Applicability of cellulosic-based *Polyalthia longigolia* seed filler reinforced vinyl ester biocomposites to tribological performance

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Abstract

The focus of this work was to analyze the effect of weight percentage (wt.%) of *Polyalthia longigolia* seed filler (PLSF) on the wear responses (wear loss and coefficient of friction) of the vinyl ester (VE) matrix composites, using signal-to-noise (SN) ratio and analysis of variance (ANOVA) methods. The *Polyalthia longigolia* seed filler/vinyl ester (PLSF-VE) composites were produced by compression molding (CM) technique. Scanning electron microscopy (SEM) analysis showed that the PLSF content was homogeneously distributed in the matrix. Pin on disc (POD) wear tester was employed to carry out the experiments. Both SN ratio and ANOVA were performed to determine the process parameters that exhibited lower wear responses. The wear of the composite was minimized by optimizing the four diverse process factors: load, *P* (N), sliding speed, *N* (rpm) and filler content (wt.%), based on Taguchi's L_9 orthogonal array. The process parameters at which minimum wear loss (WL) occurred were identified with 25 wt.% sample at 10 N and 300 rpm. Also, the process parameters for minimum coefficient of friction (COF) were observed with 25 wt.% sample at 5 N and 700 rpm. From the ANOVA results obtained, it was evident that *P* mostly influenced the WL and COF of the PLSF-VE composites during POD wear testing. This kind of lightweight composite can be a suitable alternative for small scales loading conditions, such as brake bad and clutch plates in the automobile industry.

Keywords: Sliding wear, polymer matrix composites, particles, friction.

1 INTRODUCTION

Composite materials are being established to increase the properties and announce novel products.[1,2] Polymers are used as both smart engineering materials and matrix materials for composite material, since they have high specific strengths and corrosion resistances. Polymer-based composites are considered or used majorly, because they have variety of compositions and flexibility in design.[3] Different types of filler materials are utilized as reinforcement materials to produce polymer matrix composites. Plant and animal-sourced types are used as natural fillers. Natural fibers are mostly from vegetables. Natural fibers are environmentally friendly materials and these have outstanding properties preferable to many man-made fibers.[4] Natural fiber polymer composites contain polymer as matrix that entrenched through extraordinary property of the natural fibers, such as bagasse, banana ribbon, Indian mallow, kusha grass, red banana peduncle, nendran, vetiver, banana peduncle, jute, flax, oil palm, kenaf and sisal.[5-13]

In addition, bio reinforcements have been used to produce polymers composites, because of their better mechanical and temperature properties. Hence, they are used in a variety of applications. For instance, many researchers used red mud, fly ash, rice husk and sawdust as reinforcement materials.[14] Stalin et al. studied the tamarind seed filler-composites and observed that the composites can be used to manufacture various engineering products.[15] Rajamuneeswaran et al.[16] investigated the accumulation of chitosan particles in coir fiber/vinyl ester (VE) polyester composites and reported a greater increment in their mechanical characteristics. Nagarajan et al.[17] studied the influence of agro-waste α-cellulosic filler on thermomechanical behaviors of epoxy composites. The tensile, flexural and impact behaviors of the composites increased, following a linear trend. Agunsoye et al.[18] studied into the mechanical behaviors of *Delonix regia* seed filler/polymer composites. Babu et al.[19] investigated the tribological and physical behaviors of wear debris powder reinforced epoxy composites for structural application. Suresha et al.[20] conducted the investigation on mechanical and tribological behaviors of VE composites. The hybrid composites showed better tribological and mechanical performances, which was suggested for bearing used in the automobile industries.

Moreover, Shivamuthy et al.[21] reported the sliding wear and mechanical behaviors of jatropha seed cake waste/epoxy composites. They suggested that the composites were well suitable for high-load and high-speed applications. Madhu and Balasubramanian[22] reported that plant-based natural fibers could be used in automobile application, due to their low weight and high fiber strength. They concluded that these fibers improved the composite properties. Anand et al.[23] analyzed the wear properties of glass fiber/Al_2O_3 nanowire fillers reinforced VE composite for structural application. Suresha et al.[24] examined the tribological behaviors of glass/carbon fiber reinforced VE composites. Their results depicted that the coefficient of friction (COF) and wear rate increased with an increment in load, sliding velocity and based on reinforcement of fiber content and interphase temperature. And the reported carbon-VE composite was suitable for bearing applications.

Also, Chauhan et al.[25] reported the effect of fly ash particulate on tribological behavior of glass fiber reinforced VE composites in water-lubricated and dry sliding environments. The study established that COF decreased with an increase in applied normal load. With the addition of fly ash particles, the specific wear rate of composite was significantly reduced. Suresha and Kumar[26] studied the two-body abrasive wear characteristics and mechanical performance of VE composites reinforced with glass/carbon fiber. The assessed wear volume loss increased with an increase in the abrasive particles size and abrading distance. Therefore, the specific wear rate declined as the abrading distance increased and the abrasive particle size decreased.

The present study investigated into the wear loss (WL) and COF of a newly developed composite material with an enhanced tribological properties. Some studies have been conducted on wear behaviors of some polymer matrix composites,[27–29] as subsequently summarized. Iyer et al.[30] reported the wear of the nano-HA and nano-carbon fibers filled polymer-based composites for medical products. Sudeepan et al.[31] investigated the wear and COF of ABS/ZnO polymer composite, utilizing Taguchi technique and reported that COF and wear were expressively affected by the intensification of filler content, load, P (N) and sliding speed, N (rpm). Kumar and Reddy[32] reported the wear of nylon-BN composites for varying filler, sliding distance, P and N, using the Taguchi's design of experiment. From the aforementioned studies among other experimental investigations, it is observed that the tribological study of an innovative *Polyalthia longigolia* seed filler/vinyl ester (PLSF-VE) composites has not been reported, despite of their various possible benefits or potential applications. Paul et al.[33] reported the tribological behavior and extensive applications of natural filler/fiber reinforced polymer-based composites in the automotive industry, including body and door panels, lightweight seats, engine covers, seat surfaces, roof covers, parcel shelves, headliner and insulation panels, interior carpets, dashboards and other interior and exterior parts.

Therefore, this present study aimed to examine the influence of weight filler contents, P and N on the WL and COF of the PLSF-VE composites, using Taguchi method. L_9 orthogonal array was used for the three parameters at three levels, each. Analysis of variance (ANOVA) and signal-to-noise (SN) ratio were used to establish significance of the process parameters used and validity of the experimental results obtained.

2 EXPERIMENTAL DETAILS

2.1 Materials

The mast tree (*Polyalthia longigolia*) belongs to the family of *Annonaceae* and its seeds were sourced from Madurai, Tamil Nadu, India. Manual method was used to remove coats from the seeds, and later cleaned up using distilled water. The two-week dried *Polyalthia longigolia* seeds were ball milled to fine powder. The average particle size of the filler was 25-50 µm. Vinyl ester resin was used as a matrix. Bisphenol-A-epoxy VE resin along with N-Dimethylaniline as an accelerator, methyl ethyl ketone peroxide as catalyst and cobalt naphthenate as a promoter were utilized. They were purchased from Coimbatore Seenu Company, Tamil Nadu, India. Compression molding (CM) technique was used to produce the PLSF-VE composites with 0, 25 and 50 wt.%.

