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RESEARCH ARTICLE / ARAŞTIRMA MAKALESİ

A Design of Experiments-Based Investigation of Solid Lubricants in Heavy Vehicle Brake Pads

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Abstract

Brake pad formulations for heavy-duty vehicles must balance wear resistance, fade resistance, and friction stability under extreme braking conditions. This study systematically investigates the impact of solid lubricant compositions, including graphite, antimony trisulfide (Sb_2S_3), molybdenum disulfide (MoS_2), zinc sulfide (ZnS), and calcium fluoride (CaF_2), on the tribological performance of copper-free brake pads. A Design of Experiments (DOE) approach was employed to develop 29 unique formulations and evaluate them under the ECE R90 test procedure using a Krauss friction tester. Statistical analyses were used to determine the effects of lubricant combinations on performance metrics. The findings revealed that increasing CaF_2 content led to a significant rise in the coefficient of friction (up to 0.46), whereas Sb_2S_3 enhanced fade resistance by reducing loss of friction coefficient to as low as 0.03 but increased wear. MoS_2 was associated with superior thermal stability, while graphite primarily contributed to reduced wear loss (minimum 0.009 g). The formulation containing 10 wt% graphite, 2.5 wt% MoS_2, and 2.5 wt% CaF_2 demonstrated the most optimal balance of performance. These results underscore the importance of synergistic solid lubricant combinations in improving the durability and effectiveness of environmentally friendly brake pad formulations for heavy-duty applications.

Keywords: Brake pads, solid lubricants, tribology, friction, ANOVA

I. INTRODUCTION

Brake friction materials are essential for ensuring the safety and efficiency of heavy vehicle braking systems. Unlike passenger vehicles, heavy-duty vehicles operate under significantly higher loads, extreme braking conditions, and prolonged service durations. As a result, brake pads for heavy vehicles must exhibit high thermal resistance, minimal wear, stable friction coefficients, and excellent fade resistance to ensure long-term durability and safety in severe operating environments [1, 2]. The performance of disc brake systems in heavy commercial vehicles is determined by the friction and wear characteristics of the brake pad-disc interface, which are heavily influenced by the tribological properties of the friction material [3–5].

Friction materials in brake pads are complex composite systems typically formulated from more than ten different components, each tailored to fulfill a specific function in braking performance. These components generally include binders, reinforcing fibers, abrasives, fillers, and most critically, solid lubricants. Solid lubricants are particularly essential for reducing wear, stabilizing friction, and minimizing stick-slip behavior across a wide range of operating conditions[6–9]. In heavy-duty vehicles, where brake systems are exposed to elevated loads, frequent braking, and high thermal stress, the choice and optimization of solid lubricant combinations are vital [10]. Brake pads must maintain consistent friction behavior and minimal wear despite these harsh conditions and achieving high fade resistance, defined as the ability to retain braking performance at elevated temperatures, is a critical design requirement. Variations in braking response can be minimized by carefully selecting and optimizing solid lubricant compositions, as factors such as chemical structure, particle size, and phase transformations significantly influence tribological behavior. Recent studies indicate that synergistic combinations of metal sulfides and fluorides enhance braking stability by modifying wear mechanisms and regulating interfacial temperature fluctuations [11]. During breaking, the oxidation of metallic and carbon-based materials, along with a series of thermochemical reactions, leads to the formation of a tribofilm at the pad-disc interface [12]. This tribofilm plays a vital role in stabilizing friction, minimizing wear enhancing thermal stability and improving overall braking efficiency [9, 13, 14]. Its formation, thickness, and composition are influenced by the type and the amount of solid lubricants in the brake pad formulation [13]. Therefore, a well-balanced selection of solid lubricants is essential for optimizing the performance of heavy-duty brake pads [2, 13].

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Graphite is one of the most extensively used solid lubricants in friction materials due to its ability to provide consistent lubrication under both dry and wet conditions [15]. Studies have shown that graphite particle size and structure significantly influence braking performance, with finer particles promoting better dispersion and tribofilm formation, leading to more stable friction behavior [1, 16]. MoS_2 is widely utilized for friction stability, attributed to its layered structure that facilitates easy shear between contact surfaces and excellent thermal resistance [17, 18]. The particle size of MoS_2 has been found to influence tribological properties significantly, with larger particles exhibiting improved wear resistance and thermal endurance [18].

SnS_2 has been a preferred solid lubricant in brake pads due to its ability to form a stable boundary layer at high temperatures, reducing wear and preventing sudden variations in the coefficient of friction [6]. Studies suggest that combining Sb_2S_3 with MoS_2 and SnS_2 can significantly enhance fade resistance and durability while mitigating these environmental issues [9, 13]. Additionally, research indicates that dual and triple metal sulfide combinations provide superior tribofilm stability compared to single-lubricant formulations, leading to improved braking performance under extreme conditions [13, 19].

