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Comparative performance assessment of pineapple and Kevlar fibers based friction composites



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ABSTRACT

Novel friction composites materials using pineapple fiber as a sustainable alternative for automotive industry were developed by increasing its amount from 5–20 wt.% in the step of 5 %. To compare the performance of pineapple fiber, friction composites with 5–10 wt.% of Kevlar fiber were also manufactured. The results of physico-mechanical properties reveal that density, hardness and ash content decrease whereas water absorption, porosity and compressibility increase with the increased pineapple/Kevlar fiber contents. Further, the friction and fade performance were found to decrease whereas the recovery performance and wear was found to increase with increased pineapple fiber content. Among pineapple fiber reinforced composites, the best composite is the one having 5 wt.% pineapple fibers that exhibits the highest performance in terms of coefficient of friction (0.548), lowest fade-% (36.31 %) along with the lowest specific wear rate ($3.49 \times 10^{-8} \text{ cm}^3/\text{N-m}$). Nonetheless, the results show that the 5 wt.% Kevlar fiber based composite reveals good performance in terms of coefficient of friction (0.592) with slightly lower fade-% (35.98 %), recovery-% (107.43 %) and specific wear rate ($3.46 \times 10^{-8} \text{ cm}^3/\text{N-m}$) when comparing to 5 wt.% pineapple fiber based composites. Finally, the possible wear mechanisms were discussed with the help of composites worn surface morphologies.

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1. Introduction

Today, the novel innovative brake friction materials made of natural and waste resources are able to replace the classical materials which are difficult to machine and help to protect

the environment [1,2]. A brake friction material is designed to meet a wide range of performance requirements, including high and stable coefficient of friction, high recovery, low fade, wear, noise and vibration over a varying range of working environments [3]. The brake friction materials frequently contain in excess of fifteen ingredients, which are categorized into five prime classes of abrasives, binder, filler, fibers and lubricants [4]. Among them, the fibers play a central role because they can control the physical, mechanical and tribological properties of brake friction materials [5]. The development of brake

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Table 1 – Details of composites designation and composition.

Composition (wt.%)	Composite designation					
	PF-1	PF-2	PF-3	PF-4	KF-1	KF-2
Parent formulation ^a	45	45	45	45	45	45
Pineapple fiber	5	10	15	20	0	0
Barium sulphate	50	45	40	35	50	45
Kevlar fiber	0	0	0	0	5	10

^a Parent formulation = Property modifiers (15 wt.%; graphite: alumina: vermiculite = 1:1:1), phenol formaldehyde resin (10 wt.%), lapinus fiber (20 wt.%).

Table 2 – Processing conditions adopted for composite fabrication.

Procedure	Conditions
Mixing	Sequence: (i) Fibrous ingredients with phenolic resin for 5 min (ii) Powdered ingredients for next 5 min.
Curing	Temperature = 155 °C, Pressure = 15 MPa, Time = 10 min with 5 breathings to expel volatiles.
Post-curing	Temperature = 170 °C, Time = 3 h.
Specimen size	Specimens of 25 mm × 25 mm × 6 mm sizes were used in tribological assessment.

friction materials were started with the marked utilization of asbestos fiber [6]. But the prolonged exposure to asbestos fiber proved carcinogenic and it was reported that nearly 0.2 million people die due to asbestos related diseases, including mesothelioma, lung cancer [7,8]. Various fibrous materials such as steel, aluminium, lapinus, wollastonite, Kevlar, carbon etc. have been studied as a substitute of asbestos fiber [9–11]. Among them, Kevlar fiber attracts noticeable attention and reported to enhance various properties of friction materials [12,13]. Apart from the benefits, Kevlar fiber reported to exhibit some drawbacks like non-recyclability, higher cost and energy consumption [14]. The world health organization reported the hazards of fourteen types of asbestos substitute materials including Kevlar fiber [15]. Moreover, Kevlar fibers were also reported to possess carcinogenic nature by various researchers [16–18]. Nonetheless, the natural fibers are very attractive solutions cause of lighter weight, renewability, biodegradability, low or zero cost, high specific modulus, availability, non-abrasive and non-toxic nature and their continuum demand for various applications [19,20].

