Mechanical properties of natural fibre reinforced polymer composites

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Abstract. During the last few years, natural fibres have received much more attention than ever before from the research community all over the world. These natural fibres offer a number of advantages over traditional synthetic fibres. In the present communication, a study on the synthesis and mechanical properties of new series of green composites involving *Hibiscus sabdariffa* fibre as a reinforcing material in urea–formaldehyde (UF) resin based polymer matrix has been reported. Static mechanical properties of randomly oriented intimately mixed *Hibiscus sabdariffa* fibre reinforced polymer composites such as tensile, compressive and wear properties were investigated as a function of fibre loading. Initially urea–formaldehyde resin prepared was subjected to evaluation of its optimum mechanical properties. Then reinforcing of the resin with *Hibiscus sabdariffa* fibre was accomplished in three different forms: particle size, short fibre and long fibre by employing optimized resin. Present work reveals that mechanical properties such as tensile strength, compressive strength and wear resistance etc of the urea–formaldehyde resin increases to considerable extent when reinforced with the fibre. Thermal (TGA/DTA/DTG) and morphological studies (SEM) of the resin and biocomposites have also been carried out.

Keywords. Composites; reinforcements; optimization; mechanical properties; thermal study.

1. Introduction

Increased environmental awareness and consciousness throughout the world has developed an increasing interest in natural fibres and its applications in various fields. Natural fibres are now considered as serious alternative to synthetic fibres for use in various fields (Chauhan *et al* 2000a; Singha *et al* 2006, 2008; Kaith and Kalia Susheel 2007; Singha and Thakur 2008a). The use of natural fibres as reinforcing materials in both thermoplastic and thermoset matrix composites provides positive environmental benefits with respect to ultimate disposability and best utilization of raw materials (Singha *et al* 2004; Kaith *et al* 2007). Currently, studies on use of lignocelluloses bio fibres in place of synthetic fibres as reinforcing materials are being pursued vigorously (Singha *et al* 2002; Panthapulakkal *et al* 2006). These bio fibres are being extensively used for the production of cost effective ecofriendly biocomposites (Sain and Kokta 1994; Misra *et al* 1996; Gassan *et al* 1997; Hornsby *et al* 1997).

The advantages of natural fibres over traditional reinforcing materials such as glass fibre, carbon fibre etc are their specific strength properties, easy availability, light weight, ease of separation, enhanced energy recovery, high toughness, non-corrosive nature, low density, low cost, good thermal properties, reduced tool wear, reduced dermal and respiratory irritation, less abrasion to processing equipment, renewability and biodegradability (Lawton and Fanta 1994; Simon *et al* 1998; Chauhan *et al* 1999, 2001; Joshi *et al* 2004; Singha *et al* 2004). It has been observed that natural fibre reinforced composites have properties similar to traditional synthetic fibre reinforced composites. Natural fibre composites have been studied and reviewed by a number of researchers (Dufresne 1997; Dufresne and Vignon 1998; Mao *et al* 2000; Kaith *et al* 2003; Nakagaito *et al* 2004, 2005; Bhatnagar and Sain 2005). During the past decade, a number of significant industries such as the automotive, construction or packaging industries have shown massive interest in the progress of new biocomposites materials. One of the most appropriate examples of this is the substitution of inorganic fibres such as glass or aramid fibres by natural fibres (Bledzki and Gassan 1999; Chauhan *et al* 1999; Chakraborty *et al* 2006). All these properties have made natural fibres very attractive for various industries currently engaged in searching for new and alternate products to synthetic fibre reinforced composites.

The properties of natural fibres can vary depending on the source, age and separating techniques of the fibres. *Hibiscus sabdariffa*, an annual fibre plant, has been found to be an important source of fibres for a number of applications since good old days. The bast fibre has high potential as a reinforcing fibre in polymer composites. *Hibiscus sabdariffa* plant fibre is abundantly found in the Himalayan region, especially in Himachal Pradesh. Traditionally,

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this fibrous material belonging to Himalayan region is being used by the local people for making low cost articles like socks, boots, mats, ropes, bags etc. The literature review has shown scanty information on the application of this fibre as reinforcing material in the polymer composites. Our research group has successfully improved the properties of a number of fibres such as flax, *Saccharum cilliare*, *Hibiscus sabdariffa*, rayon and *Cannabis indica* by various techniques such as graft-copolymerization etc (Chauhan *et al* 1999, 2000a, b, 2001; Singha *et al* 2002, 2004, 2006; Kaith *et al* 2003, 2007a; Kaith and Kalia Susheel 2007). Keeping in view the easy availability of this new fibre a comprehensive research work has been initiated in our laboratory on synthesis and study of properties of *Hibiscus sabdariffa* fibre reinforced urea–formaldehyde (U–F) resin matrix based biocomposites.