Calcium fluoride (CaF_2) is another widely studied solid lubricant, valued for its ability to improve wear resistance and thermal stability, particularly in high-load braking conditions [20]. Additionally, tungsten disulfide has been explored as an alternative solid lubricant, demonstrating superior thermal stability and tribofilm formation, which contributes to enhanced fade resistance, wear control, and recovery performance compared to SnS_2 [8]. Further investigations into graphite, coke, and ZnS-based formulations have highlighted the importance of solid lubricant ratios in tribofilm formation, showing that higher graphite content leads to more stable tribofilms and improved durability [11].

As environmental regulations evolve, heavy vehicle brake pad formulations must now comply with stricter standards while maintaining optimal performance. One such regulation is the Better Brakes Rule, enacted by the Washington State Department of Ecology in 2010 and implemented in phases starting from 2014 [21]. This legislation restricts the use of harmful substances such as copper, asbestos, and heavy metals in brake components to reduce environmental pollution caused by brake wear debris. The rule mandates the gradual elimination of copper and other toxic materials from friction materials, thus accelerating the shift towards safer and eco-friendly alternatives [22]. Thus, the alternative brake pads formulations require further investigation, particularly in terms of their ability to compensate for the loss of traditionally used materials while maintaining or enhancing tribological properties. In this regard, understanding the synergistic effects of different solid lubricants, their impact on tribofilm formation, and their long-term stability under extreme braking conditions is crucial for developing next-generation brake pads that meet both performance and regulatory requirements.

This study aims to systematically investigate the effects of different solid lubricant compositions on the tribological performance of copper-free, asbestos-free organic composite brake pads specifically formulated for heavy-duty vehicles. The formulation used in this study is based on a proprietary organic polymer matrix designed to comply with REACH directives and Better Brakes legislation. The main matrix is a non-metallic, fiber-reinforced composite system that excludes copper and asbestos entirely. The environmental impact of the selected lubricants was also considered. None of the additives used in this study are classified as Substances of Very High Concern (SVHC) under the REACH Regulation (EC) No 1907/2006 [23]. Sb_2S_3, although containing antimony, is used in minimal controlled amounts and in a chemically bound form within the matrix. ZnS and CaF_2 are widely used industrial lubricants with no known harmful emissions during operational use. By analyzing the interaction effects between graphite, Sb_2S_3, ZnS, MoS_2, and CaF_2, this research seeks to optimize solid lubricant formulations that provide superior braking performance under extreme operating conditions. Additionally, Analysis of Variance (ANOVA) based statistical modeling techniques are employed to quantify the significance of each solid lubricant and its interaction with others.

II. MATERIALS AND METHODS

2.1. Materials and Design of Experiments

The experimental approach employed a constrained full factorial Design of Experiments (DOE) framework to evaluate the effects of solid lubricant combinations on tribological properties. The base material matrix was a non-metallic, asbestos-free organic composite composed primarily of phenolic resin, reinforcing fibers, and proprietary fillers. This matrix was fixed at 85 wt% across all samples and represents a standard commercial-grade copper-free heavy-duty brake pad formulation. To systematically investigate the influence of solid lubricants, a mixed-level factorial array, with graphite was included in all formulations at two levels (5% and 10%), while other lubricants, Sb_2S_3, ZnS, MoS_2, and CaF_2, were examined at three levels (0%, 2.5%, and 5%). The total solid lubricant content was constrained to 15 wt% in every formulation, ensuring that all combinations adhered to this compositional limit. Based on this design logic, 29 unique brake pad compositions were generated to represent a constrained full factorial coverage of feasible solid lubricant mixtures. These combinations are detailed in Table 1.

Table 1. Design of Experiments (DOE) matrix for solid lubricant amounts in the brake pad formulations used in the study.

Sample #	Graphite (%)	Sb_2S_3 (%)	ZnS (%)	MoS_2 (%)	CaF_2 (%)
1	5	0	0	5	5
2	5	0	2.5	2.5	5
3	5	0	2.5	5	2.5
4	5	0	5	0	5
5	5	0	5	2.5	2.5
6	5	0	5	5	0
7	5	2.5	0	2.5	5
8	5	2.5	0	5	2.5
9	5	2.5	2.5	0	5
10	5	2.5	2.5	2.5	2.5
11	5	2.5	2.5	5	0
12	5	2.5	5	0	2.5
13	5	2.5	5	2.5	0
14	5	5	0	0	5
15	5	5	0	2.5	2.5
16	5	5	0	5	0
17	5	5	2.5	0	2.5
18	5	5	2.5	2.5	0
19	5	5	5	0	0
20	10	0	0	0	5
21	10	0	0	2.5	2.5
22	10	0	0	5	0
23	10	0	2.5	0	2.5
24	10	0	2.5	2.5	0
25	10	0	5	0	0
26	10	2.5	0	0	2.5
27	10	2.5	0	2.5	0
28	10	2.5	2.5	0	0
29	10	5	0	0	0

To characterize the properties of the investigated solid lubricants, various analytical techniques were employed. Particle size distribution (PSD) analysis was performed using the Malvern Panalytical Mastersizer 3000 to ensure precise measurement of lubricant particle dimensions. X-ray diffraction (XRD) analysis was conducted using the Malvern Panalytical X'Pert³ MRD to identify the crystalline phases present in the lubricants and their interaction within the composite matrix. Furthermore, thermogravimetric analysis (TGA) was carried out to assess the thermal stability and decomposition behavior of the lubricants. These characterization techniques provided comprehensive insights into the physical and thermal properties of the solid lubricants, aiding in the evaluation of their tribological performance.