Over the past few years, numerous research groups dealt with natural fiber based friction composite materials. They suggested that the numerous advantages of natural fibers will provide a cheaper and ecofriendly alternative to expensive fiber such as Kevlar used in the brake friction material industries. M.A. Maleque and A. Atiqah [21] concluded that the addition of 5 vol.% coir fibers into friction formulations resulted into highest wear resistance. Z. Fu et al. [22] studied the tribological properties of flax fiber reinforced friction composites. They revealed that the incorporation of 5.6 vol.% flax fibers stabilize the friction coefficient and help to improve the wear resistance at elevated temperature. Y. Liu et al. [23] studied the influence of abaca fiber of friction and wear performance on phenolic resin-based composites. They concluded that not only the amount but also the length of fibers can play a significant role to enhance tribological properties. In addition, sisal fiber [24], hemp [25], bamboo fiber [26], kenaf fiber [27] and more recently cow dung fibers [28] have been reported to improve the tribological properties.

There, the use of natural fibers allows replacing the asbestos and other carcinogenic material. One of most abundant raw materials is the pineapple fiber produced from the pineapple fruit plant (*Ananas Comosus*) leaves that is extensively used in textile industries [29]. Although, the pineapple fibers reinforced composites were reported to demonstrate good physical and mechanical properties [30], to the authors knowledge no information is available on the use of these fibers in the brake friction materials design. Therefore, here, we propose a novel brake friction material as an economic and ecologic alternative for synthetic fiber such as Kevlar used in automotive industry. The natural pineapple fibers were used to design the novel composite as reinforcement in phenol formaldehyde resin. The performance of pineapple fiber based friction composites were also compared with Kevlar fiber reinforced composites. The results from the experiments prove suitable performances in terms of friction and wear performance that make the novel composite as a potential candidate for brake components.

2. Experimental procedure

2.1. Materials and fabrication details

The friction composites are made of parent formulation (45 wt.%). It contains phenol formaldehyde resin, lapinus fiber, graphite, alumina, and vermiculite. The remaining 55 wt.% was adjusted by varying barium sulphate, pineapple fiber and Kevlar fiber as is stated in Table 1.

The Kevlar fiber (fiber length = 0.5–1 mm) was procured from DuPont India, while the pineapple fiber was procured from Chandra Prakash & Co., Jaipur, India. The procured pineapple fibers were treated with 5 wt.% of sodium hydroxide solution for 24 h. After washing them with distilled water, the fibers were oven dried for 5 h at 60 °C. Then, the fibers were cut to a length of 2–6 mm and used for the composite fabrication.

During composite manufacturing, the ingredients were sequentially mixed in a mechanical mixer and thereafter heat treated in a compression molding machine as per the details

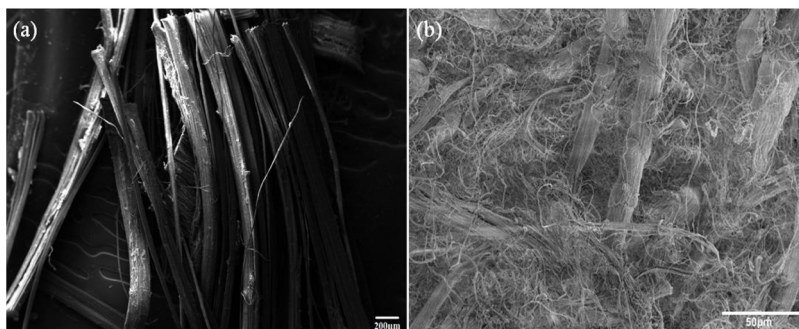


Fig. 1 – SEM images of (a) pineapple fiber and (b) Kevlar fiber.

presented in Table 2. The image of pineapple and Kevlar fibers obtained by using the scanning electron microscope are shown in Fig. 1.

2.2. Physical, mechanical and chemical properties

The density of manufactured composites was determined, at room temperature, by applying Archimedes method using a density determination kit (Wensar Weighing Scales Ltd., India). The porosity was determined by soaking the composite sample in preheated SAE 90-grade oil for 8 h as per JIS D 4418 standard [31]. In the developed composites, the amount of uncured resin was determined by acetone extraction using ASTM D 494 standard [32]. The ash content of the composites was found out by roasting the sample weight of 2–4 g in a muffle furnace ($850 \pm 25^\circ\text{C}$ temperature) for two hours. Water absorption was found by immersing the composite sample ($25\text{ mm} \times 25\text{ mm} \times 5\text{ mm}$) in distilled water at room temperature for 24 h as per ASTM D 570-98 standard [33]. The water absorption was calculated by normalizing the difference between the initial and final weight to initial weight. The compressibility performances were carried out accordingly to ISO 6310 standard [34] by using a compressibility testing machine (Hind hydraulics, India). The hardness was measured on a digital hardness tester (Model: TRSN-BD; Fine Manufacturing Industries, India) on Rockwell-R scale using a steel ball indenter (12.7 mm diameter) with minor and major loads of 10 kg and 60 kg, respectively.