2. Experimental

2.1 *Material and methods*

Urea (Qualigens Chemicals Ltd), formaldehyde solution (Qualigens Chemicals Ltd.) and sodium hydroxide (Qualigens Chemicals Ltd.) were used as received.

2.1a *Matrix polymer*: Amino resin such as urea–formaldehyde (U–F) was used as matrix polymer.

2.1b *Reinforcing material*: The ligno-cellulosic material used as the reinforcing material in the biocomposite was *Hibiscus sabdariffa* fibre of different dimensions.

2.2 *Instruments used*

Weights of the samples were taken on Shimadzu make electronic balance (LIBROR AEG-220), curing of samples was done on compression molding machine (SANTECH INDIA Ltd), thermal studies were carried out using Thermal Analyzer (LINSEIS, L 81-11) and SEM micrographs were taken on LEO 435VP.

2.3 *Synthesis of urea–formaldehyde resin*

Urea and formaldehyde were taken in different molar ratios (1·0 : 1·0, 1·0 : 1·5, 1·0 : 2·0, 1·0 : 2·5 and 1·0 : 3·0) by weight, in the reaction kettle and were mixed with the help of a mechanical stirrer. NaOH solution was added slowly with constant stirring and heating to the urea–formaldehyde solution till pH of 8 was attained. Since the reaction was exothermic, proper care was taken to maintain the temperature between 50 and 60°C, for initial 2 h. Then the temperature was increased to 80–85°C at pH 5·5–6 and the mixture was heated at this temperature, till the resinification started. The reaction mixture was con-densed at this temperature for 3–4 h till the complete resinification. Then heating was stopped and the resin was cooled and samples were then cured at 130°C in compression molding machine. The cured samples were then subjected to various mechanical and thermal studies.

2.4 *Fabrication of biocomposites*

2.4a *Particle reinforced composites*: *Hibiscus sabdariffa* fibres were grinded to a powder and filtered through a sieve of pore size, 200 microns. The specific amount of resin and fibres was taken and mixed thoroughly by suitable loading (1·0 : 0·01). Curing of samples was done in compression molding machine at 130°C and were subjected for different mechanical and thermal studies.

2.4b *Short-fibre reinforced composites*: The fibres were chopped into 3 mm size and mixed with weighed amount of UF resin in a dish. Curing of samples was carried out at 130°C in compression molding machine.

2.4c *Long fibre reinforced composites*: In this case, fibres were chopped into 6 mm length and mixed with weighed amount of resin. Curing of samples was done as per the method discussed earlier.

2.5 *Mechanical testing*

Testing of samples for tensile and compressive strengths were done on Computerized Universal Testing Machine (HOUNSFIELD H25KS) and wear testing was done on Wear & Friction Monitor (DUCOM-TR-20L).

2.5a *Tensile strength test*: The tensile strength test was conducted on Computerized Universal Testing Machine (HOUNSFIELD H25KS). The sample of 10 cm length was clamped into the two jaws of the machine. Each end of the jaws covered 2 cm of the sample. Tensile strength was studied over the rest of 6 cm gauze length. Reading of the tensile strength test instrument for Newton force and extension was initially set at zero. The test was conducted at the constant strain rate of the order of 10 mm/min. Tensile stress was applied till the failure of the sample and load-extension curve was obtained. Each sample was tested for five times and average results have been reported.

2.5b *Compressive strength test*: Compression strength of samples was also tested on Computerized Universal Testing Machine (HOUNSFIELD H25KS). Composite sample was held between the two platforms and the strain rate was fixed at 10 mm/min whereas the total compression range was 7·5 mm. The compression stress was applied till the failure of sample. Total compression per unit force was noted.

2.5c *Wear test*: The wear test of the testing sample was conducted by Wear & Friction Monitor (DUCOM-TR-20L). The disc was cleaned with emery paper and it was fixed at 500 rpm. The inner diameter of steel disc was 80 mm. Initial weight of the sample was noted and the sample pin was fixed in the jaws of wear testing machine. Then machine was set to display zero wear and friction. The samples were tested with different loads varying from 1–3 kg. For each load the machine was allowed to run for 15 min and the readings were recorded. After 15 min the sample was taken out from the machine and weighed again. Then loss in weight due to abrasion was calculated and this weight loss was used as the measure of wear.

2.6 *SEM analysis of samples*

Scanning electron micrographs (SEM) of resin sample and its respective composites were taken on Leo 435 VP. These micrographs clearly show the difference in the morphology of UF resin and respective fibre reinforced polymer composites.

2.7 *Thermal analysis of samples*

Thermal analysis of natural and synthetic polymers gives us good account of their thermal stability. Thermal analysis comprises of various methods such as thermogravimetric analysis (TGA)/differential thermal analysis (DTA), derivative thermogravimetry (DTG) etc. Thermogravimetric analysis (TGA) and differential thermal analysis (DTA) studies of samples were carried out in nitrogen atmosphere on a thermal analyser (Perkin Elmer) at a heating rate of 10°C/min.