2.2 Sample preparation

The preparation phases of *Polyalthia longigolia* seed filler (PLSF) is shown in Figures 1(a)-(d). Measured quantity of PLSF, such as 5 wt.% was put into the resin and constantly stirred for 20 minutes. Then, the resin was blended with accelerator, catalyst and promoter of 1.5 wt.% each. It was gradually decanted into a wax coated mold cavity size of 200 x 200 x 3 mm. After it has been wholly poured into the mold cavity, the mold was closed with upper die and compressed with a pressure of 100 KPa. Lastly, the whole set-up was left for 24 h at room temperature for better curing.[34-35]

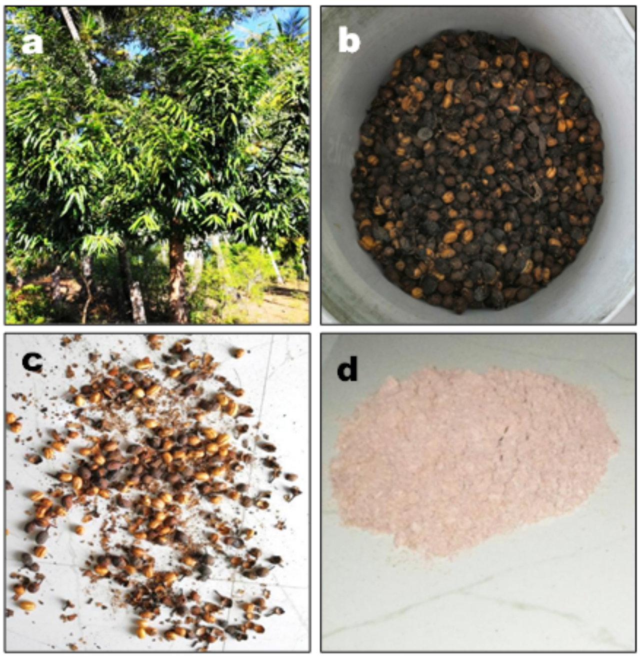

FIGURE 1 Preparatory stages of *Polyalthia longigolia* seed filler, showing its (a) tree, (b) seed, (c) extraction of seed and (d) final seed filler

2.3 Wear analysis

The wear loss tests were executed on POD apparatus. The POD machine set-up and wear samples are depicted in Figures 2(a) and (b), respectively. The process parameters and levels used are highlighted in Table 1. The PLSF-VE composites of 0, 25 and 50 wt.% with the dimension of 8 x 8 x 30 mm each, were used as test samples. The weights of pin and disc were measured in a digital balance with an accuracy of 0.0001 mg. The full details of the parameters used and the WL and COF results obtained are depicted in Table 2. *P* (N), Wt.% of PLSF and *N* (rpm) were taken as the parameters, while WL and COF were taken as the wear responses from the present study. Minitab 17 software was utilized to carry out the SN ratio and ANOVA.

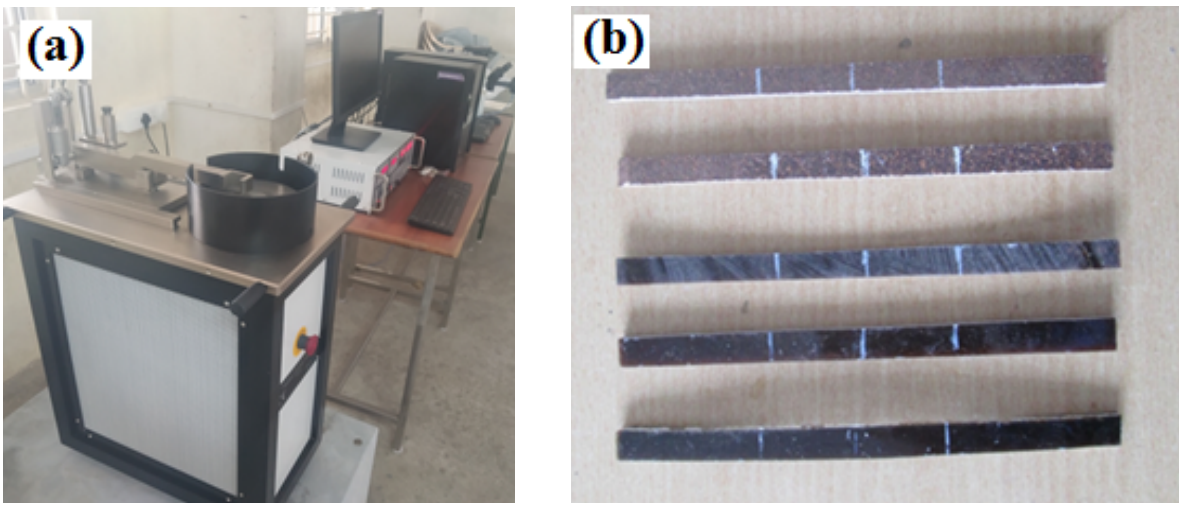

FIGURE 2 (a) POD machine and (b) fabricated PLSF composite samples

TABLE 1 Factors and their levels

Factor	Type	Level	L1	L2	L3
Load, *P* (N)	Fixed	3	5	10	15
Filler content (wt.%)	Fixed	3	0	25	50
Sliding speed, *N* (rpm)	Fixed	3	300	500	700

TABLE 2 Experimental plan, WL and COF results obtained

Ex. No	Load, *P* (N)	Filler content (wt.%)	Sliding speed, *N* (rpm)	Wear loss, WL (μm)	COF	SNRA1	SNRA2
1	5	0	300	52.72	2.71	-34.4395	-8.65939
2	5	25	500	50.31	2.11	-34.0331	-6.48565
3	5	50	700	48.84	3.7	-33.7755	-11.3640
4	10	0	500	42.65	5.76	-32.5984	-15.2084
5	10	25	700	47.05	3.97	-33.4512	-11.9758
6	10	50	300	33.11	4.5	-30.3992	-13.0643
7	15	0	700	87.78	1.83	-38.8679	-5.24902
8	15	25	300	51.05	2.75	-34.1599	-8.78665
9	15	50	500	72.55	5.78	-37.2127	-15.2386

3 RESULTS AND DISCUSSION

3.1 SEM analysis

Figures 3(a) and (b) depict the SEM images of PLSF-VE composite samples with 25 and 50 wt.% filler contents, respectively. Figure 3 shows the consistent spreading or uniform distribution of the PLSF content in the matrix. This uniform spreading of filler content is important to obtain good microstructure of the newly proposed composite samples. Precisely, the 25 wt.% composite sample with better microstructure consequently exhibited improved wear resistance properties (Figure 3a), in addition with an expected enhanced mechanical behavior. This was achieved by the proper selection of fabrication parameters and method.

