



Study of Mechanical and Frictional Behaviour of the PDMS Elastomer

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ABSTRACT

Polydimethylsiloxane (PDMS) is a versatile elastomer widely used in biomedical devices, microfluidics, and sensors. To extend its applicability, this study investigates the influence of the base-to-curing agent ratio on its mechanical and tribological properties. PDMS samples with ratios of 20:1, 10:1, 5:1, 3:1, and 1:1 were analysed using nanoindentation. The results revealed that both elastic modulus and hardness increased from 20:1 to 5:1 and then decreased at higher curing agent concentrations (3:1 and 1:1). The effects of curing ratio and MoS₂ incorporation on adhesion and friction were further examined using linear reciprocating ball-on-disc tests against polypropylene (PP) and AISI 304 stainless steel (SS304) balls. Tests were performed under normal loads of 1-5 N at a constant sliding speed of 1 mm/s. For pristine PDMS, the coefficient of friction (CoF) followed the same trend as the mechanical properties, while MoS₂ addition, owing to its solid lubricating nature, reduced the CoF, by approximately 41% and 85 % with the PP ball and SS304 ball respectively.

KEYWORDS: Friction force, Adhesion force, characteristics of PDMS: elastomer, hardness, roughness.

Introduction;

Polymers are large molecules formed of many repeating units called monomers [1–3]. Polymers are widely used in engineering and scientific fields because of their adaptable properties and broad functionality [4–12]. Polymers are dream materials for engineers as its properties can be tuned using many different pathways like varying molecular weight [13,14], crosslinking [15,16], entanglements [17,18], branching [19], grafting [20,21], crystallinity [22], blending [23], embedding nanoparticles [24–26] are a few to name. Among polymers, polydimethylsiloxane (PDMS) has emerged as one of the most versatile silicone-based elastomers. It combines several desirable characteristics such as optical transparency [27], chemical stability [28,29], non-toxicity [30] [10], and mechanical flexibility [31] [11], which make it suitable for applications ranging from microfluidic devices [32–35], adhesives [36–43], wetting [44–46], patterning [47] to drug delivery systems [48,49].

One of the unique advantages of PDMS is that its mechanical performance can be tailored by adjusting the ratio between its base polymer and curing agent. Changing the ratio alters the crosslinking density, which in turn affects properties of polymers [50,51]. The stiffness, elasticity, and surface response of PDMS are strongly governed by its cross-link density. The standard Sylgard 184

formulation at 10:1 (base: curing agent by weight) typically shows an elastic modulus close to 1 MPa [52] [19]. Increasing the proportion of curing agent generally raises the cross-link density and thus increases stiffness; however, studies have revealed that beyond an optimal point, the modulus decreases, suggesting over-crosslinking or internal stress relaxation [53] [20]. The hardness of PDMS also changes with formulation. Flaminio et al. [54] reported that samples with different base-to-curing ratios displayed Shore A hardness values ranging from about 37 to 43, again with an optimal curing agent content showing maximum hardness, thus demonstrating the complex balance between elasticity and strength [55].

Similarly, interfacial properties as well as tribological behaviour, including friction and wear, is highly dependent on the mixing ratio as well as presence of nanoparticles [56]. Kim and co-workers observed that the elastic modulus, surface roughness, and frictional characteristics of PDMS surfaces vary noticeably when altering the curing concentration, highlighting the importance of reproducible synthesis protocols in tribological studies [53].

Adhesion force is critical for applications involving contact mechanics, soft robotics, and biomedical interfaces.

Modifying the base to curing agent ratio and processing parameters alters the elastic modulus of PDMS, which in turn affects its adhesive behaviour [41,42,57]. Additionally, variations in the surface roughness[58,59], as well as the incorporation of nanoparticles[60,61] into the PDMS matrix, can further influence the extent of adhesive interactions with counter surfaces [26].