TGA was used to characterize the decomposition and thermal stability of materials under a variety of conditions, and to examine the kinetics of the physico-chemical processes occurring in the sample. Basically in this method a change in thermal stability was examined in terms of percentage weight loss as a function of temperature. The mass change characteristics of a material were strongly dependent on the experimental conditions such as sample mass, volume, physical form; shape and nature of the sample holder, nature and pressure of the atmosphere in the sample chamber and the scanning rate, all have important influences on the characteristics of the recorded TG curve. At the same time, DTA involves comparing the precise temperature difference between a sample and an inert reference material, while heating both.

DTG is a type of thermal analysis in which rate of material weight changes upon heating vs temperature is plotted and is used to simplify reading of weight versus temperature thermogram peaks that occur close together. DTG peaks are characterized by the peak maximum (T_{max}) and the peak on set temperature (T_e). The area under DTG curve is proportional to the mass change and the height of the peak at any temperature gives the rate of the mass change at that temperature. DTG curves are frequently preferred when comparing results with DTA curves because of the visual similarity.

3. Results and discussion

The importance of static mechanical analysis (SMA) as a tool in the study of the behaviour of polymer biocomposites is of paramount importance. It has been proved to be an effective method to study the behaviour of materials under various conditions of tension, compression, stress–strain, and phase composition of fibre composites and its role in determining the mechanical properties. Static mechanical properties of fibre reinforced composites depend on the nature of the polymer matrix, distribution and orientation of the reinforcing fibres, the nature of the fibre–matrix interfaces and of the interphase region.

Tensile strength: The ability of a material to resist breaking under tensile stress is one of the most important and widely measured properties of materials used in structural applications. The force per unit area (MPa or psi) required to break a material in such a manner is the ultimate tensile strength or tensile strength at break.

Compressive strength: The ability of a material to resist breaking under compression stress is also one of the most important and widely measured properties of materials used in various applications.

The value of uniaxial compressive stress reached when the material fails completely is designated as the compressive strength of that material. The compressive strength is usually obtained experimentally by means of a compressive test. The apparatus used for this experiment is the same as that used in a tensile test.

3.1 *Mechanism of synthesis of urea–formaldehyde resin*

Mechanism of polymerization reaction follows two steps. First step is the formation of methylol urea from the reaction between urea and formaldehyde. Since urea is tetra functional so initial reaction due to presence of sufficient ratio of formaldehyde to urea may lead to the formation of tetra methylol derivative of urea (as shown in figure 1A). The rate at which methylol urea (I) and (II) formation takes place depends upon temperature, pH and the ratio of formaldehyde to urea (F/U). To control the reaction and to favour the formation of dimethylol urea, the reaction condition must be slightly alkaline and a pH of 7–8 is employed (Singha and Thakur 2007). Since methylol ureas are not adhesives, condensation does not take place. So next step is the condensation of monomeric methylol urea to form polymer molecules. The reaction is carried
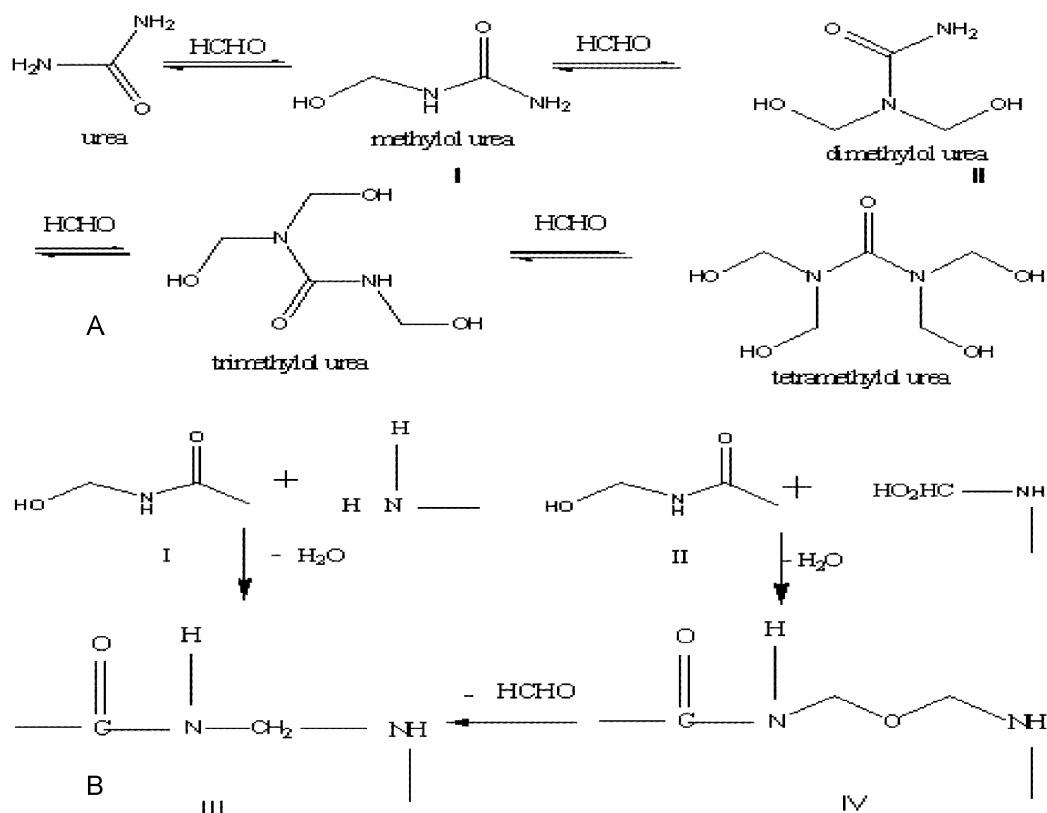
Figure 1. **A**. Formation of methylol derivatives of urea and **B**. condensation of methylol urea by elimination.