2.2. Production of Samples

The 85 wt% base formulation for brake pads consisting of phenolic resin, reinforcing fibers, abrasives and fillers was prepared based on a proprietary recipe provided by Eren Balata INC specifically for heavy-duty commercial vehicle brake pads. The solid lubricant compositions were added to the base formulation according to Table 1. The main manufacturing steps and conditions can be summarized as below:

1. All raw materials were weighed and added into a mechanical mixer, where they were blended at 750 rpm for 14 minutes to ensure homogeneous distribution.
2. The blended mixture was transferred into a mold, where it was compacted using a hot hydraulic press at 165°C and 300 kgf/cm² for 5 minutes.

The compacted samples were post-cured in an industrial oven at 165°C for 8 hours to enhance bonding and mechanical properties.

The final sample size was approximately 85 mm × 52 mm × 13 mm. The samples were ground to achieve uniform thickness after curing.

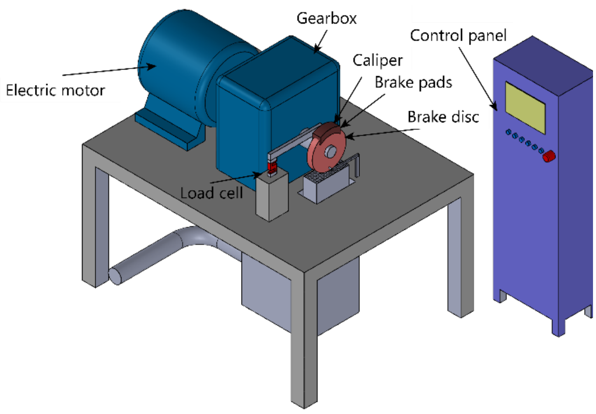

Figure 1. Schematic diagram of Krauss-type test setup used in this study. The device simulates dynamic braking events under controlled thermal and mechanical loading as outlined in ECE R90 Annex 9.

2.3. Tribological Characterization

The tribological performance of the developed friction materials was evaluated using a computer-controlled Krauss-type test machine, schematically illustrated in Figure 1. This setup simulates dynamic braking conditions under controlled thermal and mechanical loading. Testing was performed according to the ECE R90 regulation, specifically Annex 9, Section 3.2.2, which outlines standardized procedures for evaluating replacement brake linings [24]. The program began with an initial bedding phase consisting of 30 consecutive brake applications to stabilize the contact surfaces. This was followed by a sequence of 11 braking stages at varying initial and maximum disc temperatures to assess key performance indicators such as fade behavior, friction stability, and wear resistance under progressively severe conditions. Each stage was designed to replicate real-world thermal and mechanical stresses encountered during heavy vehicle operation, with or without active cooling. The full test schedule is detailed in Table 2.

Table 2. ECE R90 Annex 9 Section 3.2.2 brake testing sequence applied on Krauss-type test rig. Each stage simulates a defined thermal condition by specifying the number of brake applications, initial brake temperature, and target disc temperature. "Cooling" indicates whether active cooling was applied between applications.

Stage	No. of Brake App.	Initial Brake Temp. (°C)	Target Max. Disc Temp. (°C)	Cooling
1	5	100	300-350	✓
2	5	≤200	300-350	X
3	5	200	300-350	X
4	5	≤300	300-350	X
5	5	300	500-600	X
6	3	250	300-350	✓
7	3	200	300-350	✓
8	3	150	300-350	✓
9	10	100	300-350	✓
10	5	≤300	500-600	X
11	5	300	300-350	X

The friction coefficient and temperatures on the brakes were continuously monitored throughout the tests to evaluate the fade resistance and recovery characteristics of each material. To compare wear behavior of different compositions, the masses of individual brake pads were measured prior to and after each test.

2.4. Analysis

The experimental results were comparatively investigated though statistical approaches. A General Factorial Regression model was employed to establish the relationship between solid lubricant compositions and the tribological performance of the friction materials. This statistical approach was utilized to analyze the effects of solid lubricant type and amount on key response variables, namely friction coefficient, wear rate, and fade resistance.

The model was constructed using Minitab statistical software, incorporating main effects as well as selected two-way interactions. Analysis of Variance (ANOVA) was performed to quantify the contribution and significance of each input factor and their interactions. F-values and P-values were used to evaluate the statistical relevance of the parameters, while contribution percentages were calculated to understand their relative impact on performance. This approach enabled the identification of the most influential lubricant components for each tribological metric and provided insight into synergistic effects between additives.