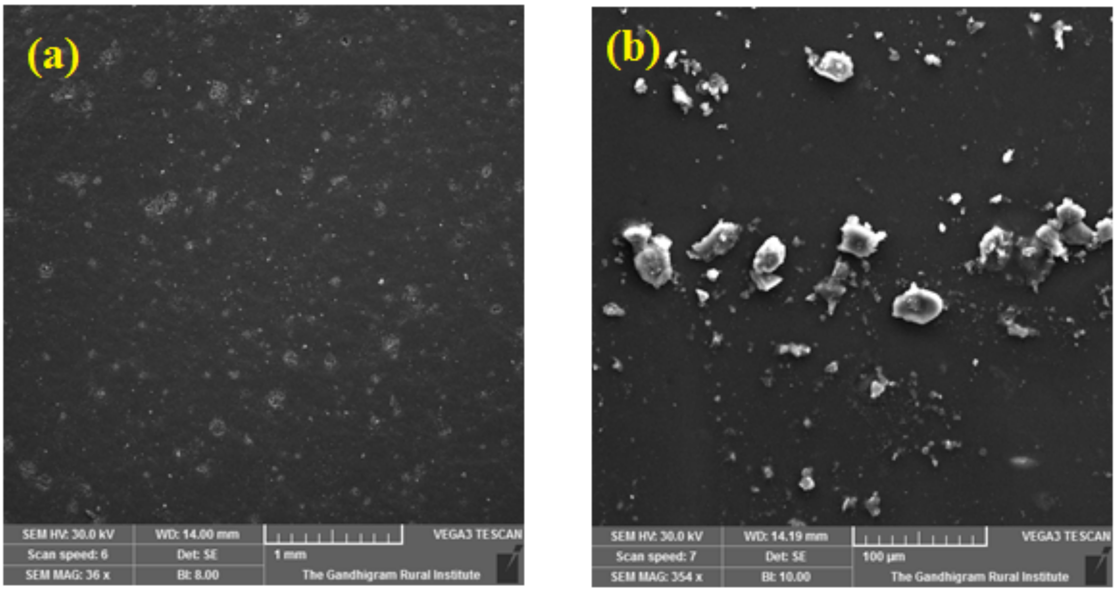

FIGURE 3 SEM images of PLSF-VE composites at (a) 25 and (b) 50 wt.% fillers

3.1 SN ratio analysis for wear and COF

Tables 3 and 4 show various responses from wear using SN ratio and Means methods, respectively. The rank is provided based on the delta value given in both Tables. Both SN ratio and Means Tables provide the same ranking for the parameters. Figures 4 and 5 show the SN ratio and Means plot for the wear response, respectively. From the Tables and plots, it was evident that the *P* was the most dominant factor for the wear response. Pattanaik et al. reported similar results for epoxy resin-fly ash composites. The *N* was the second factor which affected the wear of the PLSF-VE composites, followed by filler content.[36] The same observations were obtained from the Means Table and plots.

TABLE 3 Responses from wear using SN ratio method

Level	Load, *P* (N)	Filler content (wt.%)	Sliding speed, *N* (rpm)
1	-34.08	-35.30	-33.00
2	-32.15	-33.88	-34.61
3	-36.75	-33.80	-35.36
Delta	4.60	1.51	2.37
Rank	1	3	2

TABLE 4 Responses from wear using Means method

Level	Load, *P* (N)	Filler content (wt.%)	Sliding speed, *N* (rpm)
1	50.62	61.05	45.63
2	40.94	49.47	55.17
3	70.46	51.50	61.22
Delta	29.52	11.58	15.60
Rank	1	3	2

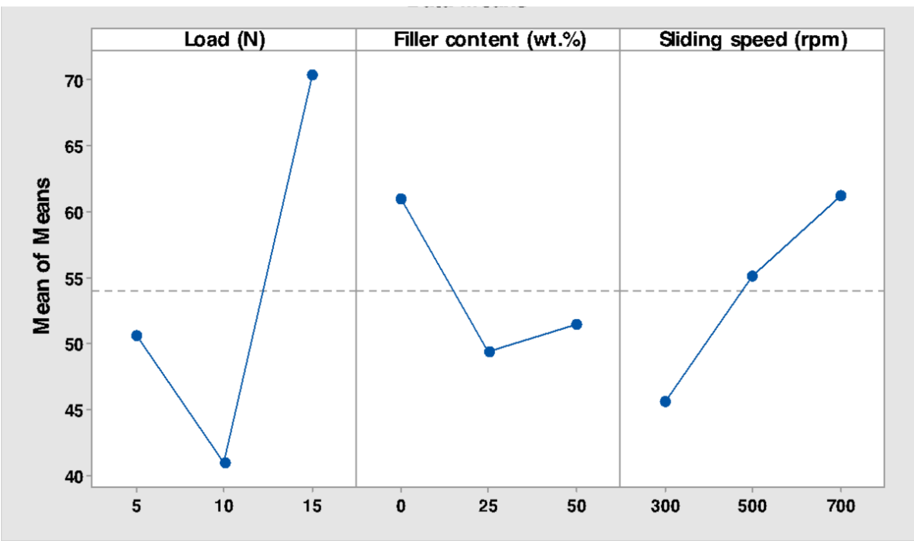

FIGURE 4 Main effect plot for wear (SN ratio)

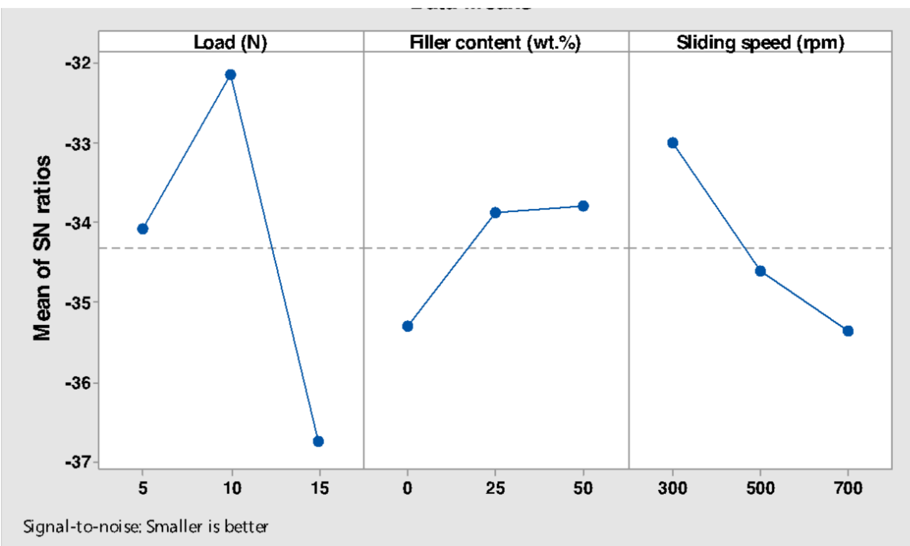

FIGURE 5 Main effect plot for wear (Means)

Tables 5 and 6 show the COF responses, using SN ratio and Means methods. Both SN ratio and Means Tables provide the same ranking for the parameters with respect to the COF responses. Figures 5 and 6 depict the SN ratio and Means plot for the COF, respectively. From the SN ratio Tables and plots, it was evident that the *P* was the most dominant factor for the COF of the PLSF-VE composites, followed by *N* and filler content. The same observations were obtained from the Means Table and plots for the COF responses.

TABLE 5 COF response using SN ratio method

Level	**Load, *P* (N)**	**Filler content (wt.%)**	**Sliding speed, *N* (rpm)**
1	-8.836	-9.706	-10.170
2	-13.416	-9.083	-12.311
3	-9.758	-13.222	-9.530
Delta	4.580	4.140	2.781
Rank	1	2	3

TABLE 6 COF response using Means method

Level	**Load, *P* (N)**	**Filler content (wt.%)**	**Sliding speed, *N* (rpm)**
1	2.840	3.433	3.320
2	4.743	2.943	4.550
3	3.453	4.660	3.167
Delta	1.903	1.717	1.383
Rank	1	2	3

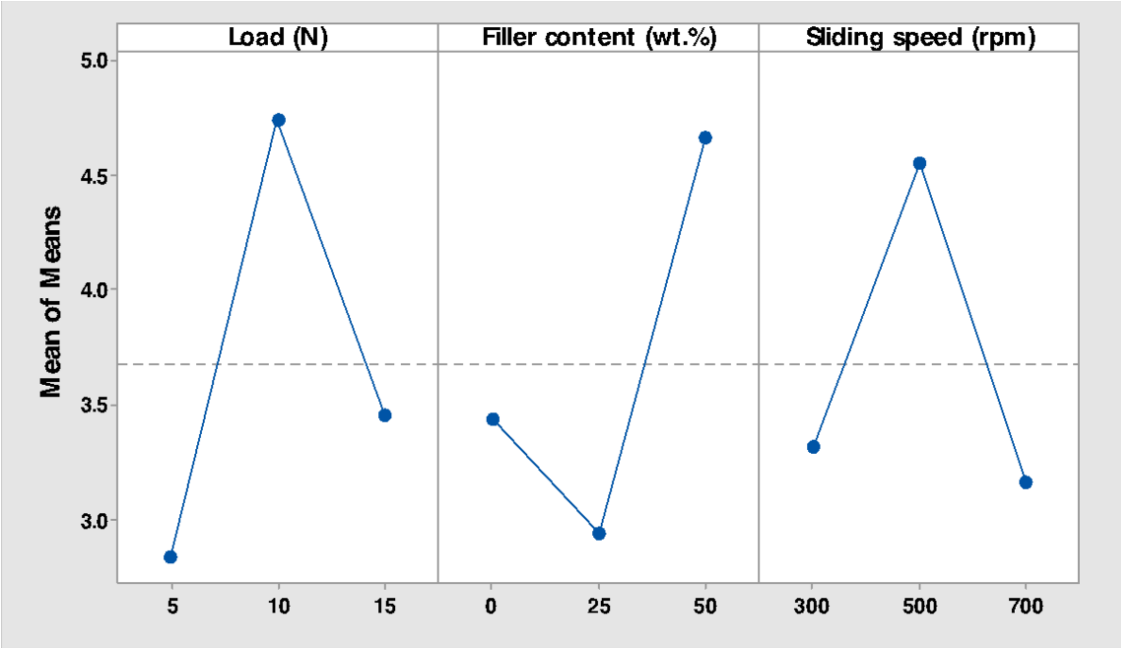

FIGURE 6 Main effect plot for COF (SN ratio)

