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Full Length Article

Crystallinity dependent tribological behavior of molybdenum disulfide: Insights from experiments and atomistic simulations

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ABSTRACT

The frictional and wear performance of molybdenum disulfide (MoS_2) is strongly governed by its crystallinity, yet this influence remains largely unexplored. In this study, we present the first integrated experimental and atomistic study that establishes how structural order governs the tribological performance of MoS_2. A wide spectrum of crystallinity, from amorphous to highly crystalline states, is investigated to evaluate its influence on the coefficient of friction (COF) and wear resistance. Experiments reveal a strong dependence of COF and wear on crystallinity, with crystalline MoS_2 achieving ultralow friction and enhanced wear resistance. In contrast, amorphous and polycrystalline films display significantly higher COF and pronounced tribo-polishing effect. Complementary reactive molecular dynamics simulations capture the same trend and uncover the underlying mechanics: crystalline MoS_2 maintains smooth sliding interfaces and enables frictional anisotropy through interlayer shear. However, sliding of disordered structures increases surface roughness, energy dissipation, and material removal. Furthermore, we examine the wear mechanisms under high normal loads, demonstrating that crystallinity enhances wear resistance by mitigating material deformation. The strong agreement between experiments and simulations confirms crystallinity as a dominant factor governing the tribological response of MoS_2. Our findings provide atomic-scale insights into the mechanisms of superlubricity in layered materials and offer a fundamental basis for designing advanced solid lubricants and wear-resistant coatings.

1. Introduction

Transition metal dichalcogenides (TMDs) have shown exceptional mechanical [1], electronic [2], and tribological properties [3], making them suitable for a wide range of applications, including solid lubricants, energy storage devices, and nanoelectronics [4,5]. Molybdenum disulfide (MoS_2), a two-dimensional TMD, has attracted significant attention due to its unique combination of low friction [6,7], high wear resistance [8], and thermal stability [9,10]. These properties have led to the usage of MoS_2 in sectors such as aerospace and automotive industries [11,12]. However, its performance significantly depends on crystallinity, which dictates the arrangement of atoms and layers within the material [13,14].

Several experimental studies on MoS_2 have reported excellent mechanical properties, including elastic modulus [15], bending modulus [16] and fracture toughness [14]. The exceptional tribological performance of MoS_2 is attributed to its layered structure, where weak van der Waals interactions between layers enable easy shear [17]. This leads to low friction and superlubricity under specific conditions [6,18]. Superlubricity, characterized by friction coefficients below 0.01 [19,20], occurs when the sliding surfaces are atomically flat, leading to incommensurate contacts that minimize frictional forces [6]. However, the presence of humidity [21], oxygen [22], variations in external temperature [23], and degree of crystallinity [13,14] govern the frictional and wear performance of MoS_2. Khare & Burris showed that the friction in MoS_2 increases with the presence of oxygen [24]. The negative influence of oxygen on the tribological properties has been attributed to the formation of molybdenum oxides on the sliding surface [24,25]. Several experimental studies have also reported an increase in friction due to the presence of water [21,26,27]. John F. et al. studied the influence of microstructure on the oxidation and friction behavior of MoS_2 [28]. Recently, Liu et al. fabricated different crystal structures of MoS_2 by altering the grain boundaries [13]. Their study showed that a higher degree of crystallinity is beneficial for oxidation resistance and longevity of MoS_2. The wear study conducted by Hesam et al.

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on amorphous and polycrystalline (PC) structures revealed that the amorphous form exhibits wear resistance four times greater than that of its crystalline counterpart [29].

Although the effects of humidity and oxygen on the tribological properties have been extensively studied, the influence of varying degrees of crystallinity on its frictional and wear behavior remains poorly understood. A key challenge lies in the difficulty of fabricating MoS_2 with precise control over crystallinity [30,31], which is essential for a consistent comparison of friction and wear behavior. Moreover, experimental techniques often face limitations in capturing the atomic-scale mechanisms governing friction and wear. Equally, experimental techniques alone cannot fully resolve the atomic-scale mechanisms governing frictional dissipation and material removal, necessitating complementary atomistic simulations.

With the continuous advancement in computational power, researchers are increasingly utilizing computational techniques to study the mechanics of materials. Various computational methods, including molecular dynamics (MD) [32,33], coarse-grained simulations [34], finite element methods [35], and multiscale modeling, are employed to study material behavior across different length and time scales [36,37]. Among these approaches, MD simulations have proven to be particularly powerful in exploring atomic-scale phenomena, providing insights into fundamental mechanisms such as fracture, crack propagation, friction, and wear [38,39]. Earlier MD simulations on MoS_2 have focused on estimating key mechanical properties including elastic modulus [40], ultimate strain [41], and fracture toughness [42]. For instance, Bao et al. investigated the crack propagation mechanism for single layer MoS_2 using atomistic simulations and proposed modifications to the Griffith criterion [43]. Additionally, MD simulations have been extensively used to study the influence of grain boundaries and defects on the mechanical properties and failure pattern in PC MoS_2 [31,44,45]. Recently, we employed MD simulations to study MoS_2 with varying degrees of crystallinity, demonstrating the dependence of fracture toughness and failure mechanisms on crystallinity [46].