Table 3. Characterization test results of solid lubricants

Lubricant	PSD	XRD		TGA
	d50 (μm)	Main component (%)		Decomposition Temperature (T ^{0}C)
Synthetic Graphite	453	C	100.0	750
Antimony Trisulfide	10	Sb_2S_3	100.0	750
Zinc Sulfide	7	ZnS	97.3	450
Molybdenum Disulfide	11	MoS_2	100.0	450
Calcium Floride	43	CaF_2	98.0	550

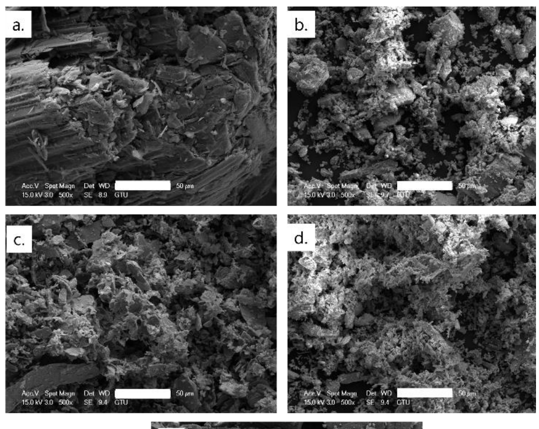

Figure 2. SEM micrographs of solid lubricants: (a) Graphite, (b) Sb_2S_3, (c) MoS_2, (d) ZnS, (e) CaF_2. The images depict morphology prior to blending into the brake pad formulation. (Scale bar corresponds to 50 μm)

III. RESULTS AND DISCUSSION

3.1. Materials Characterization

To evaluate the properties of the investigated solid lubricants, a series of characterization tests were performed. Particle size distribution, main chemical component of the lubricants obtained from X-ray diffraction (XRD), and decomposition temperatures extracted from thermogravimetric analysis (TGA) results for each solid lubricant are summarized in Table 4. In addition, the morphological characteristics of solid lubricants were examined using scanning electron microscopy (SEM). Figure 2 presents the SEM images of graphite, Sb_2S_3, MoS_2, ZnS, and CaF_2, respectively, providing insights into their microstructural properties and surface morphology.

3.2. Tribological Performance Evaluation

A key requirement for an effective friction material is maintaining a stable coefficient of friction regardless of variations in pressure, speed, torque, or temperature. The 29 formulated samples underwent a fixed-pressure test using the Krauss-type friction tester in accordance with ECE R90 Annex 9. The raw test data, including coefficient of friction and surface temperature responses, were automatically recorded and are presented in Figure 3. Figure 3a illustrates the evolution of the friction coefficient (μ) over successive braking cycles, while Figure 3b shows the corresponding brake disc temperature responses. The initial 30 brake applications represent the bedding period. While disc temperature profiles followed a similar trend due to consistent test settings, μ-values showed marked variation, indicating the strong influence of solid lubricant composition on friction behavior. And the surface temperature has a negative effect on the coefficient of friction.

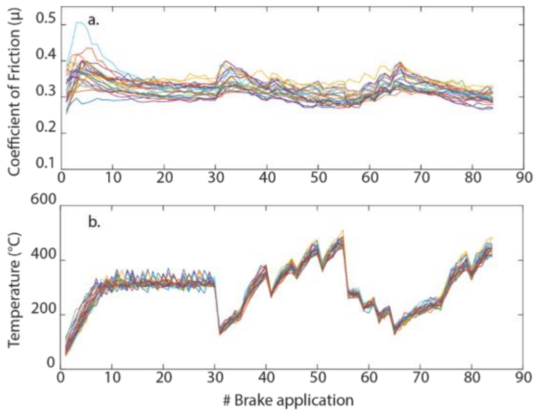

Figure 4. Evolution of (a) friction coefficient and (b) brake disc surface temperature as a function of sequential brake applications during the Krauss test of all tested specimens.

Based on the complete test data, three key performance indicators; fade resistance, average friction coefficient, and total wear rate, were extracted and summarized in Figure 4. Among all samples, the most favorable fade resistance was observed in samples 21, 29, 27, and 11. The highest coefficient of friction (0.46) was recorded in sample 14, while sample 21 demonstrated the lowest value (0.31). Wear rates varied substantially across samples, with sample 16 showing the highest wear and samples 21, 22, and 24 exhibiting the least. Overall, sample 21 (10 wt% graphite, 2.5 wt% MoS_2, and 2.5 wt% CaF_2) provided the most balanced tribological performance across all metrics.

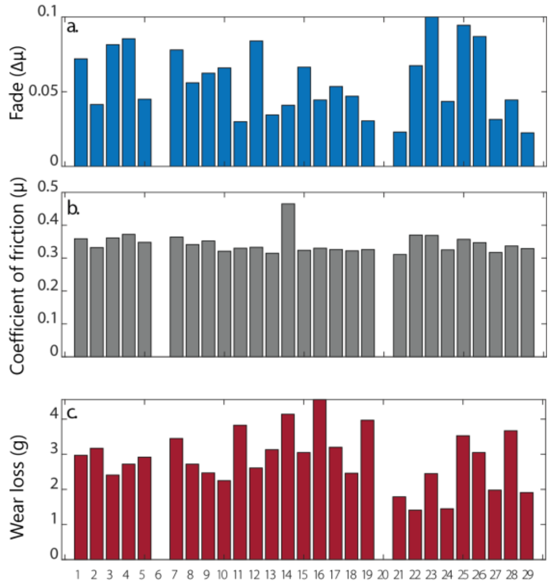

Figure 3. a. fade resistance, b. average friction coefficient, and c. wear amount obtained from the experiments.

