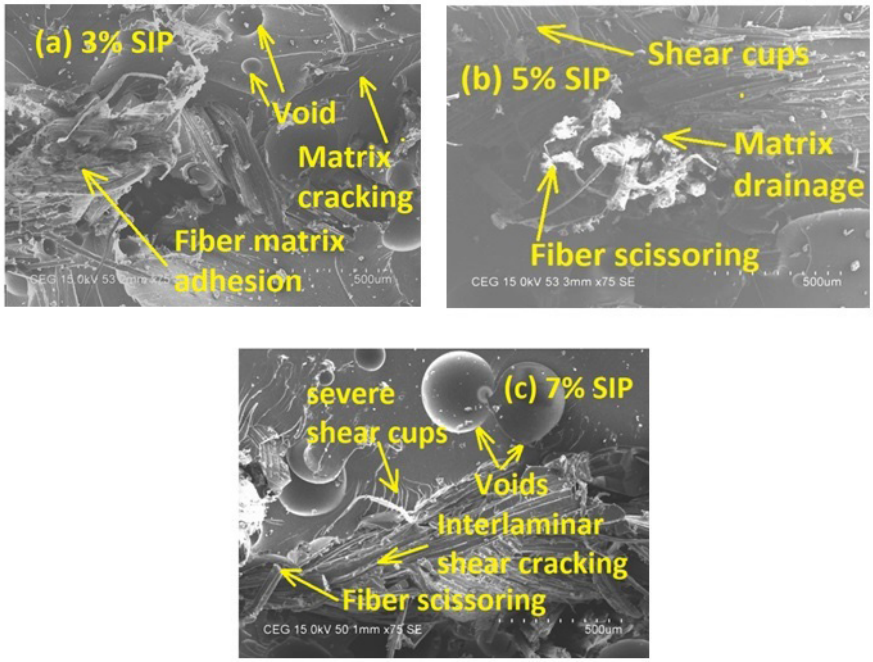

Figure 10. SEM analysis of sisal fiber reinforced (a) 3% TiO_2 (b) 5% TiO_2 (c) 7% TiO_2 (impact fractured) particulate IPN composite.

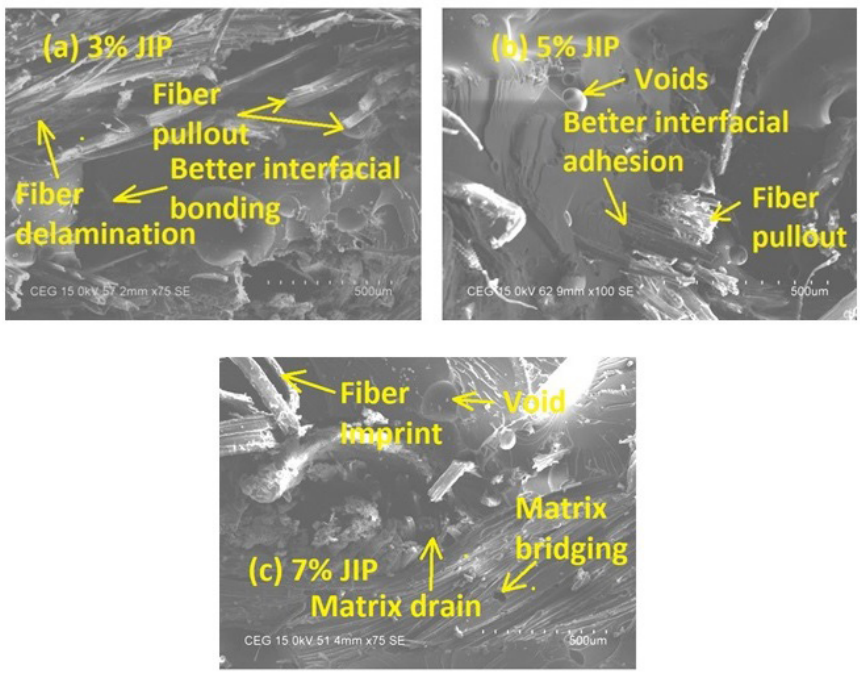

Figure 11. SEM analysis of jute fiber reinforced (a) 3% TiO_2 (b) 5% TiO_2 (c) 7% TiO_2 (impact fractured) particulate IPN composite**.**

on the IPN composites. As much as addition of particulate loading increases the better interfacial adhesion among the IPN composites. Additionally, fiber delamination was frequently noticed on all the specimens, initially matrices tended to absorb all the forces and transmit the same to the fibers. Once the threshold limit of the laminate exceeds, finally it leads to fiber delamination and fiber scissoring[41].