2.3. Tribological characterization

The tribological tests were performed under a Chase friction tester in accordance with IS 2742 standard [35]. The main cycles used in the trial procedure are burnish, reset, initial baseline, first fade run (FFR), first recovery run (FRR), wear run, second fade run (SFR), second recovery run (SRR) and ending by final baseline. The test procedure and the working condition of the machine is detailed elsewhere [36]. The friction results reported in this paper refer to the progress of fade and recovery runs. The composite samples trials was initiated by burnishing it at 308 rpm speed, imposing a load of 440 N and a temperature of 93°C for 20 min. Therefore, the composite material may attain at least 95 % of contact with the drum. The FFR is simulated for around 10 min drag at 411 rpm speed and a load of 660 N. Once the FFR was finished, the FRR was

promptly initiated by turning off the heating system and friction values were recorded at 261°C , 205°C , 149°C and 93°C during the continuous cooling. On the SFR trials, a continuous drag was applied at 411 rpm speed and 660 N of load with heating on. This step was run for 10 min in which 345°C drum temperature was attained, whichever occur first. The friction values were recorded at intervals of 28°C , starting from 93°C . For SRR, friction values were recorded for each interval of 56°C , starting from 317°C after initiating cooling. The fade-recovery response was further studied taking into account various performance that define the attributes of the friction composites (see details in Table 3).

3. Results and discussion

3.1. Physical, mechanical and chemical properties

Table 4 presents the results of various characterizations of the composites. The density of the composites was found in the range of $2.23\text{--}2.44\text{ g/cm}^3$. When the amount of pineapple or Kevlar fiber was increase the density decreases. The decreasing trend in the density may be attributed to the inclusion of lighter pineapple fiber (1.56 g/cm^3) or Kevlar fiber (1.44 g/cm^3) which replace an equal amount of denser barium sulphate (4.5 g/cm^3).

Further, the porosity remains in the range of $4.34\text{--}7.36\%$ and it was found to increase with the increase of pineapple or Kevlar fiber content. The increased porosity may be attributed to the agglomeration and improper distribution of increased fibrous content in the phenolic matrix. The acetone extraction values were detected in a narrow range of 0.63 ± 0.19 , which is a sign of proper curing of the developed composites. Moreover, the highest ash content were found for lowest pineapple or Kevlar fiber reinforced composites and found to decrease with increased fibrous amount. This decreasing trend in ash content may be due to the replacement of higher heat resistant barium sulphate with lesser heat-resistant pineapple or Kevlar fiber. The compressibility and water absorption of the developed composites shows an increasing trend with increased pineapple or Kevlar fiber content. This trend may be correlated to increased porosity valve of the developed composites. The friction composite KF-1 with 5 wt.% Kevlar fiber exhibits lowest porosity (4.34 %) with least compressibility (0.88 %) and lowest water absorption (1.45 %) values. Instead, the fric-