It is worth noting that samples 6 and 20 exhibited inconsistent or unstable behaviors upon repeated testing and were thus excluded from final comparisons. These results suggest that specific combinations of solid lubricants not only affect fade resistance and wear but also induce complex interaction effects that dominate friction coefficient behavior—an insight further explored through ANOVA modeling in the next section.

3.3. Statistical Analysis

To determine the statistical significance of various factors affecting tribological performance, an analysis of variance (ANOVA) was conducted. Table 4 provides F and P-values and percentage contribution of each factor and interaction effect for fade resistance, wear amount, and friction coefficient. For simplicity, degree of freedom, sum of squares and mean squares values were removed from the table.

The ANOVA results suggest that in terms of friction coefficient, the statistical significance of individual factors was low, with MoS_2 (20.68%) and ZnS (19.94%) showing the highest contributions. However, the relatively high P-values indicate that these effects are not strongly significant, suggesting that the coefficient of friction is governed by a more complex set of interactions among the lubricants and is not governed by individual components alone. Graphite, despite its widespread usage, contributed only 0.73%, reinforcing the hypothesis that frictional stability results more from tribofilm integrity than isolated material effects.

In the analysis of fade resistance, MoS_2 and Sb_2S_3 emerged as the dominant contributors with 18.74% and 15.65% contributions respectively, each associated with near-significant P-values (0.053 and 0.067). These findings align with the established understanding of their thermal stability and effectiveness in supporting tribofilm development at elevated temperatures. The interaction between Sb_2S_3 and MoS_2 (19.59%) and other two-way combinations further confirms that fade resistance is highly dependent on the synergistic behavior of solid lubricants. In contrast, ZnS exhibited minimal contribution, indicating a more limited role in fade performance.

Table 4. Combined ANOVA results for tribological performance

Analysis of Variance	**Coefficient of Friction**			**Fade Resistance (Δμ)**			**Wear**		
SOURCE	*Contr..*	*F-Value*	*P-Value*	*Contr.*	*F-Value*	*P-Value*	*Contr.*	*F-Value*	*P-Value*
Model	79.21%	0.91	0.611	91.98%	2.73	0.134	91.39%	2.53	0.154
Linear	46.75%	1.42	0.36	37.24%	2.83	0.135	40.47%	4.02	0.073
Graphite	0.73%	1.26	0.313	0.01%	3.18	0.134	**20.49%**	1.52	0.272
Sb_2S_3	5.40%	2.54	0.173	15.65%	4.86	0.067	9.14%	1.97	0.234
MoS_2	**20.68%**	2.88	0.148	**18.74%**	5.59	0.053	8.68%	3.18	0.129
ZnS	**19.94%**	1.36	0.339	2.84%	0.79	0.502	2.17%	1.69	0.275
2-Way Interactions	32.46%	0.56	0.821	54.74%	2.44	0.166	50.92%	2.11	0.21
Graphite*Sb_2S_3	3.09%	0.91	0.46	6.22%	1.84	0.252	11.97%	4.86	0.067
Graphite*MoS_2	3.45%	0.2	0.823	**17.54%**	0.82	0.492	**19.08%**	2.26	0.2
Graphite*ZnS	6.40%	0.16	0.857	0.67%	0.12	0.893	2.15%	0.95	0.448
Sb_2S_3*MoS_2	5.17%	0.8	0.574	**19.59%**	3.38	0.107	**13.65%**	1.89	0.25
Sb_2S_3*ZnS	**14.34%**	0.86	0.544	10.73%	1.67	0.291	4.08%	0.59	0.684
Error	20.79%			8.02%			8.61%		
Total	100.00			100.00			100.00		