Because of this tunability, PDMS is increasingly used in areas such as flexible electronics [62], strain and pressure sensors [63], and nano- or micro-electromechanical systems [64], where controlled stiffness and reliable surface interactions are critical. In addition, PDMS is extensively used in tribological applications involving soft contacts, such as biomedical implants, and soft robotic components [65,66]. It also serves as a model material for studying friction, adhesion, and lubrication in biological interfaces and engineered surfaces [67,68].

In this work, we systematically examine how the variation of the base-to-curing agent ratio affects Young's modulus, hardness, adhesion force and the coefficient of friction of PDMS. The results provide insight into optimizing PDMS formulations for applications that require a balance of mechanical robustness and favourable tribological properties.

EXPERIMENTAL DETAILS

PDMS Elastomer

SYLGARD 184 Silicone Elastomer was used to prepare the PDMS elastomer. Sylgard 184 comes as a kit having polymer base and curing agent. Schematic representation of the PDMS elastomer preparation is illustrated in Figure 1. To prepare the sample, required amount of PDMS polymer base is poured into a beaker.

The height of the sample is decided to be 2 mm so accordingly the amount of PDMS base is taken. The weight of polymer base is measured using an electronic weighing balance. The weighing balance is a Wensar Electronic Balance having least count of 0.1 mg. Keeping the beaker still on the weighing balance, the curing agent is poured drop wise into the container using a pipette until the desired amount is reached. To ease the cross-linking process, the mixture is stirred continuously for 15-20 minutes using a thin glass rod. The beaker is next kept on a vacuum desiccator for 15-20 minutes to remove the trapped air in it.

Once all the trapped air has escaped, the desired amount of mixture is poured into a clean acrylic mould. The mould is also kept in the desiccator for 20-30 minutes to remove the trapped gas. Meanwhile the oven is preheated to temperature of 120°C. Also, the surface of oven is checked for flatness using a 3 Vial Spirit Level to avoid the difference of height in the sample. Once the temperature of oven has reached the desired temperature and all

the gas has escaped from mould, it is kept inside the oven. The sample is kept in the oven for 120 minutes. After the sample has cured in the oven, it is carefully removed from oven and kept in a flat surface until it reaches room temperature. When the sample has cooled down completely, it is carefully de-moulded using a scalpel and tweezers. The sample is then cut into pieces of dimensions as required for the test using a sharp cutter, and remaining sample is kept in a clean plastic container to avoid contamination.

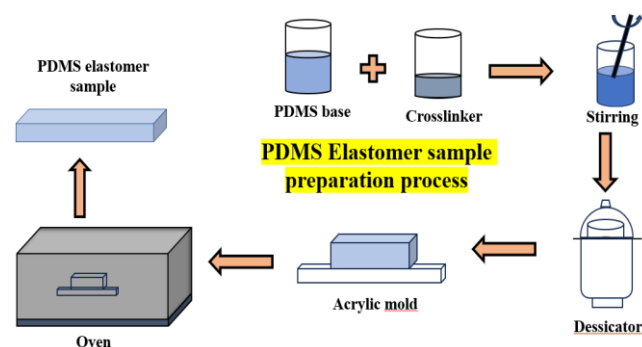


Figure 1: Schematic representation of the PDMS elastomer preparation procedure.

Nano particle reinforced PDMS

Dry blending does not result in uniform dispersion of nano particle in PDMS, therefore organic solvent method is used here. The solvent being used is toluene.

First MoS₂ was weighed using an electronic balance (about 1 wt.% of PDMS polymer) which was later dissolved in toluene solvent weighing about three times that of MoS₂ for better dissolution. The mixture was then stirred with a mechanical stirrer (at 500 rpm) for 1 hour at room temperature.

Similarly, PDMS base is mixed in toluene solvent with a mechanical stirrer (at 500 rpm) for 1 hour at room temperature. Post stirring both the solution are mixed and stirred with a mechanical stirrer (at 500 rpm) for 2 hours at 70°C. This ensures that all the toluene has evaporated from the solution and it contains only MoS₂ dispersed in the PDMS base.