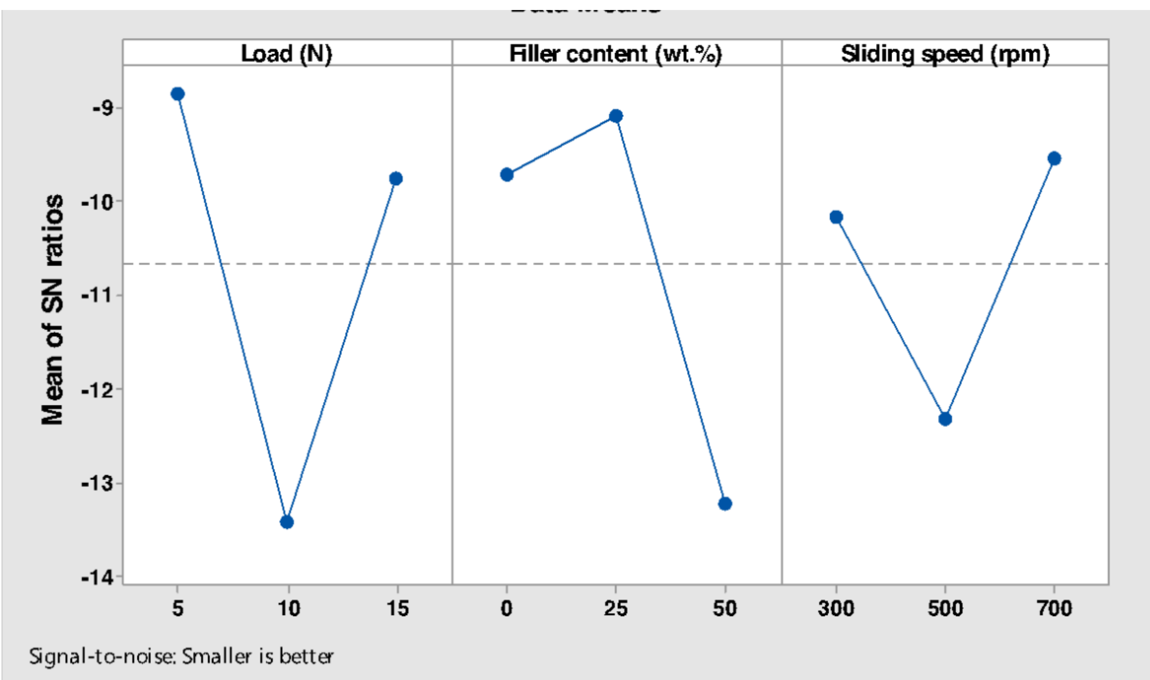

FIGURE 7 Main effect plot for COF (Means)

3.2 Interaction plot for wear and COF

Figures 8 and 9 depict the interaction plot for the wear and COF, respectively. From the plots, it was observed that the *P* was the most dominant factor for the responses of wear and COF, when interacting with filler content and *N*. When interacting with filler content and *N*, *N* recorded higher dominating influence on the COF and wear of the PLSF-VE composites. The reason was that when the *P* was increased, more wear was observed, because of the compressive action of the pin. Addition of filler content decreased the wear response of the PLSF-VE composite samples, irrespective of the values of *P* and *N*. Dalbehera and Acharya[37] reported that an addition of fillers (jute-glass) reduced the wear of the fabricated composite, using Taguchi experimental design.