out at 80–90°C in an acidic medium (pH 5·5–6). As the reaction proceeds larger molecules with cross linked structures are formed. After the completion of the reaction, neutralization (pH 7·5–8) is carried out. The condensation must be closely watched and controlled at the stages of production because if the reaction is allowed to continue cross linking will lead to the gelatization of the resin. Under acidic conditions methylol urea condense by elimination of water between either of four steps (as shown in figure 1B).

3.2 Optimization of urea–formaldehyde resin

3.2a *Tensile strength*: It has been observed that U–F samples of ratio 1 : 2·5 bear maximum load at a particular applied load as compared to samples of other ratios. This ratio (1 : 2·5) could bear a load of 128·125 NF with an extension of 1·84 mm (figure 2A). On the other hand, samples of other ratios bear low loads.

3.2b *Compressive strength*: It is evident from figure 2B that the samples of ratio 1 : 2·5 could bear a load of 991 N at a compression of 3·51 mm.

3.2c *Wear resistance*: It has been observed that wear rate of samples of ratio 1 : 2·5 was less as compared to

any other samples. Loss of material was due to abrasion and friction of samples with disc (figure 2C).

As evident from figures 2A–C samples of ratio 1 : 2·5 show maximum tensile and compressive strengths. Moreover, at this ratio wear rate was also very less. Therefore, this ratio was taken for further preparation of urea–formaldehyde resin and respective biocomposites.

3.3 Effect of reinforcement on mechanical properties of U–F based biocomposites

3.3a *Tensile strength*: It has been observed that composites with particle reinforcement showed more tensile strength which was followed by short fibre and long fibre reinforced composites (figures 3A–C). It is clear from figures 3A–C that the samples of (i) particle reinforced composite could bear a load of 332·8 N at an extension of 2·2 mm, (ii) short fibre reinforced composite could bear a load of 307·6 N at an extension of 2·23 mm and (iii) long fibre reinforced composite could bear a load of 286·1 N at an extension of 2·28 mm.

3.3b *Compressive strength*: Compressive strength of UF resin matrix has been found to increase when reinforced with fibre. It has been found that with particle re-
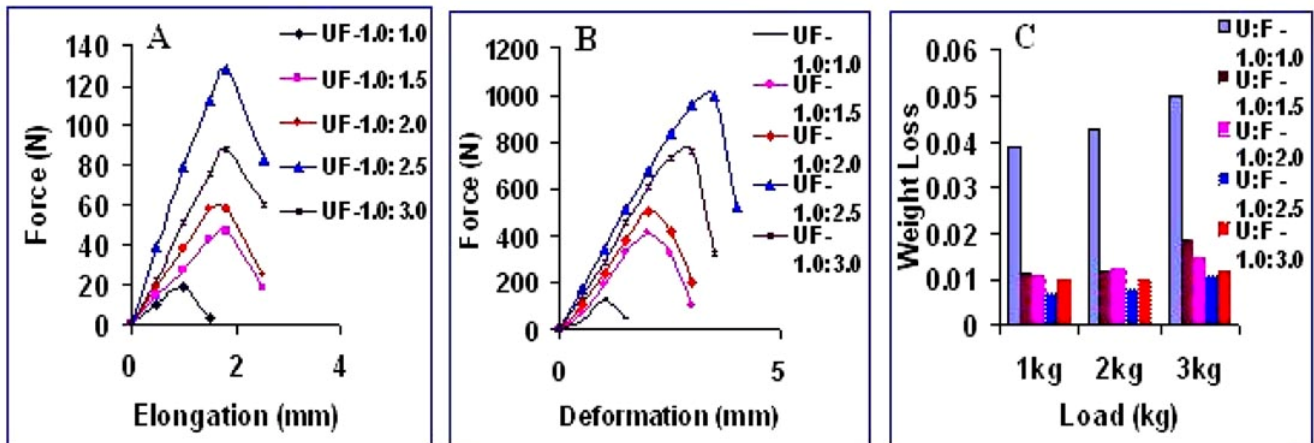
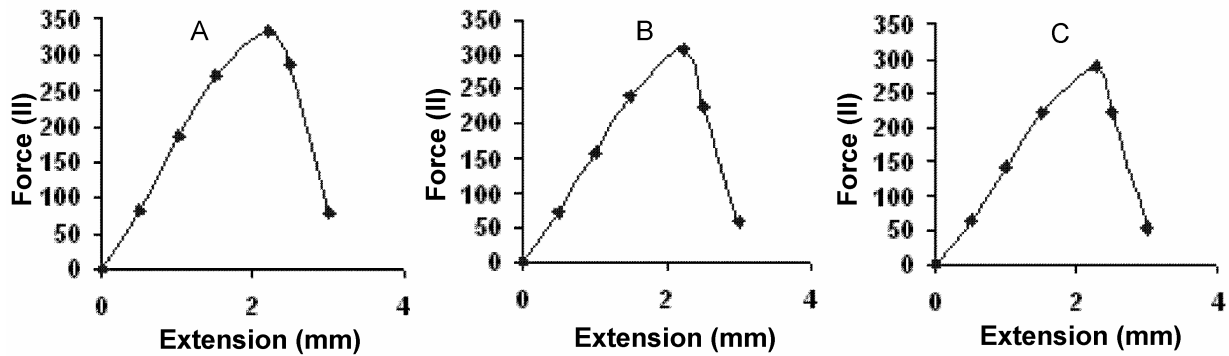**Figure 2.** Tensile/compression strength and wear resistance curve of UF resin (**A**, **B**, **C**).