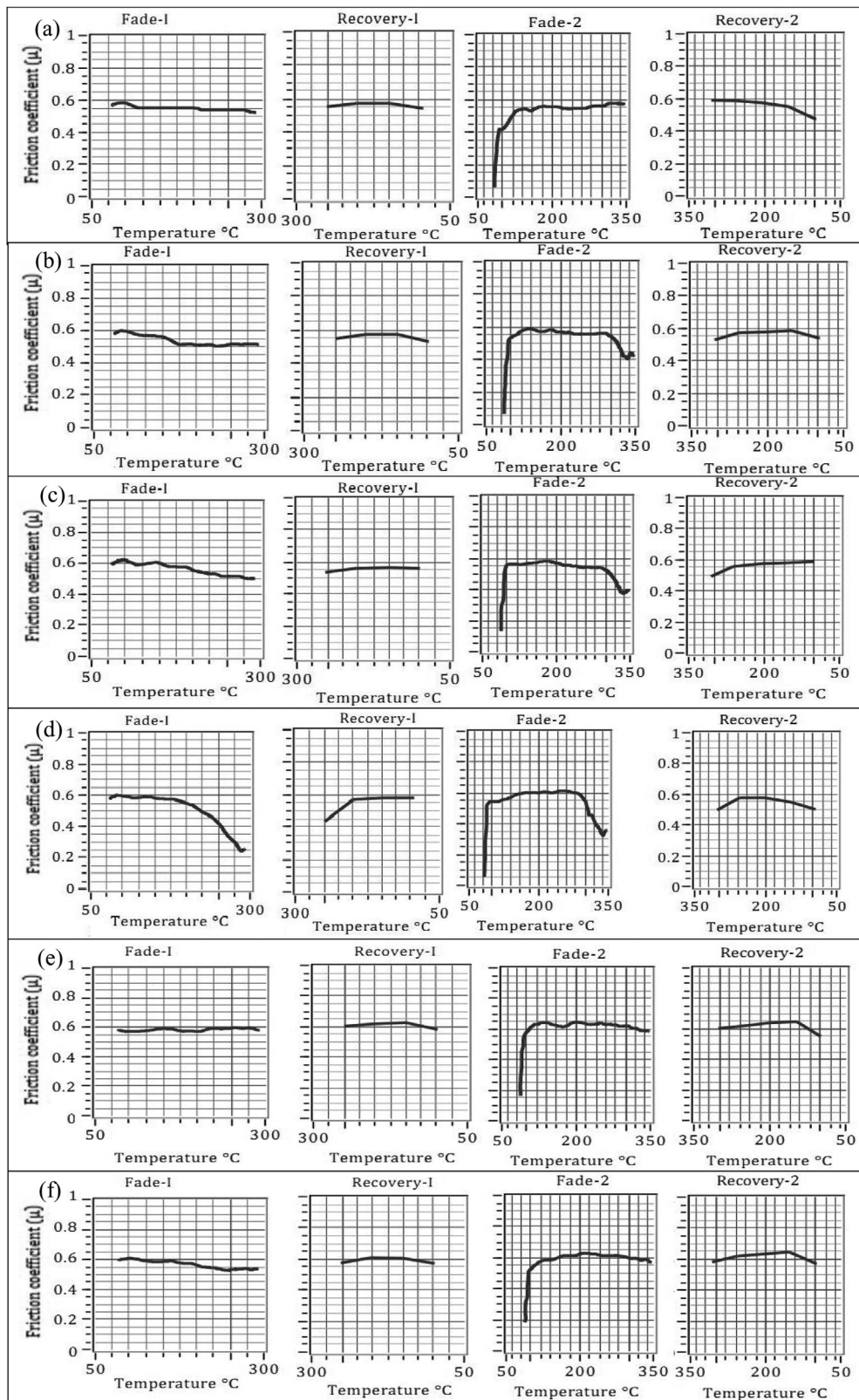


Fig. 2 – Fade and recovery response of the friction composites, (a) PF-1, (b) PF-2, (c) PF-3, (d) PF-4, (e) KF-1 and (f) KF-2.

Table 3 – Attributes used for the performance assessment.

Attribute	Explanation
μ_P	It is taken as the average μ recorded for fade and recovery cycles.
μ_F	It is taken as the lowest μ registered for the fade cycles.
Fade-%	$\frac{\mu_P - \mu_F}{\mu_P} \times 100$, lower value is enviable for good friction composites [37].
μ_R	It is taken as the highest μ registered for the recovery cycles.
Recovery-%	$\frac{\mu_R}{\mu_P} \times 100$, higher value is desirable for good friction composites [38].
Specific wear rate	Specific wear rate is computed by using following equation [39]: $\frac{w}{\rho \times t \times v_s \times \eta}$, where, w is composite weight loss (g), ρ is composite density (g/cm ³), t is the testing time (s), v_s is sliding velocity (m/s) and η is applied load (N).

Table 4 – Various properties of the developed composites.

Properties	PF-1	PF-2	PF-3	PF-4	KF-1	KF-2
Density (g/cm ³)	2.44	2.36	2.32	2.23	2.38	2.26
Porosity (%)	4.68	5.58	6.12	7.36	4.34	6.54
Acetone extraction (%)	0.64	0.60	0.74	0.82	0.44	0.52
Ash content (%)	81.46	78.94	73.12	68.96	81.62	75.41
Water absorption (%)	2.04	2.64	3.18	3.62	1.45	1.92
Compressibility (%)	1.16	1.34	1.78	2.02	0.88	1.16
Hardness (HRR)	108	104	98	96	115	112

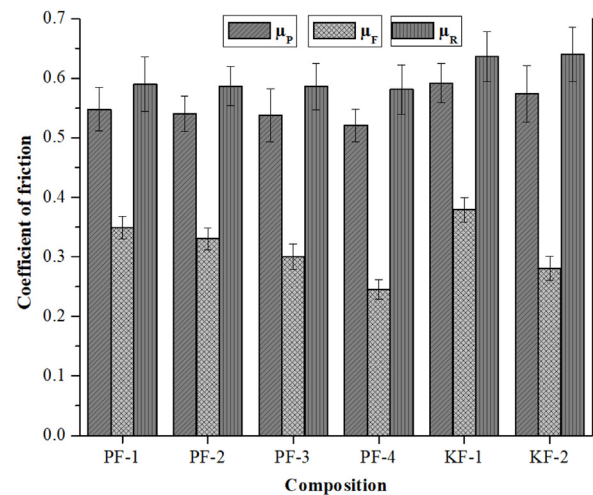
tion composite PF-4 with highest porosity value (7.36 %) may exhibits highest compressibility (2.02 %) and water absorption (3.62 %) values. The hardness (HRR) remains in the range of 96–115 and found to decrease with the increased pineapple or Kevlar content. The increased fibers content may bring structural discontinuities in the composite structure which results in a reduced hardness [40–42].