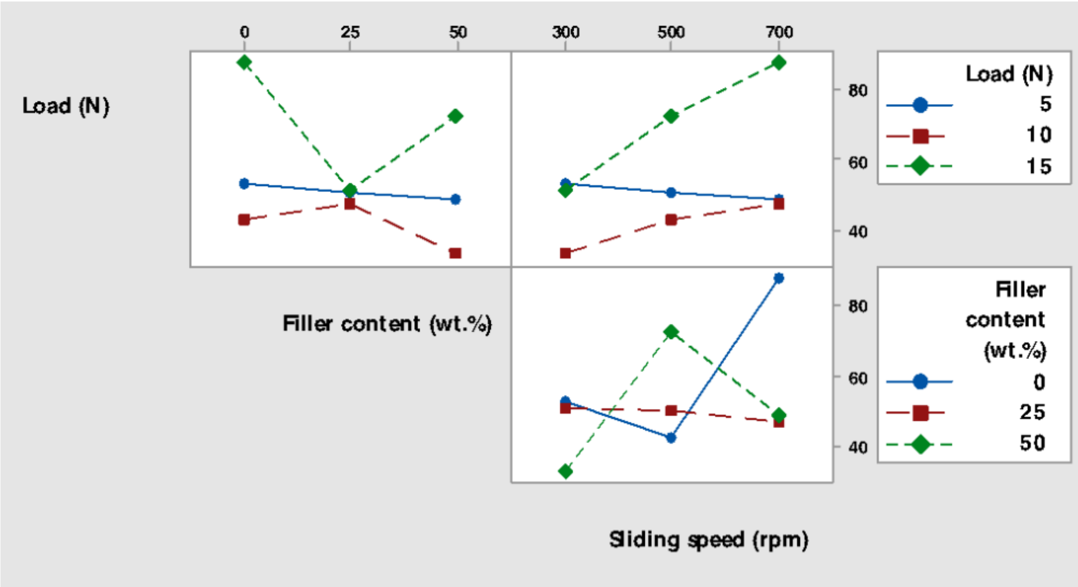

FIGURE 8 Interaction plot for wear

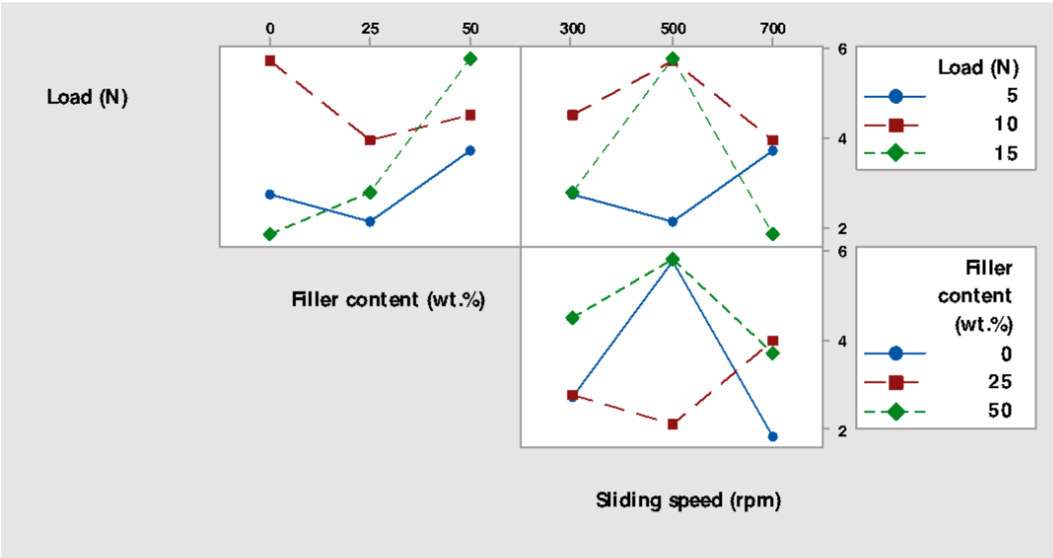

FIGURE 9 Interaction plot for COF

3.3 Contour plots analysis

Figures 10-12 and 13-15 show the contour plot for the wear rate and COF of the PLSF-VE composites, respectively after the analysis. Figure 10 depicts the contour plot for wear, drawn between *P* and filler content. From the plot, it was observed that the *P* was the dominant factor impacting wear of the PLSF-VE composites, during wear testing. An increase in the PLSF content in the VE matrix decreased the wear. Figure 11 depicts the contour plot for wear, plotted between *N* and filler content. The low filler content and high *N* led to high wear. Filler content was the most influencing factor for the wear of the tested composite samples, similar to the report of Balan and Ravichandran.[3] Figure 12 presents the contour plot for wear, developed between *P* and *N*. From the plot, it was evident that an increase in *P* resulted to an increment in the wear, irrespective of the value of *N*. Figure 13 provides the contour plot for COF, obtained between P and filler content. The plot shows that an increase in *P* caused an increase in the COF. Also, an increase in filler content reduced the COF during testing. Figure 14 shows the contour plot for COF, plotted between *N* and filler content. The plot depicts that an increment in filler content produced a decrease in the COF. For the higher filler content, a lower COF was observed. Figure 15 displays the contour plot for COF, plotted between *P* and *N*. *P* recorded a higher significant effect on COF, when compared with *N* during the sliding of the composite against the pin, similar to the previously reported studies.[31,38-40]