Figure 3. Tensile strength curves of P–Rnf/SF–Rnf and LF–Rnf composites (**A**, **B**, **C**).
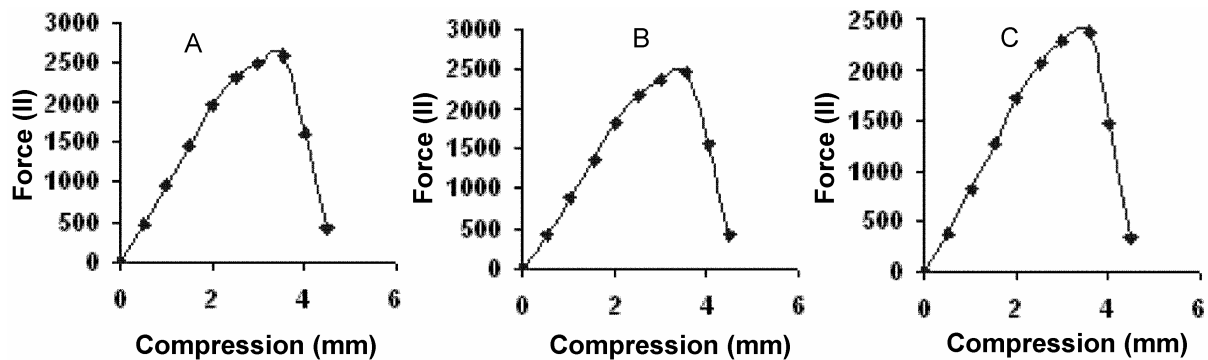

Figure 4. Compressive strength curves of P–Rnf/SF–Rnf and LF–Rnf composites (**A**, **B**, **C**).

inforcement, compressive strength increases to a much more extent than short and long fibre reinforcement (figures 4A–C). It is clear that the samples of (i) particle reinforced composite could bear a load of 2586·5 N with a compression of 3·51 mm, (ii) short fibre reinforced composite could bear a load of 2466·5 N with a compression of 3·55 mm and (iii) long fibre reinforced composite could bear a load of 2376·5 N with a compression of 3·58 mm.

3.3c *Wear test*: As evident from figures 5A–C wear rate of UF matrix decreases appreciably when it is reinforced with *Hibiscus sabdariffa* fibre. It was observed that particle reinforcement decreases the wear rate to a much more extent than short and long fibre reinforcements.