3.2. Tribological characterization of developed friction composites

3.2.1. Fade-recovery response

The fade and recovery response of the investigated composites are shown in Fig. 2. During the FFR, the coefficient of friction starts to decrease from the initial temperature i.e. 93 °C for all the composites. The composites PF-1/PF-2/PF-3 having between 5–15 wt.% pineapple fiber content indicates that the μ decreases slowly and then remain in the range of 0.513 ± 0.011 until to the end of the FFR. However, for composite PF-4 with 20 wt.% pineapple fiber content this decrease in the μ was drastic above temperature 233 °C. However, it shows a value of 0.246 at the end of FFR. Interestingly, the μ start to increase at the beginning of SFR for all the composites. This build-up in the μ was continuous until the end of SFR for composite PF-1. Above 289 °C a considerable decay in μ was observed for the composites PF-2/PF-3/PF-4 which remain in the range of 0.3–0.4 at the end of SFR. In the FRR, the composites PF-1/PF-2/PF-3 exhibit μ well above 0.5 at the end of the test, while it decreased to 0.4 for the PF-4 composite.

The SRR of PF-1 showed a continuous increase in μ value from 0.48 to 0.59 at the end of SRR whereas a substantial decay in μ was observed for PF-2/PF-3/PF-4 composites above 250 °C. The FFR of KF-1 composite reveals good frictional stability (0.58 ± 0.01) with small variation in obtained μ values as the spectrum remains almost flat whereas for KF-2 composite the μ value start diminishing after 150 °C and reached to 0.52 at the end of the run. Similar to pineapple fiber based composites; the μ start to increase at the beginning of SFR

**Fig. 3 – μ_P , μ_F and μ_R response of the composites developed.**

remains identical (0.6) in between 150–300 °C. At higher temperature >300 °C the μ value suffered a small reduction and dropped to 0.57 . The FRR and SRR of the Kevlar fiber based composites (i.e. KF-1, KF-2) remains almost identical. The μ value starts increasing in the beginning of the recovery runs and start decreasing at the termination of the recovery runs. The tribological performances computed from fade-recovery tests are presented in Figs. 3–5.

3.2.2. μ_P , μ_F and μ_R response

Fig. 3 presents the responses of μ_P , μ_F and μ_R generated by the composites considering the attributes from Table 3. The μ_P , μ_F and μ_R values of the composites were found to decrease with increased pineapple fiber content. When the fiber-based composite contains 5 wt.% (i.e. PF-1) the μ_P value was high (0.548). However, by adding of 10–15 wt.% fiber the μ_P decreases with 2 %. Further inclusion of 20 wt.% pineapple fiber reduced it

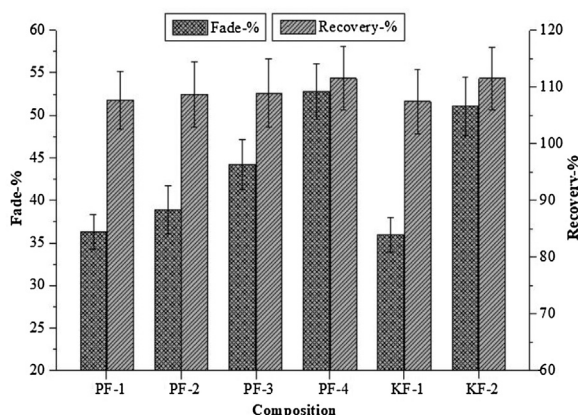


Fig. 4 – Fade-% and recovery-% response of the composites studied.