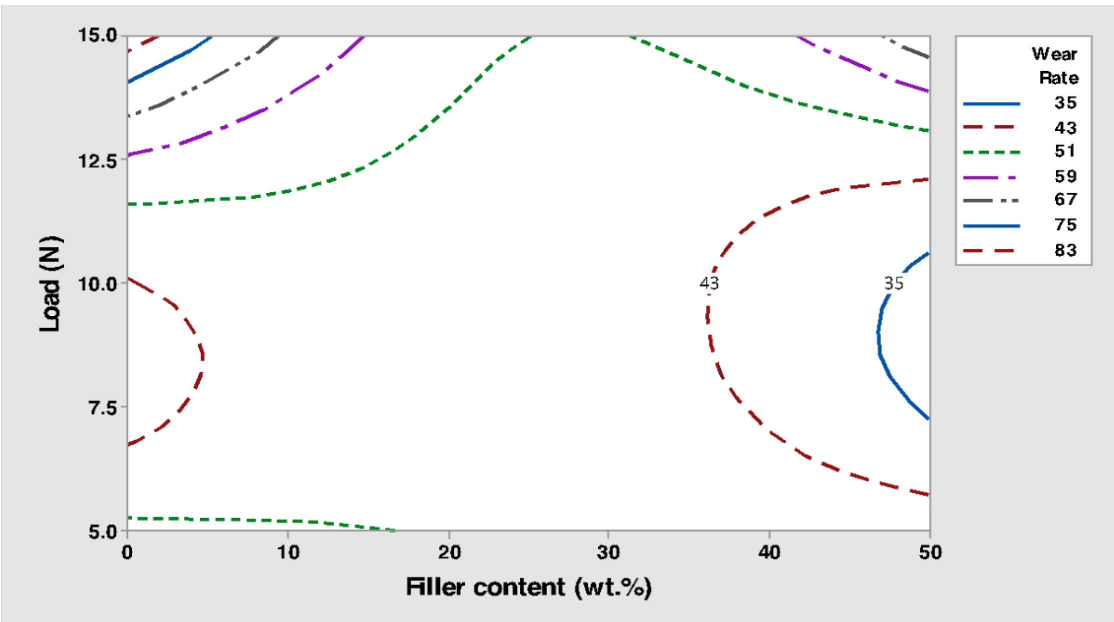

FIGURE 10 Contour plot for wear: Load *versus* filler content

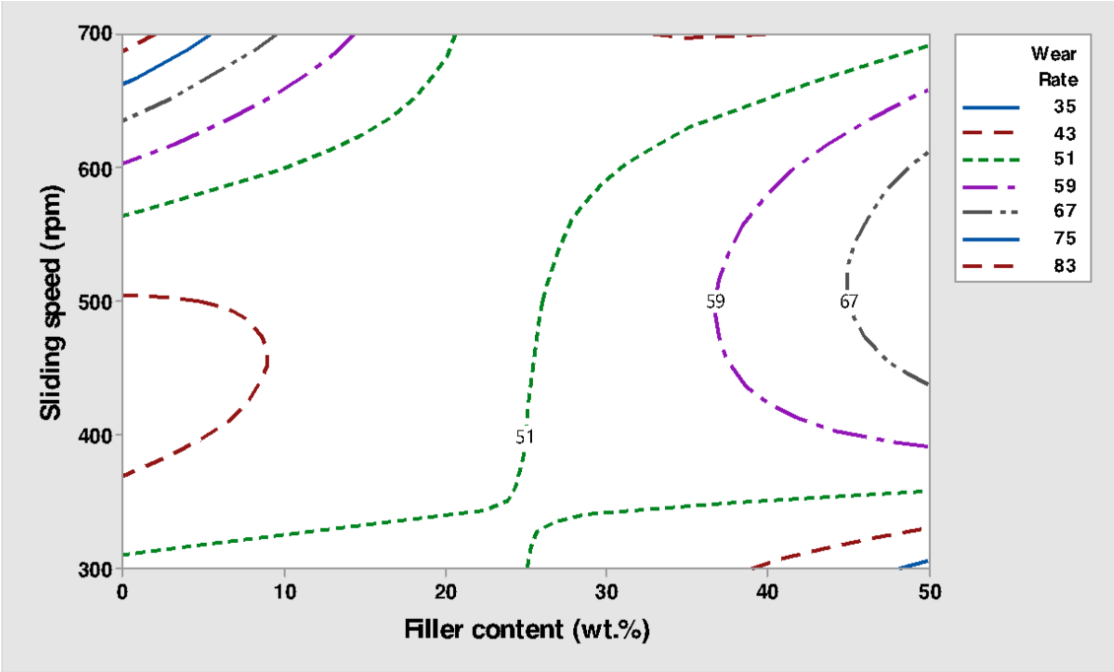

FIGURE 11 Contour plot for wear: Sliding speed *versus* filler content

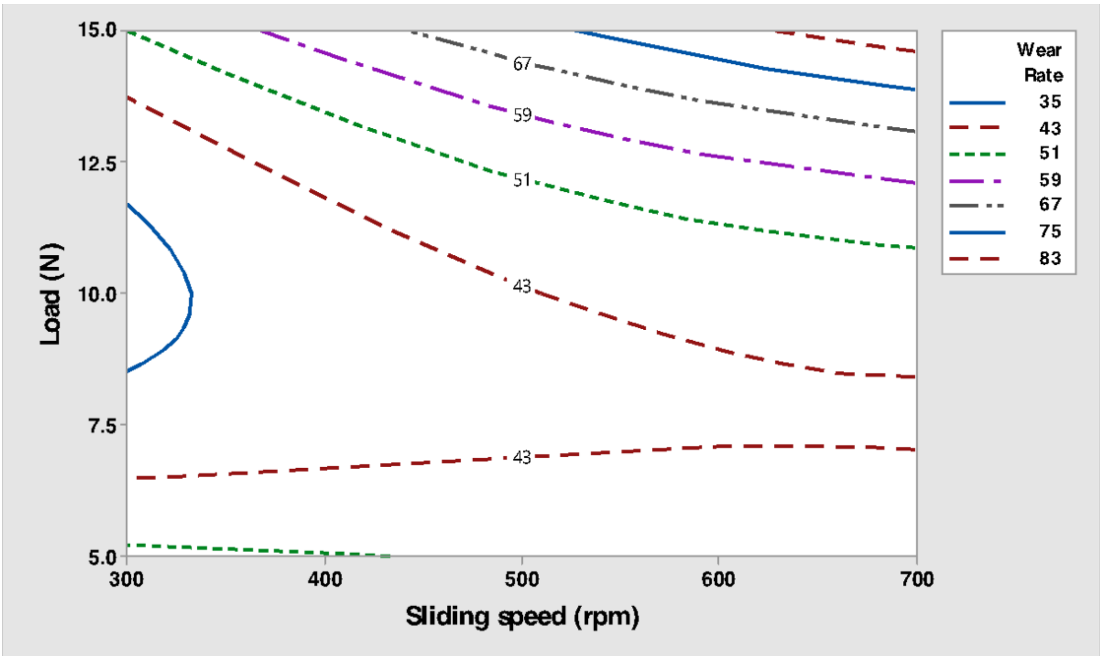

FIGURE 12 Contour plot for wear: Load *versus* sliding speed

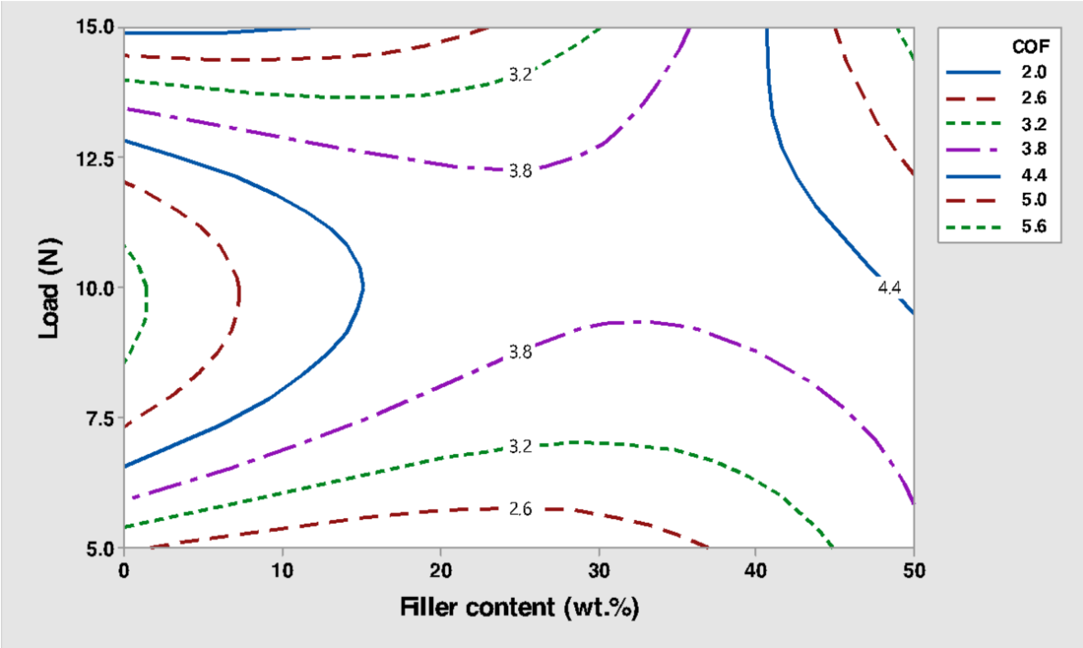

FIGURE 13 Contour plot for COF: Load *versus* filler content

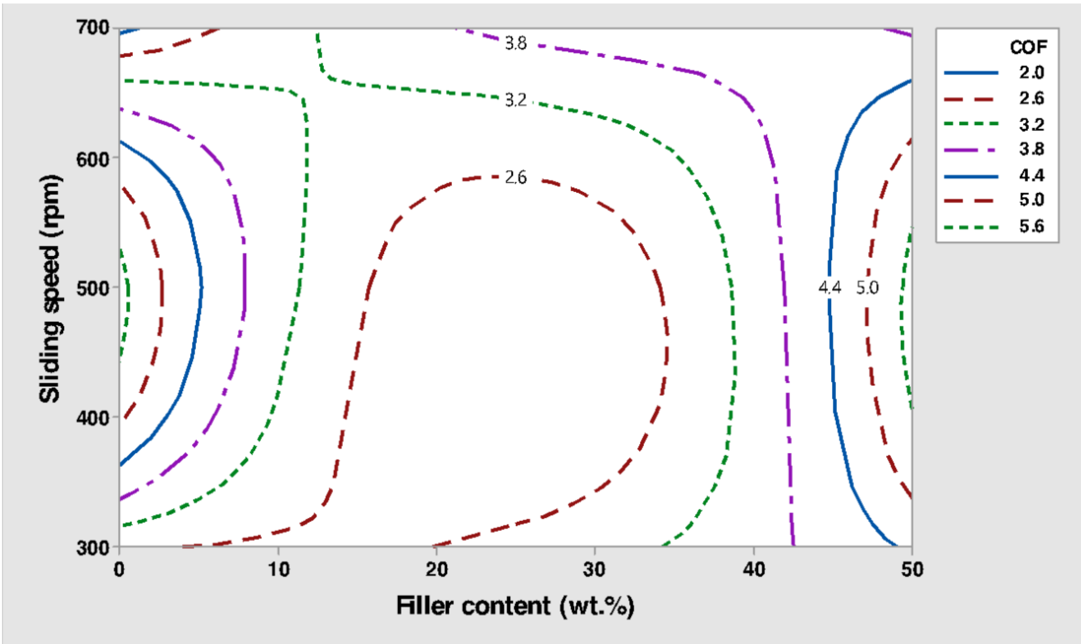

FIGURE 14 Contour plot for COF: Sliding speed *versus* filler content

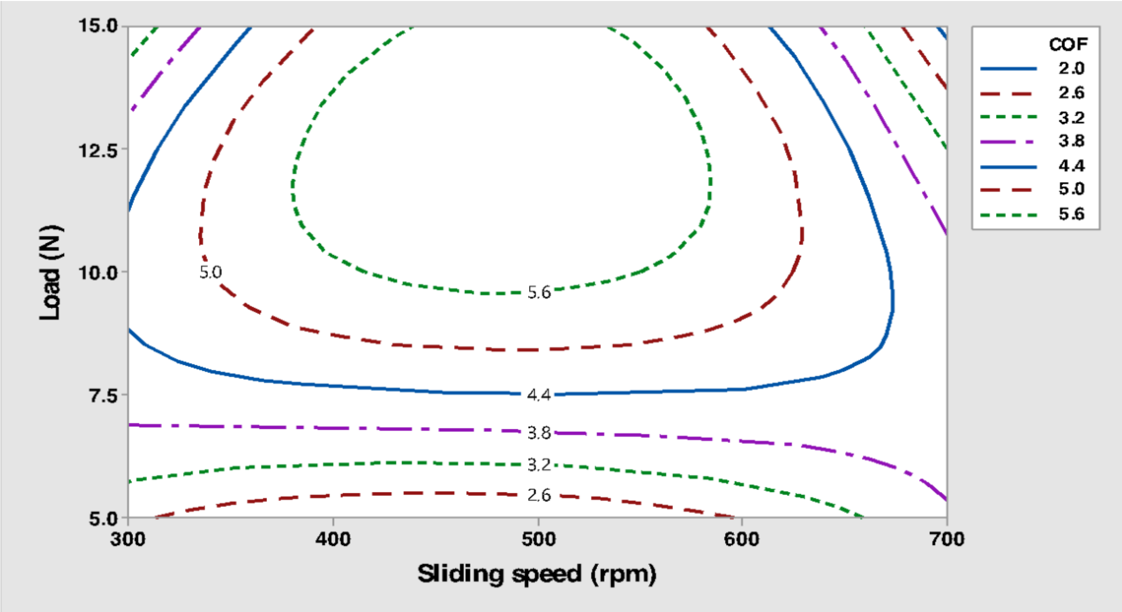

FIGURE 15 Contour plot for COF: Load *versus* sliding speed

3.4 ANOVA analysis for wear and COF

ANOVA, a statistical technique was utilized to establish the order of importance of the process parameters on the wear and COF of PSLF-VE composite. The results of the ANOVA on the SN ratios for the wear and COF values are presented in Tables 7 and 8, respectively. The significance of controlling factors was determined by juxtaposing their P-values. Based on the P-values obtained, as shown in Table 7, the wt.% of filler content was observed to be the significant determinant factor for wear, while *N* was the substantial factor for the rate of COF [22], as depicted in Table 8.