From these results it is clear that particle reinforcement is more effective than short and long fibre reinforcements. This may be due to larger surface area and more fibre/matrix interaction in case of particle reinforced composites.
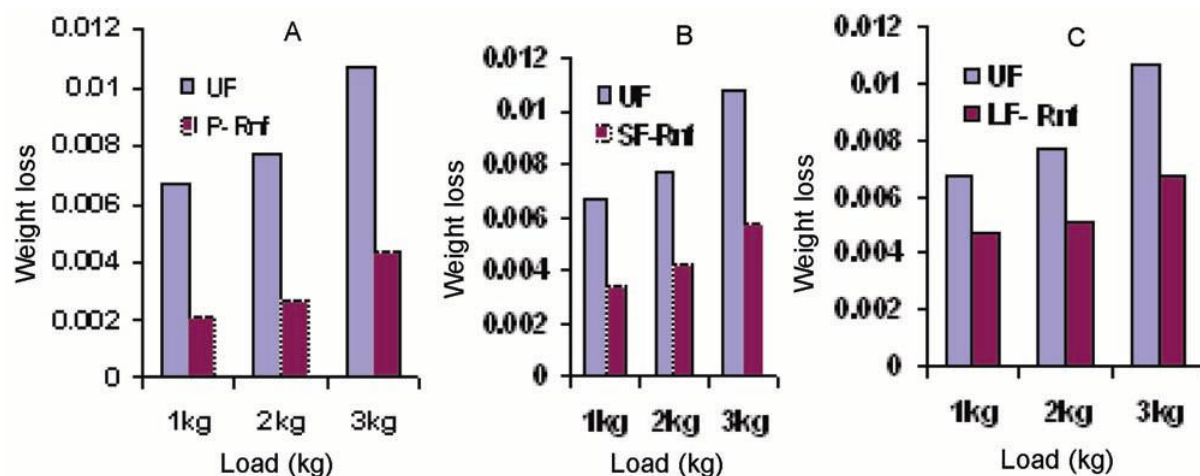
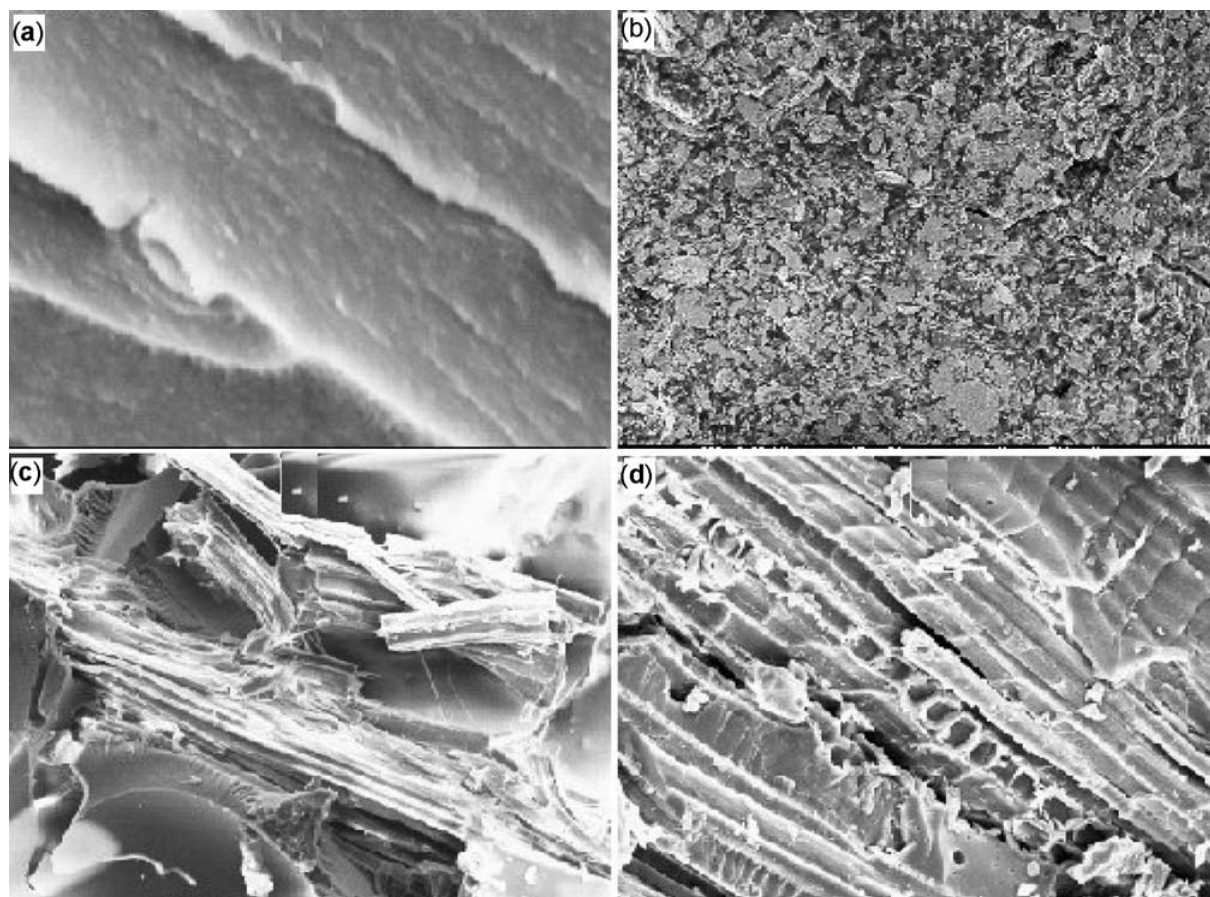**Figure 5.** Wear resistance curves of P–Rnf/SF–Rnf and LF–Rnf composites (**A**, **B**, **C**).