In the context of tribology, significant progress has been made in understanding the friction and wear mechanisms of MoS_2 using MD simulations. For instance, Hu et al. studied the friction mechanism in multilayer MoS_2 under variable loads and shearing velocities [47]. Their results highlighted the irreversible deformation caused due to heavy load and shear velocity. Wei et al. explored the influence of grain boundary defects on the wear resistance through scratch simulations [48]. Other studies have examined the impact of interlayer spacing and temperature on frictional forces during sliding [49], as well as the effects of oxygen content in altering the tribological properties of MoS_2 [50]. Notably, Serpini et al. performed sliding simulation of ordered and disordered MoS_2, uncovering several nanoscale mechanisms for amorphous and crystalline configurations [51]. However, their study compared only two cases — fully ordered and fully disordered MoS_2 — without examining intermediate crystallinity levels or wear phenomena. Additionally, studies have reported superlubricity in crystalline MoS_2 and detailed its nanoscale frictional behavior [52,53]. Recently, research has focused on the abrasive wear due to sliding and rotating of a rigid body on crystalline MoS_2 [54]. Furthermore, previous studies have explored the influence of normal load and temperature on the wear mechanism during reciprocating friction [55]. However, despite these advancements, the influence of crystallinity on the frictional and wear properties of MoS_2 remains largely unexplored, highlighting a critical gap in the understanding of its tribological behavior. Furthermore, no prior study has systematically combined experimental measurements and MD simulations to establish a direct correlation between crystallinity and the tribological response of MoS_2. Such an integrated approach is essential to connect nanoscale mechanisms with macroscopic observations and to validate simulation insights against real surfaces.

In this study, we bridge this gap by performing a combined experimental and atomistic investigation of MoS_2 with controlled degrees of crystallinity — ranging from amorphous to highly crystalline structures — to unravel how structural order dictates friction and wear. Experiments are conducted to measure the COF and wear under nanoscale loading conditions. Complementary reactive MD simulations provide a mechanistic understanding of the experimental trends, revealing the atomic-scale origins of frictional anisotropy, surface roughness evolution, and deformation processes. Friction and wear mechanisms are simulated by sliding a rigid carbon body over MoS_2. Unlike previous studies, which primarily examined either crystalline or amorphous forms in isolation, the present work systematically explores the full spectrum of crystallinity and establishes a direct structure–property relationship. The integrated insights demonstrate that crystallinity is the governing parameter controlling the transition from high-friction, wear-dominated behavior to ultralow-friction in MoS_2.

2. Materials and methods

In this section, we describe the experimental methods used to create MoS_2 with varying degrees of crystallinity, along with the details of the MD simulations.

2.1. Sample preparation

MoS_2 samples with varying degrees of crystallinity were prepared to investigate their tribological performance. All samples were prepared on Si (100) wafers and their thicknesses were measured using a Zygo 7200 profilometer. A crystalline multilayer MoS_2 sample was obtained by mechanical exfoliation from a commercial bulk MoS_2 crystal. Thin films with varying crystallinities were synthesized by magnetron sputtering under different deposition conditions. An amorphous MoS_2 film was deposited by direct current magnetron sputtering from an MoS_2 target at room temperature in an Argon (Ar) atmosphere (5 mTorr ≈ 0.67 Pa) using a power of 150 W. A polycrystalline MoS_2 film was produced by reactive co- sputtering of two Mo targets (High-power impulse magnetron sputtering (HiPIMS) + ratio frequency (RF)) at 500 °C in an Ar/H_2S mixture. During this deposition, the HiPIMS source operated at 208 W with a pulse frequency of 189 Hz, while the RF source was maintained at 167 W. The working pressure was 1.1 Pa, with gas flow rates of Ar at 20 sccm and H_2S at 5 sccm. The crystallinity of the samples were confirmed by Raman spectroscopy (Fig. 1), which clearly distinguishes the crystalline, PC, and amorphous states of MoS_2. Experimental synthesis does not permit precise fabrication of intermediate crystallinity levels; therefore, only three possible structures were examined.

2.2. Nanotribological tests

Nanotribological measurements were performed using a scanning probe microscope operated in contact mode under ambient conditions. Load-dependent nanofriction tests were conducted over an area of 10 μm × 10 μm with normal load incrementally increased from 20 to 120 nN. Nanowear tests were carried out over 2 μm × 2 μm regions under a normal load of 600 nN for five consecutive scans. Sharp uncoated probes (240AC-NA, OPUS, k = 1.7 N/m) were used for all nanotribological measurements. To ensure consistency, all friction tests were performed with a single probe, while new probes were used for nanowear tests. The lateral and normal spring constants for the cantilever were calibrated following the procedures Sader & Green [56,57]. The calibration of lateral and normal force sensitivity are performed using the beam-geometry method [58,59]. All measurements were performed under an ISO Class 5 clean room conditions with constant temperature (about 25 °C) and humidity (RH ≈ 40%). At these low loads and micron-scale contact areas, tribo-oxidation of MoS_2 is expected to be minimal, as also reported in earlier studies [53,60].