TABLE 7 ANOVA for wear

Source	**DF**	**Seq SS**	**Adj SS**	**Adj MS**	**F**	**P**
Load, *P* (N)	2	1358.95	1358.95	679.48	7.15	0.123
Filler content (wt.%)	2	229.42	229.42	114.71	1.21	0.453
Sliding speed, *N* (rpm)	2	370.97	370.97	185.49	1.95	0.339
Error	2	189.94	189.94	94.97		
Total	8	2149.29				
S = 9.74536, R-Sq = 91.16% and R-Sq(adj) = 64.65%						

TABLE 8 ANOVA for COF

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Load, *P* (N)	2	5.663	5.663	2.831	1.67	0.374
Filler content (wt.%)	2	4.692	4.692	2.346	1.39	0.419
Sliding speed, *N* (rpm)	2	3.450	3.450	1.725	1.02	0.495
Error	2	3.382	3.382	1.691		
Total	8	17.186				
S = 1.30034, R-Sq = 80.32% and R-Sq(adj) = 21.29%						

Figures 16 and 17 illustrate the residual plot for wear and COF, respectively. Figure 18 depicts the probability plot for wear and Figure 19 provides the probability plot for COF. The errors were distributed in straight. This is shown in the normal probability plot for wear and COF. The arrangements of points are judiciously straight and inside the confidence intervals lines.[32]