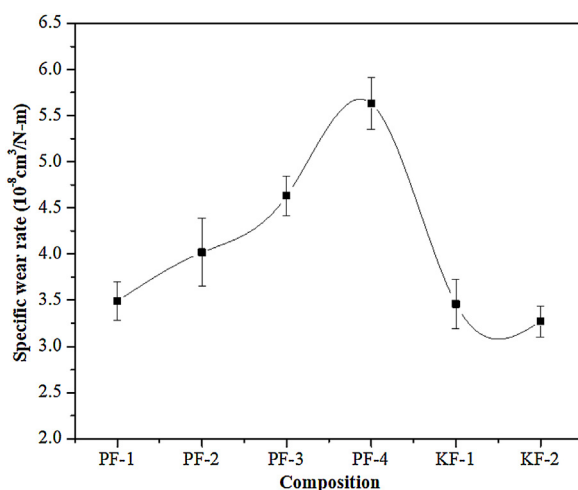


Fig. 5 – Specific wear rate of the composite.

by 5 % to a lowest value (0.521). The μ_F found to remains in the range 0.246-0.349. Were noted that the value of μ_P (0.543 ± 0.005) and μ_F (≥ 0.3) remains appreciably high in case of pineapple fiber ≤ 15 wt.%, however, the μ_R response remains comparable (0.585 ± 0.005) in the investigated composites. The decline in μ_F and μ_P , response may be attributed to the presence of an increased content of pineapple fiber in phenolic matrix that enhances the presence of adhesive component on the sliding interface. The adhesive component boost the formation of contact film by the degradation of organic ingredients during the increase in the temperature which then result in the decreased friction performance [40,41].

Further, the μ_P and μ_F remains 0.592 and 0.379 respectively for 5 wt.% Kevlar fiber based composite (i.e. KF-1) and suffered a considerable drop as the Kevlar fiber content increased to 10 wt.% (i.e. KF-2). This decrease in μ_P and μ_F in Kevlar fiber composites is attributed to the formation friction film at the braking interface which eases the movement of mating surfaces [42]. The μ_R performance for Kevlar based composites remained nearly comparable (0.638 ± 0.002). Comparatively to 5 wt.% pineapple fiber based composite (PF-1), the inclusion of 5 wt.% Kevlar fibres results 8 % enhancement in the μ_P and μ_F values of KF-1 composite. This increase in μ_P and μ_F was

attributed to the higher thermal stability of Kevlar fibers which results in the generation of load carrying friction films at elevated temperatures and hence resulted in increased frictional performance.

3.2.3. Fade-% and recovery-% response

Fig. 4 presents details of the fade-% and recovery-% responses of the composites as per Table 3 attributes. The extent of composite fade-%, PF-1 and PF-2, with the lower pineapple fiber (≤ 10 wt.%) have been observed to be almost similar with values in the range 37.60 ± 1.30 %.

On the other hand, the composites PF-3 and PF-4 with higher pineapple fiber (≥ 15 wt.%) the fade-% were found to increase. They have numerical values in the range 48.51 ± 4.27 %. Hence, we can note that the inclusion of higher pineapple fiber in the composite resulted in increased fade-%. Such as, the amount of 5–10 wt.% pineapple fiber leads to an optimal level of fade-% (37.60 ± 1.30 %) with around 23 % lower in comparison to the one of 15–20 wt.% pineapple fiber based composites. The improvement in the fade-% by inclusion of lower pineapple fiber may be attributed to the reduced organic content. In the literature is reported that at higher sliding temperature the degradation of organic content generates a decrease of frictional force and induces fade [42,43]. The recovery-% response of the composites with ≥ 15 wt.% pineapple fiber remain almost similar, in the range of 108.29 ± 0.63 %. While the composite, PF-4 with maximum pineapple fiber content (20 wt.%), show the highest fade-% (52.78 %), which exhibit as well as the highest recovery-% (111.52 %). On the other hand, the fade-% remains lowest (35.98 %) for 5 wt.% Kevlar fiber based composite i.e. KF-1 whereas it increased nearly 42 % for 10 wt.% Kevlar fibers based composite i.e. KF-2 and remains 51.05 %. The recovery-% of lower (5 wt.%) Kevlar fiber based composite (KF-1) remains 107.43 % and increased by 4%–111.50 % as Kevlar fiber content increased to 10 wt.% (i.e. KF-2). Overall the fade-% and recovery-% of the investigated composites was found to increase with increased pineapple and Kevlar fiber content. With the increase of pineapple or Kevlar fiber content, in the composites, the nature of the friction film that forms at the sliding surface compositionally becomes more organic. It lead to its easier degradation and shear thinning, hence, resulting in increased fade-% and recovery-% of the composites that agree with the experiments reported in literature [4,5,42].