After complete evaporation of toluene, curing agent is added to the solution and stirred using a glass rod for 15-20 minutes to accelerate cross linking. The beaker is next kept in a vacuum desiccator to remove all the entrapped air.

The solution is then poured carefully in a dry acrylic mould, in the desired amount, and mould with the solution inside is kept in vacuum desiccator for 15-20 minutes to remove the air. After removal of air, the mould is placed inside a vacuum oven, already preheated to 120°C, and baked at 120°C for 120 minutes.

Methodology

Tribological test

Dry sliding friction tests were performed using a pin-on-disc linear reciprocating tribometer (Rtec MFT-2000) under ambient conditions, at a room temperature of 25°C and relative humidity of 50%. A ball was used as the pin, resulting in a ball-on-flat contact configuration. PP and SS304 balls were used as the countersurface material. The graphical representation of the experimental set up is illustrated in Figure 2. The sample size was taken as 2cm × 2cm × 0.2cm. The load was varied from 1N, 2N, 3N to 5N. The average sliding speed was kept at 1mm/s and the total duration for each test was 20 minutes. Each test was repeated thrice.

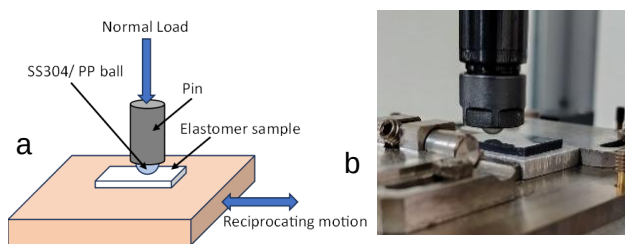


Figure 2: (a) Schematic of the linear reciprocating pin-on-disc setup and (b) a zoomed view of the tribometer showing the mounted sample and ball.

Nanoindentation test

Nanoindentation tests for Young's Modulus and Hardness were performed in Hysitron TI 750d nano-indenter. The indenter tip used was Berkovich Tip. All the samples of the different mixing ratios were tested at same depth of indentation. The sample size was taken as 1cm × 1cm × 0.2cm. Each test was repeated thrice. The machine provided reduced elastic modulus which can be converted to elastic modulus of PDMS using the formula:

$$\frac{1}{E_r} = \frac{(1-\nu_{PDMS}^2)}{E_{PDMS}} + \frac{(1-\nu_i^2)}{E_i} \quad (1)$$

where,

E_{PDMS} , ν_{PDMS} : Elastic modulus and Poisson's ratio of the material.

E_i , ν_i : Elastic modulus and Poisson's ratio of the indenter.

The value of Young's Modulus and Poisson's ratio for Diamond indenter is 1141 GPa and 0.07 respectively. Since the value of Modulus for diamond tip is very large compared to that of PDMS [69][33], therefore the second term in equation 1 can be neglected. The equation hence becomes

$$E_{PDMS} = (1 - \nu_{PDMS}^2) E_r \quad (2)$$

The value of Poisson's ratio for PDMS can be taken as 0.5 [70][34] so the equation further reduces to

$$E_{PDMS} = 0.75 * E_r \quad (3)$$

RESULT & DISCUSSION

Mechanical properties

The variation in Young's modulus (E) with different PDMS mixing ratios, as determined from the nanoindentation tests is presented in Figure 3.

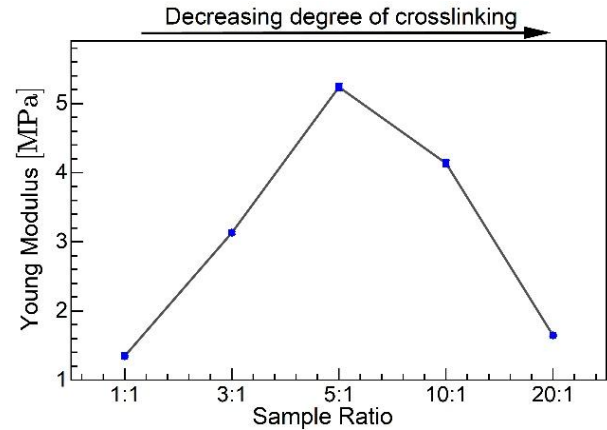


Figure 3: The plot shows the variation of Young's modulus (MPa) as a function of the PDMS base-to-curing-agent mixing ratio (sample ratio). Five different mixing ratios of PDMS, namely 1:1, 3:1, 5:1, 10:1, and 20:1, were prepared to evaluate the Young's modulus using nanoindentation measurements performed with a Berkovich indenter tip.