Figure 6. SEM images of UF resin, P–Rnf, SF–Rnf and LF–Rnf composites (**A**, **B**, **C** and **D**).

3.4 *Morphological study of biocomposites*

Morphological results (figures 6A–D) clearly show that there is proper intimate mixing of *Hibiscus sabdariffa* fibre with the synthesized resin in the biocomposites. It is clear from the micrographs that there is uniform mixing with particle reinforcement as compared to short and long fibre reinforcements.

3.5 *Thermal behaviour of U–F resin and its composites*

Thermogravimetric analysis (TGA) of raw fibre, polymeric resin and biocomposites was studied as a function of % weight loss with the increase in temperature. In case of raw fibre (figure 7), in the beginning depolymerization, dehydration and glucosan formation took place between the temperature ranges 26°C and 190°C followed by the
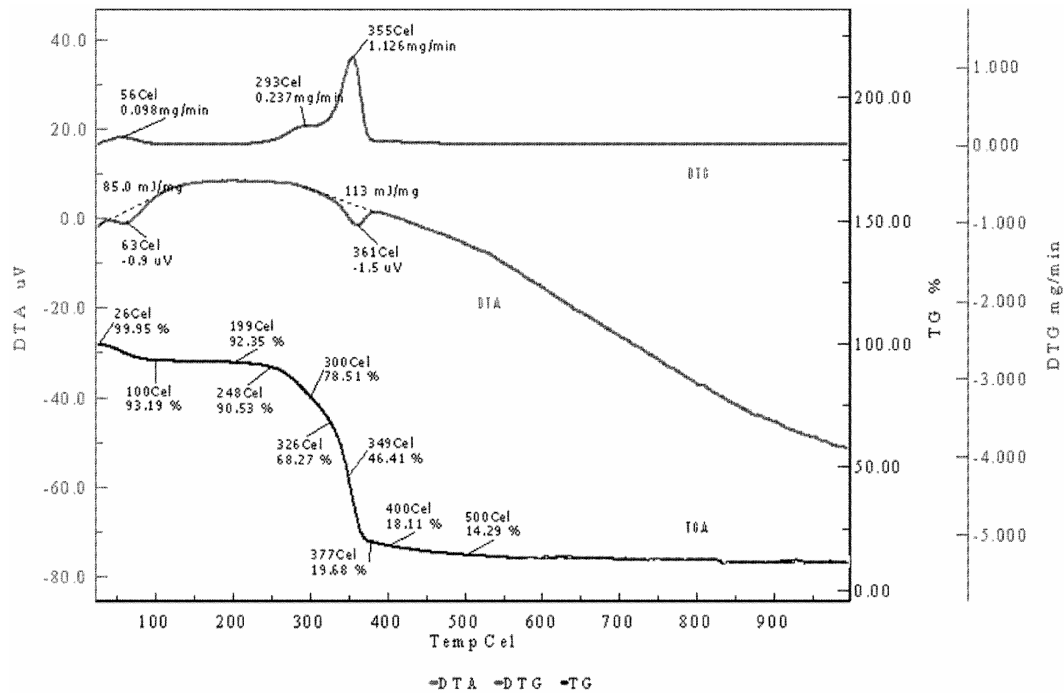
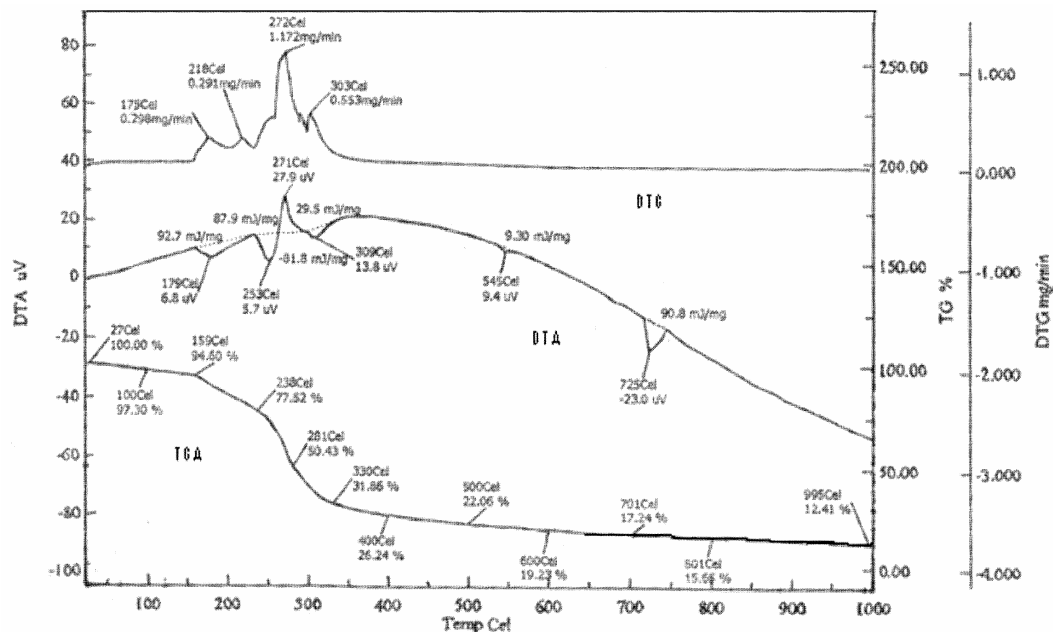**Figure 7.** TGA/DTA and DTG analysis of *Hibiscus sabdariffa* fibre.