3.2.4. Specific wear rate of the composites

Fig. 5 shows the influence of pineapple and Kevlar fiber concentrations on the specific wear rate of the friction composites. One can observe that the specific wear rate of the composites was deteriorated with the addition of increased pineapple fiber and improved by increasing the addition of Kevlar fiber concentration. It can be seen that the specific wear rate ($3.49 \times 10^{-8} \text{ cm}^3/\text{N-m}$) remains low for the PF-1 composite. Increases by 61 % (i.e. $5.63 \times 10^{-8} \text{ cm}^3/\text{N-m}$) were reported for PF-4 that is the highest overall. This was ascribed to the way that the inclusion of lower fibers alters the nature of the transfer film and its adherence to the counterface. In the literature, it is reported that wear remains lower for the transfer film that adhere evenly to the counterface. Whereas for higher fiber added to the composites; the wear rate remains

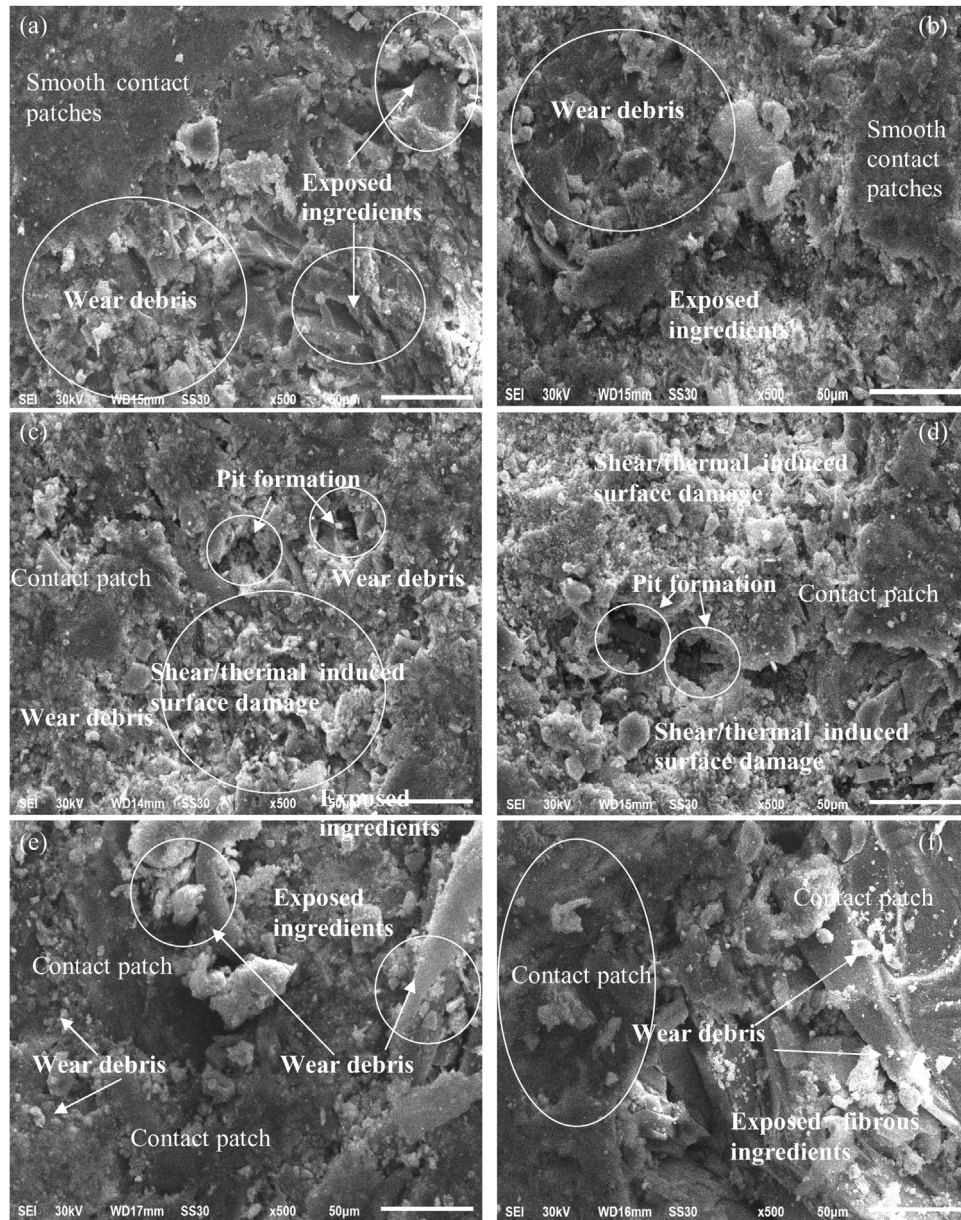


Fig. 6 – Worn surface micrographs of the composites, (a) PF-1, (b) PF-2, (c) PF-3, (d) PF-4, (e) KF-1 and (f) KF-2.

highest and may be ascribed to the degradation of the natural fibers. The natural fiber starts degrading above 200 °C, causing severe surface break and easier ingredients detachment [22–26]. Moreover, the increase of the fiber generates an agglomeration and non-uniform distribution which lead to their easy detachment and hence increased wear rate.