For each base-to-curing agent mixing ratio, three measurements were taken, and the average Young's modulus was calculated. The results, shown in Figure 3, indicate that Young's modulus increases as the ratio of base to curing agent increases from 1:1 to 5:1, reaching a maximum at 5:1. Beyond this point, as the base content continues to increase (i.e., from 5:1 to 20:1), Young's modulus decreases due to the corresponding reduction in curing agent content.

These findings are consistent with the observations of Susheel et al. [24], who also reported a peak in Young's modulus at the 5:1 mixing ratio, followed by a decline at higher base-to-curing agent ratios.

Further, as the amount of curing agent was increased from ratio 5:1 to 1:1, the Young's modulus decreased. This result is consistent with the finding of Kim et al [53]. It therefore suggests that any excess of curing agent make the elastomer less flexible.

Figure 4 illustrates the variation in hardness of PDMS as a function of the base-to-curing agent mixing ratio.

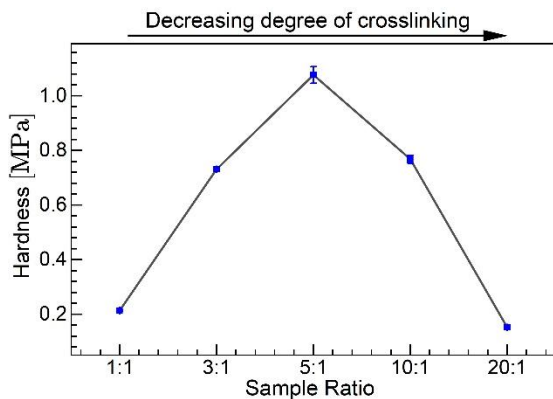


Figure 4: The plot presents the variation in hardness (MPa) as a function of the PDMS base to curing agent mixing ratio.

A trend similar to that obtained in case of Young's Modulus was found. This finding is similar to that of Lan et al. [55] for relationship between hardness and elastic modulus. Furthermore, It is observed that although the compositions (3:1 and 10:1) differ considerably in their mixing ratios, they exhibit similar hardness (Figure 4). It is possible as both can achieve comparable crosslinked network structures. Once the network reaches a certain crosslink density, further addition of the curing agent has little effect on the mechanical properties. In the 3:1 ratio, an excess of curing agent may lead to incomplete network formation, whereas in the 10:1 ratio, strong chain entanglement compensates for the lower crosslink density [54]. Further, at higher ratio of base and curing agent, more amount of polymer base is left non reacted with curing agent, making the elastomer softer and less hard [71].

CoF and adhesive force variation for plain PDMS

Figure 5 illustrates the variation in sliding frictional force with applied normal load using a PP ball as the counter-surface. To establish a trend, linear regression analysis was performed to determine the correlation between the variables.

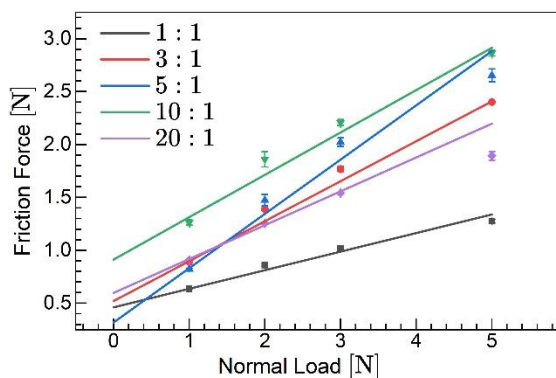


Figure 5: The plot shows the variation of friction force with normal load for PDMS samples prepared

at different base-to-curing agent ratios, when tested against a PP ball countersurface. Linear regression lines are fitted to the experimental data for each mixing ratio to determine the slope, which represents the CoF.