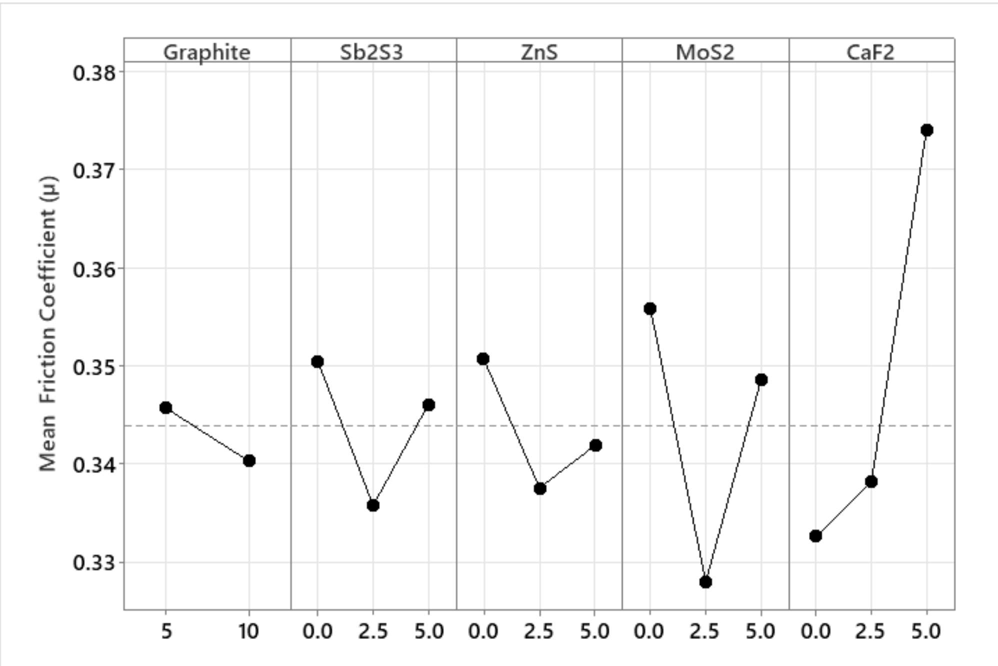

Figure 5. Main effects plot for average coefficient of friction

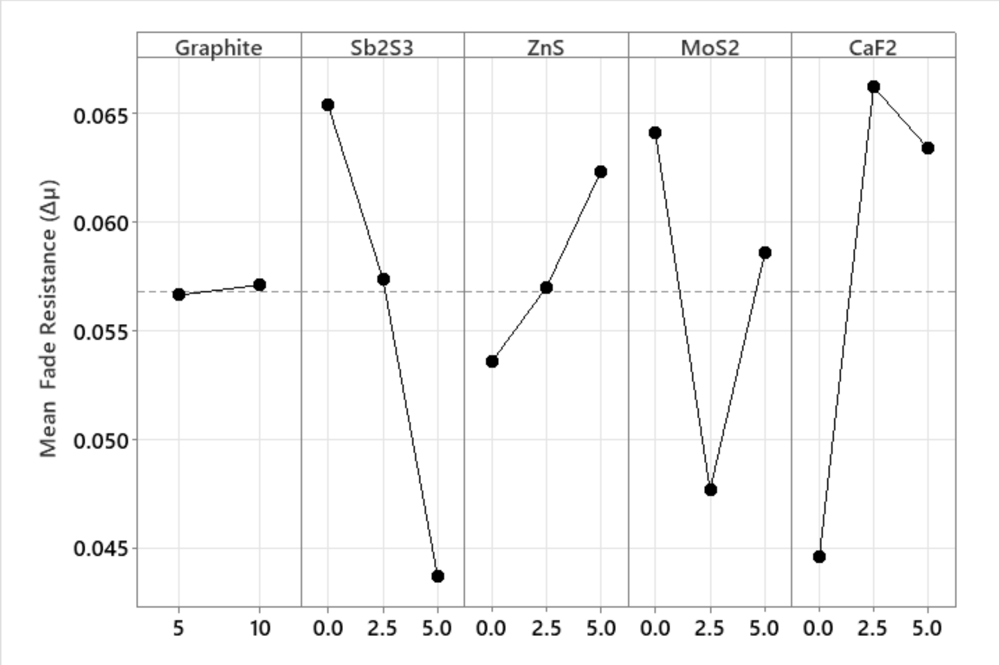

Figure 7. Main effects plot for fade resistance behavior

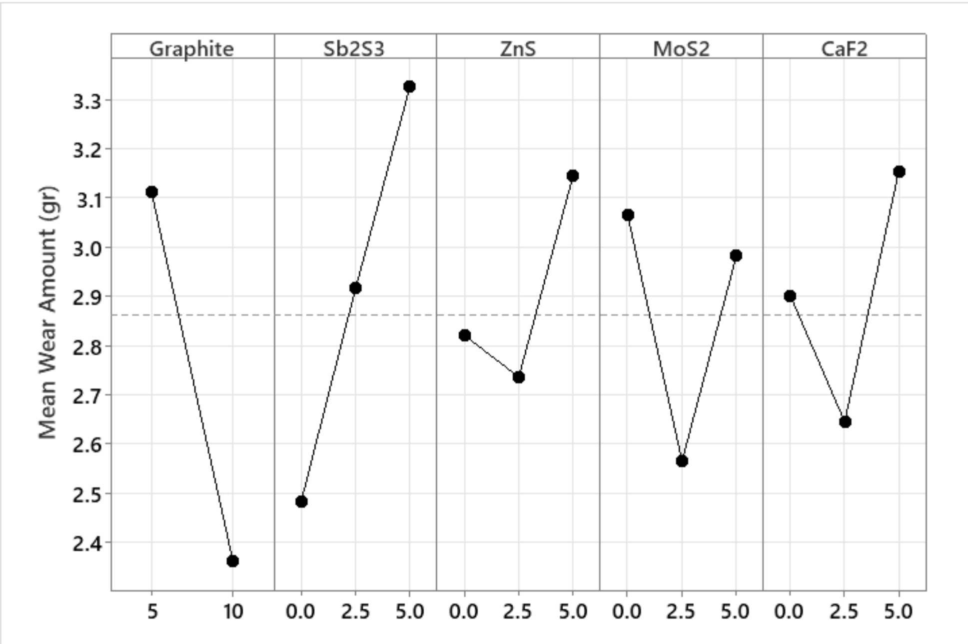

Figure 6. Main effects plot for wear amount.