Figure 8. TGA/DTA and DTG analysis of urea–formaldehyde resin.

cleavage of C–H, C–C and C–O bonds. Initial decomposition temperature (IDT) is 199°C and final decomposition temperature is 500°C. On the other hand, in case of urea–formaldehyde resin (figure 8), it is single stage decomposition and the initial decomposition temperature is 238°C, the final decomposition of the resin took place at 995°C. The degradation temperatures for natural fibre reinforced

composites fall between the degradation temperatures for the matrix and the fibres. It has been observed that for biocomposites initial decomposition temperature is 230°C and the final decomposition of the composite took place at 800°C which indicate that the presence of cellulose fibres does affect the degradation process. The data on initial decomposition (IDT) temperature and final de-
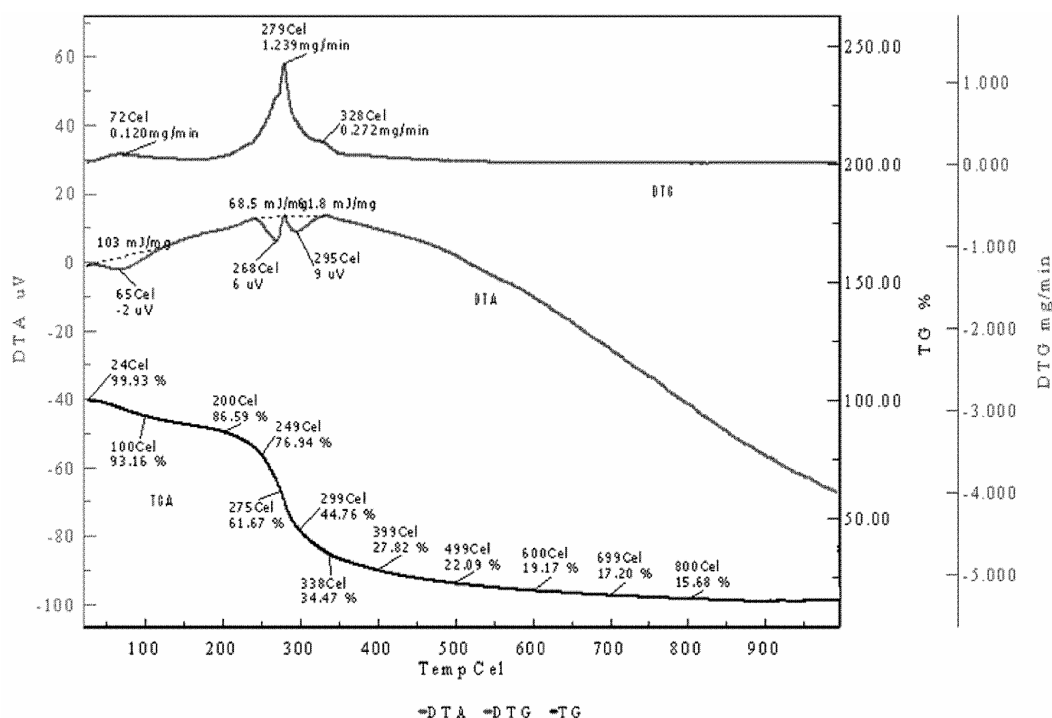
Figure 9. TGA/DTA and DTG analysis of *Hibiscus sabdariffa* fibre reinforced UF matrix based polymer composite.

composition temperature (FDT) of fibre, resin and bio-composite is shown in figures 7–9. This behaviour is consistent with the results reported earlier (Singha and Thakur 2007, 2008b). These studies are further supported by differential thermal analysis (DTA) and derivative thermogravimetry (DTG) (figures 7–9).

4. Conclusions

Various test methods were adapted for mechanical characterization of natural fibre reinforced polymer composites. In case of mechanical behaviour particle reinforcement of the UF resin has been found to be more effective as compared to short fibre reinforcement. These results suggest that *Hibiscus sabdariffa* fibre has immense scope in the fabrication of natural fibre reinforced polymer composites having vast number of industrial applications.

Acknowledgements

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