The CoF was subsequently calculated as the slope of the fitted line, plotted in Figure 6.

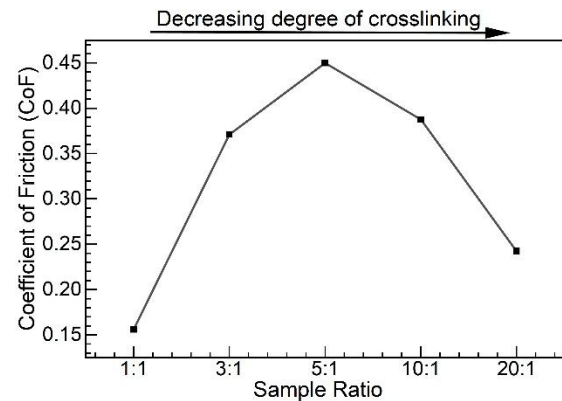


Figure 6: The plot presents the relationship between the CoF and the base-to-curing agent mixing ratio for PDMS elastomer samples, when tested against PP ball as the countersurface.

Figure 7 similarly shows variation of sliding frictional force with normal load utilizing stainless steel 304 ball. It is observed that at lower loads, the friction force for all mixing ratios is similar. While at the higher normal load i.e., 4-5 N, the difference in friction force is significant. At lower loads, the real contact area and deformation are limited, resulting in comparable frictional forces across all samples. As the load increases, the contact area and asperity interactions become more pronounced, leading to enhanced deformation, adhesion, and interfacial shear and hence, distinct frictional responses. This behaviour can be explained by observation reported by Deng, W., Kesari, H., [72] which shows that hysteretic energy loss in soft adhesive contacts increases with indentation depth (i.e., applied load). Therefore, higher loads induce disproportionately greater energy dissipation. Since the extent of hysteresis depends on the elastic modulus and viscoelasticity of PDMS, samples with different mixing ratios exhibit distinct frictional behaviour at higher loads.

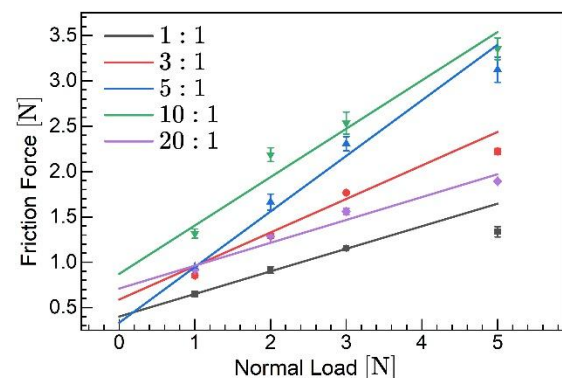


Figure 7: The plot depicts the relationship between friction force and normal load for PDMS samples prepared with different base-to-curing agent mixing ratios, when tested against SS304 ball as the countersurface.

The CoF observed for different base-to-curing agent ratios of PDMS, when tested using SS 304 ball as countersurface is plotted in Figure 8.

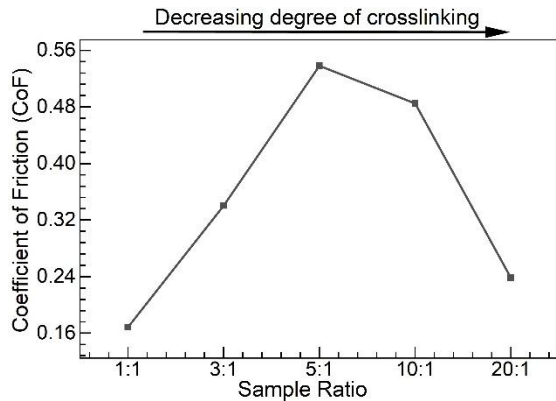


Figure 8: The plot shows the variation of the CoF with the base-to-curing agent mixing ratio for PDMS elastomer samples, using an SS304 ball as the countersurface.