Regarding the wear amount, graphite was found to be the most critical factor, contributing 20.49% to the model. These results suggest that graphite enhances load-bearing capacity and suppresses abrasive wear by promoting the formation of thermally stable tribofilms. The interactions of Graphite/MoS_2 (19.08%) and Sb_2S_3/MoS_2 (13.65%) further highlight how well-balanced combinations of solid lubricants can significantly improve wear performance. Sb_2S_3 also contributed meaningfully through its interaction with graphite (11.97%) but did not show a strong individual effect.

Figures 5 through 7 further elaborate on these statistical insights. Figure 5 illustrates CaF_2 content has a pronounced effect on the friction coefficient, where an increase in CaF_2 leads to a noticeable rise in friction levels. This finding aligns with its role in stabilizing the tribofilm under high-temperature conditions. While other lubricants such as MoS_2, Sb_2S_3, and ZnS exhibit non-linear behavior, this suggests that these lubricants contribute to tribofilm formation in a more complex, non-linear manner, dependent on their interactions with other components rather than their individual concentrations.

Figure 6 shows that an increase in Sb_2S_3 content improves fade resistance, meaning it helps maintain friction stability under high-temperature conditions. This result is consistent with literature findings, which indicate that Sb_2S_3 facilitates tribofilm formation at elevated temperatures, thereby preventing significant friction loss during prolonged braking applications [6]. Conversely, the addition of ZnS and CaF_2 appears to negatively impact fade resistance, reducing the system's ability to sustain friction levels over extended braking cycles. One possible explanation for this trend is that excess CaF_2 may lead to the formation of unstable tribofilm regions, resulting in discontinuous surface interactions. Such disruptions in the friction layer can cause inconsistent contact conditions, which, in turn, lower fade resistance.

Figure 7 shows that increasing graphite content leads to a reduction in wear amount suggesting that graphite contributes to a well-formed, thermally stable tribofilm, reducing direct surface interactions and minimizing abrasive wear. On the other hand, while Sb_2S_3 enhances fade resistance, its effect on wear behavior does not follow the same pattern. Unlike graphite, Sb_2S_3 does not consistently contribute to wear reduction, indicating that its primary function is to stabilize friction rather than directly mitigate wear. This suggests that while Sb_2S_3 strengthens the braking response at high temperatures, its interactions with other solid lubricants must be optimized to prevent excessive material loss.

The tribological performance of friction materials is inherently governed by complex interactions between mechanical, thermal, and chemical phenomena occurring at the pad-disc interface. Among these, the formation and evolution of tribofilms play a pivotal role in determining friction stability, fade resistance, and wear behavior.

The results from the ANOVA analysis and experimental evaluations reinforce the importance of not only selecting the right solid lubricant types but also formulating them in synergistic combinations. The statistical data clearly demonstrate that while individual effects of some lubricants may appear modest, their interaction effects are often substantial and sometimes dominant.

For example, graphite, traditionally recognized for its layered crystal structure and self-lubricating properties, exhibited the most consistent contribution to wear reduction. This aligns with its capacity to form stable, thermally resilient tribofilms that minimize abrasive contact [25]. However, graphite's effect on fade resistance and friction stability was less significant when considered in isolation. This suggests that graphite's benefit is primarily mechanical, buffering shear forces and suppressing wear rather than thermochemical in nature.

In contrast, Sb_2S_3 and MoS_2 emerged as critical to enhancing fade resistance. Both materials are known for their lubricating efficiency at elevated temperatures, and the ANOVA results confirm their role in preserving friction stability during thermal cycling. Importantly, the presence of these materials appears to facilitate the formation of dense, adherent tribofilms that resist degradation under cyclic thermal stress [26]. The interactions between Sb_2S_3 and graphite, and between Sb_2S_3 and MoS_2, were particularly influential in both fade and wear responses—highlighting that carefully balanced dual-lubricant strategies are more effective than single-lubricant additions.

The role of CaF_2 is more nuanced. While increasing its content led to a notable rise in the average coefficient of friction (as observed in the main effect plots), its effect on fade resistance was ambiguous. This behavior likely stems from CaF_2's tendency to contribute to friction-enhancing tribofilms at high temperatures, albeit at the risk of creating discontinuous or thermally unstable surface layers when used in excess [14]. These inconsistent tribofilm regions may be responsible for reduced fade performance in some compositions.