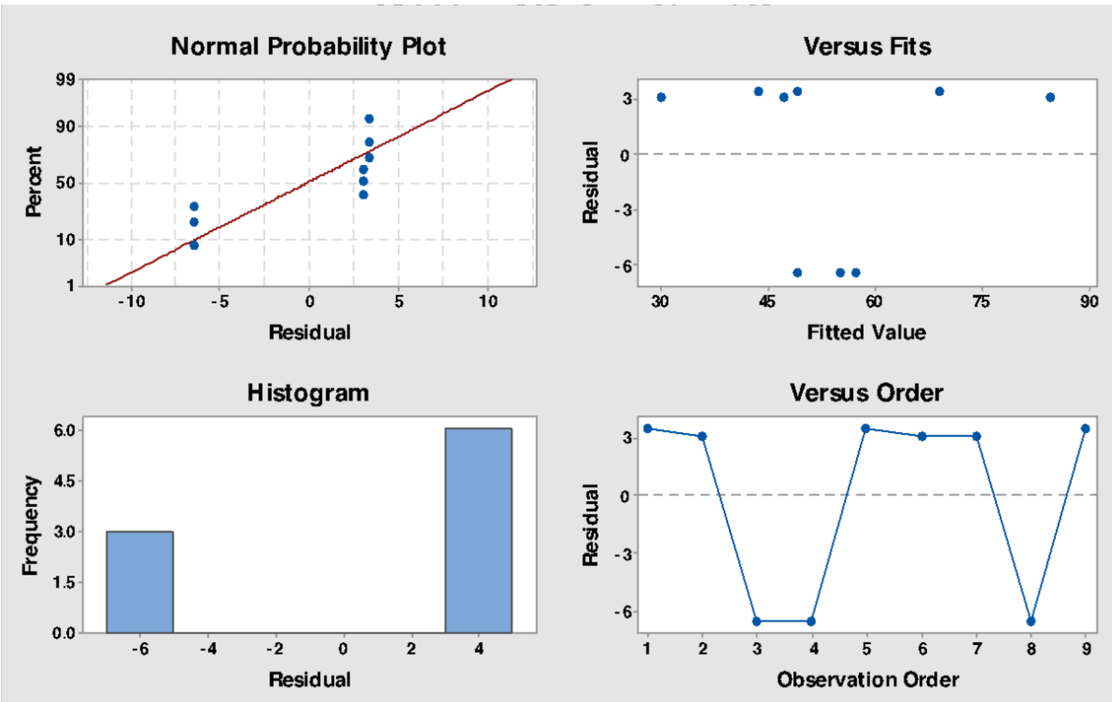

FIGURE 16 Residual plot for wear

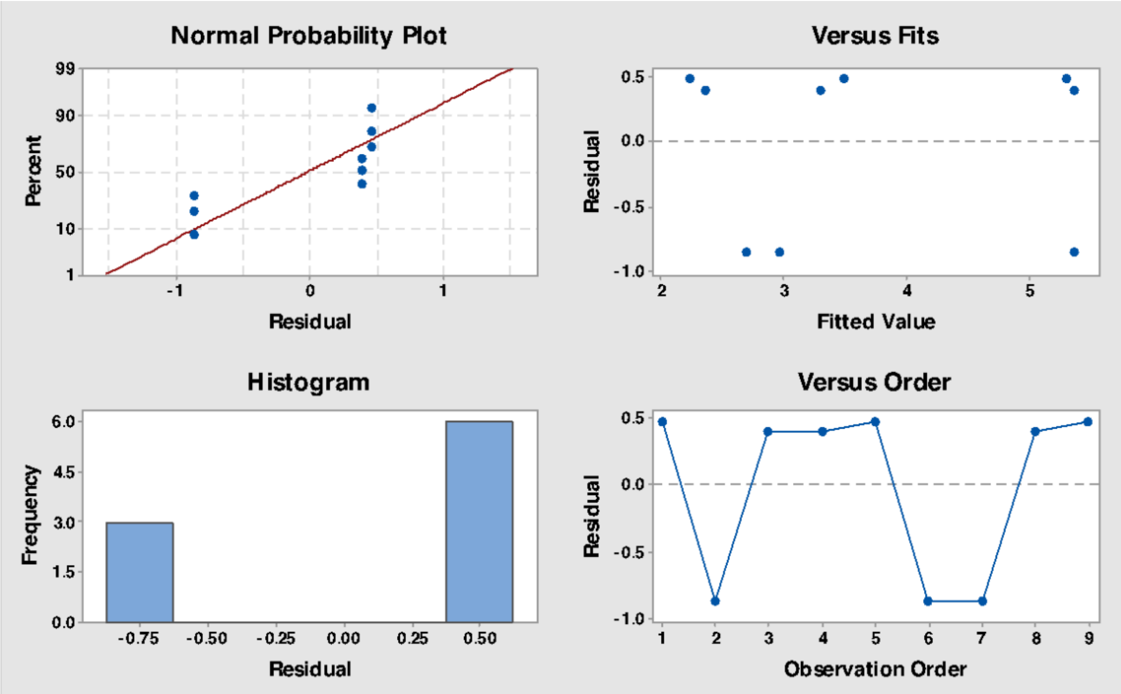

FIGURE 17 Residual plot for COF

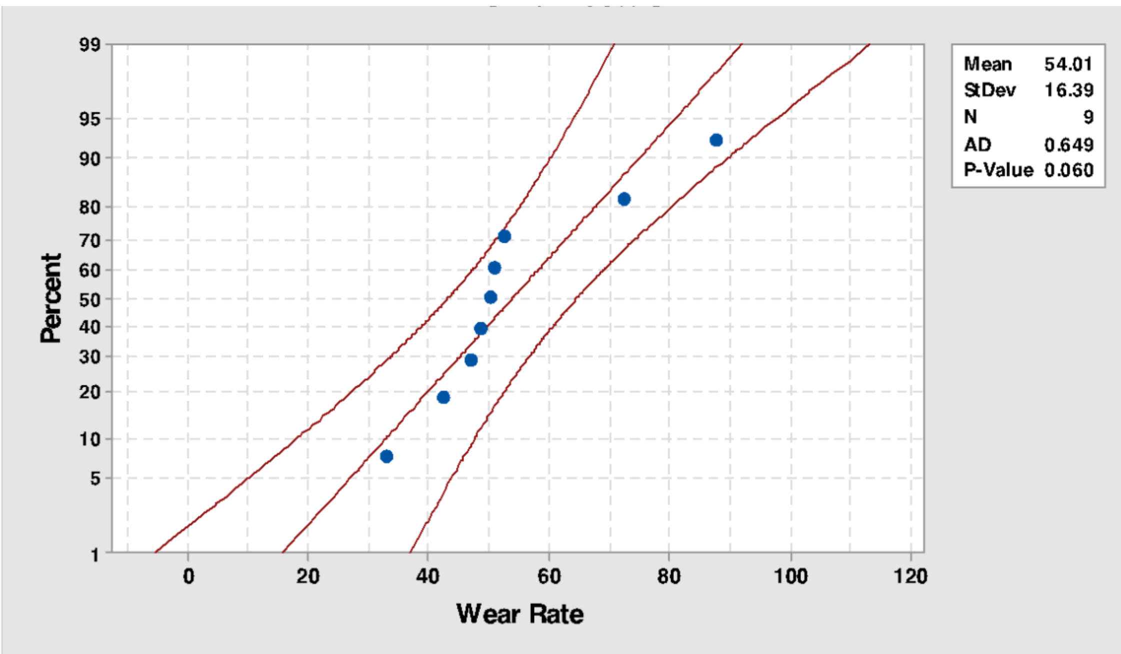

FIGURE 18 Probability plot for wear

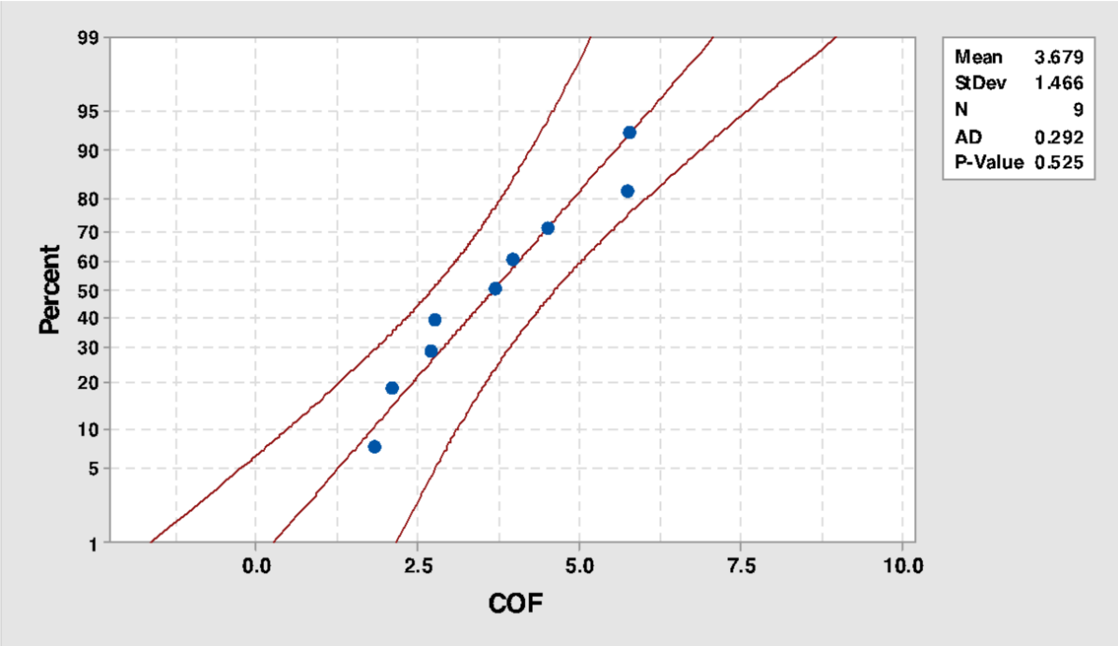

FIGURE 19 Probability plot for COF

3.5 Wear mechanisms and applications of the proposed PLSF-VE composite

The current study mainly focused on the practical aspects of friction and wear for the optimal selection of tribological testing conditions. From the aforementioned analysis, it was observed that load was the most influential factor that affected the wear of the composite specimens. The increment in filler content improved the wear resistance of the developed PLSF-VE composites. Also, the addition of PLSF fillers acted as a wear resistant agent and resisted the load during sliding. The wear mechanisms of the filler reinforced composites are expected to follow some standard wear mechanisms. Accordingly, the abrasive and adhesive wear mechanisms were evident predominantly between the filler and counter surface.

Furthermore, failures such as filler delamination, scratch, matrix plastic deformation and wear debris, due to the localized stress from thermal load led to the mass loss in the filler reinforced polymer composites, as similarly reported.[41,42] On the other hand, the deposition of polymers on the counter surface covered the asperity valleys, due to the melt flow of polymers. This self-lubricating phenomenon reduced the wear rate of the material as a result of the smooth wearing between the contact surfaces in case of a strong formation of the transfer layer. Mostly, polymer-based materials contain less filler content and experience matrix deformation during wear testing, which causes the adhesive wear. When the filler content was increased, the wear mechanism was abrasive in nature and wear resistance was improved. Agglomeration and bonding of fillers were the important factors to be considered for powder-filled composites. The presence of a good matrix-filler interfacial adhesion ensured the better wear resistance property of the composites.

Therefore, based on the tribological performance of the PLSF-VE composites, they can be used in the low temperature and loading applications, such as automotive clutches and brake pads, floorings and bearing pads at water environment, slideways in sliding doors, shoe soles and cooking vessels, including non-stick frying pans, among others.

4 CONCLUSIONS

The PLSF-VE composites were produced via compression molding technique and their tribological properties were analyzed. From the experimental and statistical analysis results obtained, the following conclusions are drawn.

- The homogeneous distribution of PLSF as a reinforcement particle in the VE matrix was evident from the SEM micrographs of the composite specimens.
- The process parameters for minimum WL were identified with filler content of 25 wt.%, at *P* and *N* values of 10 N and 300 rpm, respectively. In addition, the process parameters for minimum COF were obtained from filler content of 25 wt.%, at *P* and *N* values of 5 N and 700 rpm, respectively.
- From the ANOVA results obtained, *P* factor mostly influenced the WL and COF of the innovative PLSF-VE composites during POD wear testing, when compared with other considered factors. Therefore, engineering optimal tribological application of the different filler-reinforced composites should depend on their various responses to *P* and *N* factors.
- The PLSF-VE composites could be potentially used for automobile applications, such as body panels, lightweight seats, carpets and other interior parts, since these materials have good strength-to-density ratio, mechanical and wear properties. It can also be used for home appliances and sporting goods.

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CONFLICT OF INTEREST

There is no conflict of interest to declare.

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