The 1:1 sample was shown to have the lowest CoF value compared to any other sample, while the 5: 1 sample showed the highest CoF value compared to other samples. Also, in most of the formulations, PDMS-SS304 tribopair showed higher friction than PDMS-polypropylene pair.

The CoF increases as the base-to-curing-agent ratio decreases from 20:1 to 5:1, reaching a maximum at 5:1. With further decrease in the ratio from 5:1 to 1:1, the CoF decreases.

This non-monotonic trend can be explained by three regimes: (i) under-cross-linked PDMS (20:1) is unable to sustain high interfacial shear; (ii) an optimal cross-link density (5:1) maximizes shear strength and (iii) excess curing agent leaves unreacted oligomers that form a lubricating boundary layer. As the amount of curing agent mixed in the base polymer increased, a decrease in CoF is observed. With an increase in the amount of the curing agent, there becomes an excess of curing agent such that further no cross linking occurs. The excess curing agent is thus available as free chains that can result in adhesion. When measuring friction stress in a polymer, both adhesive forces (how well the surface sticks) and elastic deformation (how much the material deforms) contribute. Even if a polymer has a low adhesive contact, its elastic properties can compensate and still allow efficient stress transfer during friction. In case of excess curing agent, more free chains are available, which initially improve adhesion. However, these free chains create a weak

boundary layer with low shear resistance. This layer behaves like a viscous lubricant, reducing the transmission of stress to the polymers network. If a material efficiently transfers stress into its bulk structure, more energy is dissipated, increasing resistance to motion. This results in a higher friction force F_f corresponding to the applied normal load F_n , which increases the coefficient of friction:

$$CoF = \frac{F_f}{F_n}$$

Further it was found that the value of CoF was generally higher for PDMS-SS304 pair than PDMS-PP pair. It could be attributed to higher surface energy of metals and alloy compared to polymers [73][37]. Metals have higher surface energy, and a higher surface energy generally leads to easier adhesion because it indicates greater molecular attraction between materials, facilitating bonding and thus increasing the CoF.

As shown in Figures 5 and 7, when the linear fit of the friction force versus normal load curve is extrapolated to the load axis ($N = 0$), the intercept on the friction force axis corresponds to the adhesive force, $F_{adhesion}$. The total friction force can therefore be expressed as [74] [38]:

$$F = \mu N + F_{adhesion}$$

Where, F is the total friction force, μ is the friction coefficient, N is the normal force, and $F_{adhesion}$ is the adhesive force.

$F_{adhesion}$ represents the adhesive force component that arises from molecular and interfacial interactions at the contact. This contribution is independent of the externally applied normal load and reflects the inherent tendency of the surfaces to adhere, even in the absence of external pressure. Physically, it corresponds to the finite amount of frictional resistance present at zero applied load, which may originate from van der Waals forces, capillary interactions, chemical bonding, or other adhesive mechanisms active at the interface.

The values of adhesive force, determined as the intercept from the extrapolated friction-load curves, are plotted in Figure 9 and 10 for both PP and SS304 counter surfaces respectively.

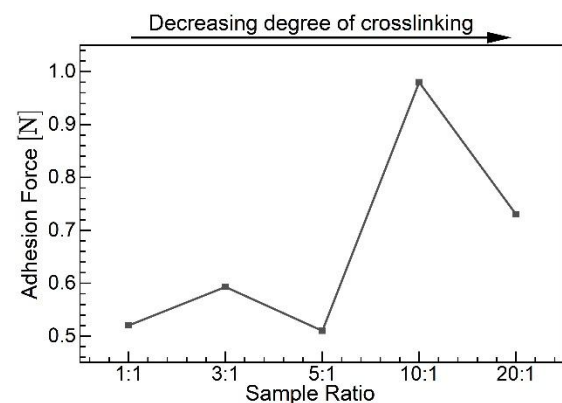


Figure 9: The plot illustrates the variation of adhesion force with the base-to-curing agent mixing ratio for PDMS elastomer samples, when tribologically

tested against a PP ball as the countersurface. Each adhesion value corresponds to a specific mixing ratio and was obtained from the intercepts of the linear fits in the friction force versus normal load plots shown in Fig. 5, where the intercept at zero normal load represents the adhesion force.