ZnS, while environmentally favorable, demonstrated minimal individual influence on fade or wear behavior in the statistical models. Its contribution appears to be more supportive, potentially enhancing tribofilm adhesion or complementing the effects of stronger lubricants like MoS_2 or Sb_2S_3 [11]. Its granular morphology and moderate thermal reactivity likely make it a passive yet beneficial filler rather than a performance driver.

From a systems-level perspective, the interplay between friction, fade, and wear performance points to a critical trade-off: compositions that resist fade often experience increased wear, particularly when tribofilms are thermally stable but abrasive [17]. However, formulations containing graphite appear to moderate this effect, achieving acceptable fade performance while substantially lowering wear rates. This moderation underscores graphite's versatility as a foundational solid lubricant in multi-component systems.

The observations are further corroborated by Figure 3 and Figure 4, where sample-specific behaviors across thermal cycles provide empirical support for the statistical findings. Sample 21 containing 10 wt% graphite, 2.5 wt% MoS_2, and 2.5 wt% CaF_2 emerged as the optimal composition, delivering low wear, high fade resistance, and moderate friction coefficient. This result highlights the effectiveness of combining mechanical lubricants (graphite) with thermally resilient additives (MoS_2, CaF_2) to achieve balanced tribological performance.

Finally, the inclusion of SEM micrographs (Figure 2) enhances the understanding of the role of solid lubricant morphology. The platelet structure of graphite, the angular grains of Sb_2S_3, and the layered features of MoS_2 each contribute to distinct tribofilm characteristics. Morphological attributes like particle size, shape, and surface texture influence dispersion, thermal contact, and film formation, all of which are critical to performance.

Together, the study confirms that optimal friction material performance cannot be achieved through the addition of a single solid lubricant. Instead, a multi-objective design approach, accounting for synergy, thermal stability, morphology, and interaction effects is essential. Future research should focus on mapping these interactions in higher resolution, including in-situ tribofilm analysis, to fully characterize the mechanisms underpinning brake pad performance.

IV. CONCLUSIONS

This study experimentally investigated the effects of solid lubricant combinations on the tribological performance of friction materials for heavy-duty brake applications. Graphite, antimony trisulfide (Sb_2S_3), molybdenum disulfide (MoS_2), zinc sulfide (ZnS), and calcium fluoride (CaF_2) were selected as solid lubricants, and their physical and chemical properties were characterized using Scanning Electron Microscopy (SEM), Particle Size Distribution (PSD), Thermogravimetric Analysis (TGA), and X-ray Diffraction (XRD) techniques.

A total of 29 different formulations were prepared by systematically varying the composition of solid lubricants while maintaining a fixed total lubricant content of 15 wt%, ensuring consistency in the base formulation. All samples were produced under identical manufacturing conditions to eliminate processing variations.

The tribological performance of the formulated friction materials was evaluated using the Krauss test machine, following the ECE R90 standard, which is widely used for assessing the braking behavior of commercial vehicle brake systems. The tests measured the relationship between temperature and friction coefficient, fade resistance, recovery performance, and wear rates of the brake pad formulations.

The experimental results revealed that:

1. Calcium fluoride (CaF_2) significantly increased the friction coefficient. Compositions with 5 wt% CaF_2 demonstrated average μ values up to 0.46, compared to lower values (~0.31) in samples without CaF_2. However, excessive CaF_2 led to reduced fade resistance due to tribofilm discontinuities.
2. Antimony trisulfide (Sb_2S_3) was most effective in enhancing fade resistance. Samples with 5 wt% Sb_2S_3 exhibited up to 37% improvement in Δμ compared to Sb_2S_3-free samples (Δμ improved from ~0.05 to 0.03). However, high Sb_2S_3 content also increased wear, with some samples showing wear losses up to 0.019 g.
3. Graphite showed the strongest influence on wear reduction. Increasing graphite from 5% to 10% decreased wear by approximately 35%, from 0.014 g to 0.009 g, without negatively affecting fade resistance or friction stability.
4. Molybdenum disulfide (MoS_2) contributed to both fade and wear improvement when combined with Sb_2S_3. The synergy between MoS_2 and Sb_2S_3 resulted in more stable tribofilms and enhanced performance, especially in mid-range compositions (2.5% each).

The best-performing sample (Sample 21), comprising 10 wt% graphite, 2.5 wt% MoS_2 and 2.5 wt% CaF_2, achieved the lowest wear loss (0.009 g), moderate average friction coefficient (μ = 0.31), and one of the best fade resistances (Δμ = 0.03), demonstrating a well-balanced tribological profile.

ANOVA analysis confirmed that MoS_2 (18.7%) and Sb_2S_3 (15.6%) had the highest contributions to fade resistance, while graphite (20.5%) was the dominant factor for wear reduction.

Overall, this study highlights the importance of strategic solid lubricant selection in developing high-performance copper-free friction materials for heavy-duty braking applications. The findings suggest that optimized multi-metal sulfide formulations offer a promising approach to enhancing brake pad performance by balancing wear resistance, fade resistance, and friction stability. Future studies should focus on long-term durability testing under real-world braking conditions and further optimization of lubricant compositions to meet evolving environmental regulations and performance demands.

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