In contrast to the pineapple fiber based composites, the wear resistance of Kevlar fiber based composites is slightly higher. Such as, the results of Kevlar fiber based composites (KF-1, KF-2) indicate a specific wear rate that vary between $3.27 \times 10^{-8} \text{ cm}^3/\text{N}\cdot\text{m}$ and $3.46 \times 10^{-8} \text{ cm}^3/\text{N}\cdot\text{m}$ which is nearly 1–6 % lower to 5wt.% pineapple fiber based composites (i.e. PF-1). It was reported in the literature that the relative amount and aspect ratio of Kevlar fiber played a vital role in improving the wear performance of the brake composite materials. Increase in wear resistance with increased aramid

fiber concentration was reported by N. Aranganathan et al. [12], whereas with the same concentration of aramid fiber (with different aspect ratio), lower wear rate was reported for shorter fiber based composites by P. Cai et al. [44].

3.2.5. Worn surface study

To understand the wear mechanism, composites worn surfaces were characterized using SEM and obtained morphologies are presented in Fig. 6. Generally, the formation of contact patches or tribo-film plays a crucial role in defining the friction and wear performance of the composites [45]. The contact patches or tribo-film formation primarily depends on the nature of ingredients as well as the composite working conditions. The contact patch or tribo-film mainly arises due to compaction of the wear debris that largely comprise organic ingredients like phenolic resin, Kevlar and pineapple fibers etc.

with increased temperature generated at the braking interface [46]. The worn surfaces of composite PF-1/PF-2 (Fig. 6a-b) with lower pineapple fiber content (i.e. ≤ 10 wt.%) shows larger extend of contact patches which helps in wear minimization. Moreover, the generated wear debris was act as third body and increases the friction coefficient by rolling abrasion mechanism [47]. The morphologies of the composite with higher pineapple fiber content i.e. PF-3/PF-4 (Fig. 6c-d) clearly shows severe surface damage which may be occur due to the thermal and shear stresses that resulted in the thermal fatigue failure of the composites. The thermal fatigue promotes the removal of the material by adhesive means and resulted in pit formation, which corresponded to low wear resistance of PF-3/PF-4 composites. Moreover, the worn surfaces exhibit rough surface morphology with higher amount of pulled-out ingredients with lesser amount of contact patches. The higher fibrous concentration resulted in increased exposure of pineapple fiber towards the tribo-interface. With increased exposure; these fibers were easily detached from the composite surface with other ingredients and resulted in increased wear as found experimentally. Hence, the adhesive and fatigue mode of wear were the main wear mechanisms of PF-1/PF-2 composites.

The worn surface of composite KF-1 (Fig. 6e) appeared to smooth as covered by contact patches but the same time it contains some wear debris and fibers. In general, the formation of smooth contact patches helps in wear minimization and meanwhile the wear debris contribute in the enhancement of friction performance by rolling abrasion mechanism. As shown in Fig. 6f, the KF-2 composite surface was mostly covered with smooth contact patches and lesser amount of wear debris were appeared to be scattered on the worn surface indicating its best wear performance. As the sliding interface temperature increased, the phenolic resin start degrading which weaken the bonding of ingredients with matrix leading to their detachment as wear debris. The compression/compaction wear debris at increased shear and temperature conditions results in the formation of a smooth contact patch on the composite surface and amply reported in wear minimization [42-47].

4. Conclusions

Friction composite materials based on pineapple and Kevlar fiber content were developed and evaluated for physical, mechanical and tribological properties. The following conclusions can be drawn from this study:

- The increase of pineapple or Kevlar fiber contents led to the decrease of density, hardness and ash content as well as it can increase the water absorption, porosity and compressibility.
- The lowest fade-% with highest friction performance and wear resistance was recorded for friction composite containing 5 wt.% pineapple fiber content i.e. PF-1, whereas friction composite PF-4 containing 20 wt.% presented the highest recovery-% with lowest wear resistance.
- A comparison of the performance of pineapple fiber and Kevlar fiber, showed that 5 wt.% Kevlar fiber based composite register highest performance coefficient of friction but

fade-%, recovery-% and wear performance remains almost identical with 5 wt.% pineapple fiber based composite.

- Finally, it can be concluded that 5 wt.% pineapple fiber exhibits comparable tribological properties to 5 wt.% Kevlar fiber and can be used in the production of non-asbestos friction composites for automotive industry.

The authors confirm that this work has not been published elsewhere and also it has not been submitted simultaneously for publication elsewhere.

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