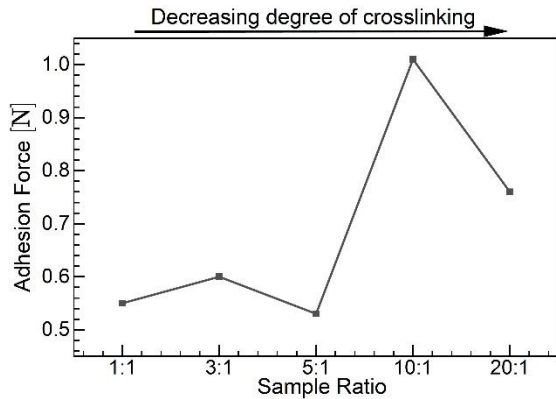


Figure 10: This plot shows the variation of adhesion force with the base-to-curing agent mixing ratio for PDMS elastomer samples tested against an SS304 ball as the countersurface. The adhesion force for each mixing ratio was derived from the friction force versus normal load data presented in Fig. 7, where the intercept of the linear regression at zero normal load corresponds to the adhesion force.

The results indicate that the PDMS sample with a 10:1 base-to-curing-agent ratio exhibited the maximum adhesive force. When the curing agent content was reduced (20:1 ratio), the adhesive force decreased, suggesting that insufficient cross-link density lowered cohesive strength and adhesion. Similarly, increasing the curing agent content (5:1 ratio) also resulted in a reduction of adhesive force, attributable to excessive crosslinking and increased stiffness. Interestingly, a slight increase in adhesion was observed when the ratio was adjusted from 5:1 to 3:1, although a further decrease was recorded at the extreme 1:1 composition. The non-monotonic trend suggests an interplay between cross-link density, residual surface chain mobility, and mechanical stiffness. As seen in Figure 3, the Young's modulus and hardness (Figure 4) is maximum at 5:1 ratio, suggesting stiffer elastomer. Excessive stiffness reduces the ability of the polymer surface to conform to counter-surface asperities, thereby lowering the real contact area and diminishing adhesion. This agrees with studies showing that higher cross-link density correlates with reduced tack and adhesion strength due to the limited mobility of polymer chains at the interface [75][39]. The slight increase in adhesion observed at 3:1 compared to 5:1 may be attributed to the reduced stiffness and enhanced chain mobility at the polymer surface, which increase conformability and intermolecular interactions with the counter surface. This suggests that at 3:1, PDMS forms a marginally larger real contact area than at 5:1.

CoF and adhesive force variation for MoS₂ reinforced PDMS

Tribological testing of MoS₂ reinforced PDMS revealed a lower CoF compared to unmodified PDMS. The tested formulation consisted of a base-to-curing-agent-to-MoS₂ ratio of 10:1:1, with PP balls employed as the countersurface under reciprocating conditions. The variation of friction force with applied normal load for this configuration is illustrated in Figure 11.

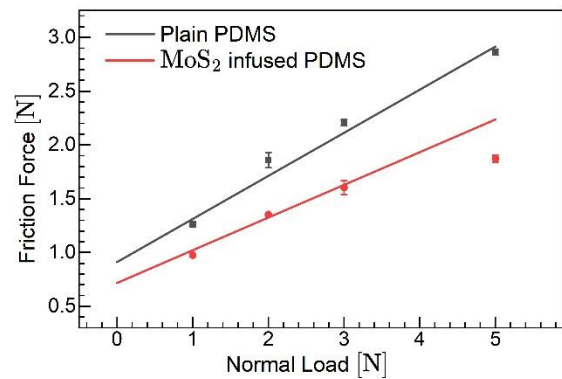


Figure 11: The plot illustrates the relationship between friction force (N) and normal load (N) for plain PDMS and MoS₂-infused PDMS elastomer samples, measured using a PP ball as the countersurface. Linear regression was applied to the experimental data to determine the slope, which represents the CoF.