The Figure 12 elucidates the fracture mechanism of the sisal/jute fiber reinforced with various amounts of particulate loaded fractured specimens. Irrespective of fiber reinforcement all impact fractured specimens showed the same trend of matrix cracking, delamination, fiber pull out and scissoring, this was generally notified in all the specimens. Potholes were also notified in all specimens due to stress concentration on void presence areas.

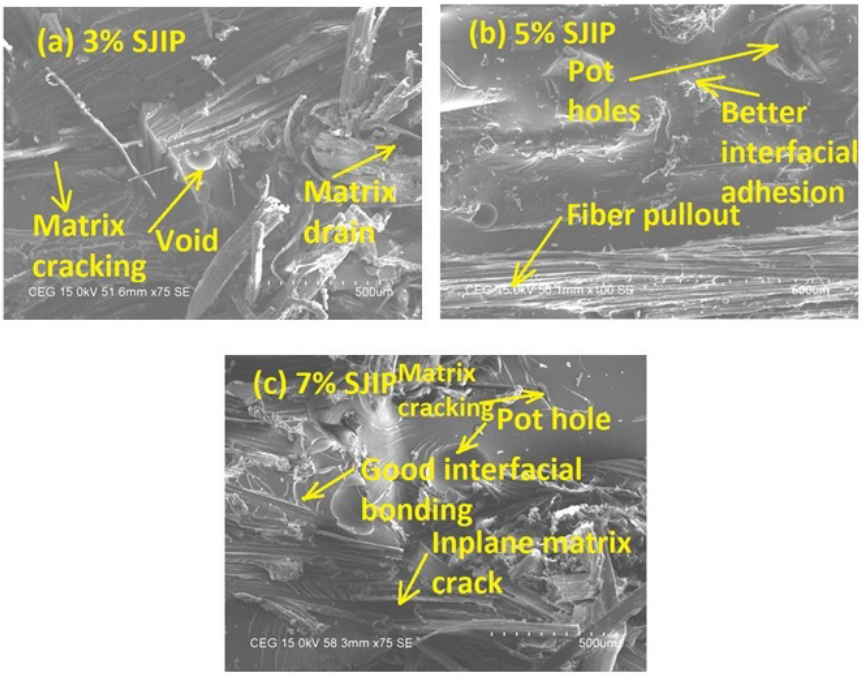

Figure 12. SEM analysis of sisal/jute fiber reinforced (a) 3% TiO_2 (b) 5% TiO_2 (c) 7% TiO_2 (impact fractured) particulate IPN composite.

The common findings from the fractrographics analysis were that, as much as addition of particulates into the matrix remarkably increases the better interfacial adhesion between the fiber and matrix, in turn strength of the composite increases specifically[42-44].

4. Conclusions

The influence of TiO_2 nano filler loading on water absorption, mechanical, and thermal characteristics of sisal, jute, sisal/jute IPN composite was the prime focus of this study. The conclusions regarding the same were written as follows.

1. The thermogravimetric analysis shows that, as much as addition of TiO_2 enhances the glass transition temperature of the composite in significant ways. As the evidence to showcase this, the 1% particulate loaded specimen shows the TGA value as 310°C and the value progressively increases (5% loaded) up to the level of 410°C upon loading of the particulate into the IPN matrix.
2. Also, mechanical properties were shown the better enhancement towards loading the particulate matters into the composite. Upon loading the fillers (TiO_2) into the composite, the tensile strength shows the growth level up to (variation from 0% to 7%) 20.3%, progressively by adding particulates. Correspondingly, flexural strength as well shows the hike in value by adding particulate into the IPN matrix, the increase in strength value was notified as (variation from 0% to 7%) 10.6%. Additionally, impact strength also proves better shock absorption characteristics by showing the increase in trend value as (variation from 0% to 7%) 50%. Finally the short beam shear strength analysis also showed the hike in strength value as 10.39%. On the contrary, in all the tests, the 5% particulate loaded specimens alone showed higher strength as compared with all other specimens. Whereas the 7% addition particulate shows the negative trend in all the physical tests, this was mainly due to the problem of agglomeration effect in the blends. Particulates could not evenly distribute into the blend beyond the limit of 5%.
3. A water absorption characteristic shows the positive trend in value, as much as addition of particulate matter into the matrix, it keeps decreases the water absorption value in agreed level. All the specimens irrespective of fiber reinforcement show the same hydrophobic nature upon particulate loading into the matrix. The variation was observed as 0% - 1.01, 7% - 1.45 towards the filler material incorporation into the IPNs.
4. Nevertheless, fractrography images revealed that, as much as addition of particulates into the matrix, increases the better interfacial adhesion between the fiber and matrix, micro crack bridging through particulates. However, the common mode of failure mechanism like fiber pull-out, interface debonding, fiber breakage and matrix cracking was observed.

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