Similarly, experiments were conducted using SS304 balls, and the corresponding results are presented in Figure 12.

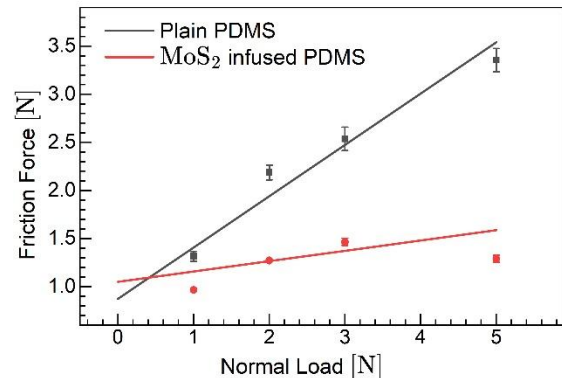


Figure 12: The figure presents the variation of friction force (N) with normal load (N) for plain PDMS and MoS₂-incorporated PDMS elastomer samples, tested using an SS304 ball as the countersurface.

As expected, addition of nanoparticles in PDMS resulted in decrease of CoF. Load is concentrated on rough surface asperities, leading to high local stress points, increased friction, and wear. Nanoparticles fill in surface roughness and asperities, creating a smoother contact

area and strengthening the matrix. They spread the applied load across a larger area, reducing contact pressure at any single point. They distribute stress more evenly, preventing high-pressure points that increase friction.

Further strong nanoparticle-matrix interactions allow stress to transfer efficiently from the matrix to the nanoparticles. This prevents crack initiation and micro-tearing in the composite, which would otherwise increase friction.

Similar to the procedure adopted for plain PDMS, the friction force versus normal load curves of MoS₂-reinforced PDMS were extrapolated to zero applied load to determine the adhesive force. The calculated adhesive force values for the reinforced samples are presented in Table A, alongside those of plain PDMS, to enable a direct comparison of adhesion behaviour.

Table A: Adhesive force values obtained from extrapolated friction–load curves for plain PDMS and MoS₂-reinforced PDMS samples.

Polymer	Adhesive force (N) with PP ball as countersurface	Adhesive force (N) with SS304 ball as countersurface
Plain PDMS (10:1)	0.982	1.014
MoS ₂ infused PDMS (10:1:1)	0.856	1.051

CONCLUSION

The variation of Coefficient of Friction, adhesion force, Young's modulus and hardness was studied by varying the mixing ratio of PDMS elastomer base and curing agent and also with addition of MoS₂ nanoparticle.

The CoF increases up to a base-to-curing-agent ratio of 5:1, beyond which it decreases with further addition of the curing agent. The CoF is also significantly influenced by the countersurface material. PDMS in contact with SS 304 exhibits higher friction compared to contact with PP. The adhesive force obtained from the intercept of the friction–load curves however, exhibited a non-monotonic trend. It increased with curing-agent content from 20:1 to a maximum at 10:1, then decreased at 5:1. A slight increase was observed at 3:1, followed by a further reduction at 1:1.

The MoS₂ reinforced PDMS composite demonstrated lower friction across all tested conditions compared to unmodified PDMS.

The value of hardness shows a trend similar to inverted bell curve corresponding to different amount of curing agent added. Hardness first increase as value of curing agent is increased up to 5:1 then decreasing as more curing agent is added. The curve of Young's Modulus Modulus showed trend similar to hardness curve. As the mixing ratio of base and curing agent is changed from 20:1 to 5:1, the value of Young's modulus gradually increases. But, after 5:1 as the amount of curing agent is increased, elastic modulus decreases steeply.

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