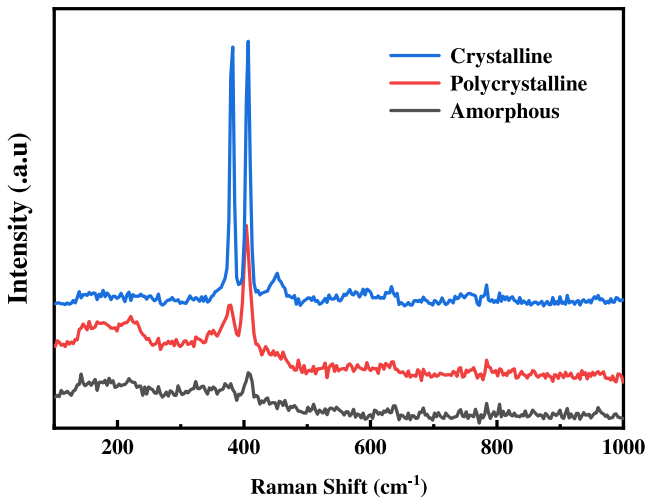


Fig. 1. Raman spectrums of MoS_2 samples with different crystallinity.

2.3. MD simulations

We modeled MoS_2 in various configurations, including crystalline, PC, and amorphous structures. Unlike experiments, the degree of crystallinity can be controlled in MD simulations. Thus, PC models were created with 75%, 25%, and 10% crystallinity, denoted as PC75, PC25, and PC10, respectively. The initial configurations were constructed by randomly placing molybdenum (Mo) and sulfur (S) atoms within a simulation box of dimensions $63 \times 40 \times 35.5$ $Å^3$. To achieve the desired PC structures, a sequence of annealing and quenching processes were employed, allowing control over the percentage of crystallinity. Detailed information regarding the annealing temperatures and the resulting crystallinity percentages are provided in Fig. 2. While PC10, PC25, and PC75 were obtained using the annealing and quenching parameters listed in Fig. 2, further adjustment of the quenching rate or annealing temperature can yield additional intermediate crystallinity levels. For a better understanding, the atomic configuration of all PC structures are placed adjacent to the plots in the figure. The perfect crystal structure with six layers is created by orthogonalizing and replicating the unit cell in X and Y directions. Here, the stable 2H phase of MoS_2 is chosen, where each Mo atom is covalently bonded to six S atoms, and each S atom is covalently bonded to three Mo atom [61]. All models were designed to maintain a constant total number of atoms. The equilibrated structure of PC25 is shown in Fig. 3(a). The red and blue colored balls represent Mo and S atoms. Following equilibration, the simulation cell is extended in the Z direction to 75 Å to accommodate a rigid body. A spherical rigid body with a diameter of 20 Å, composed of 1431 atoms is positioned on top of the MoS_2 surface. The atomic configuration of the MoS_2 model with the rigid body is illustrated in Fig. 3(b). The density of crystalline and PC MoS_2 after equilibration is obtained as 5.0 and 4.8 g/cc, which is in accordance with the values observed in experimental studies [62]. The strategy adopted to measure degree of crystallinity is given in detail in Appendix A.

All MD simulations are performed using the LAMMPS open-source package [63], and the post-processing of results are performed using OVITO [64] and MATLAB script. Atomic interactions were modeled using the reactive force field (ReaxFF) [65], implemented via the reax/c user package in LAMMPS [66]. The ReaxFF parameters employed in this study were specifically developed to accurately capture the interactions in Mo, S and carbon systems [67]. These parameters have been validated to reproduce the energetics of both crystalline Mo-S phases and amorphous MoS_2, making them well-suited for simulating the crystallization of MoS_2. While previous studies have utilized the reactive empirical bond order (REBO) potential to model atomic interactions [31], our prior work has demonstrated its limitations [68]. Specifically, the REBO potential fails to predict the enthalpies of formation for Mo-S crystalline phases other than MoS_2 and inaccurately estimates the relative energies of amorphous MoS_2 compared to its crystalline counterpart [46,68]. These shortcomings underscore the superiority of ReaxFF parameters used for this study, justifying its selection over REBO.

The simulation setup involves placing the rigid body on top of the MoS_2 surface, as illustrated in Fig. 3(b). Periodic boundary conditions are applied in the X and Y directions, while a non-periodic boundary condition is used in the Z direction. A normal load is applied in the Z direction, and a constant horizontal sliding velocity of 10^{-4} Å/s is imposed in the X direction. Five different normal loads — 22, 43, 72, 78, and 143 nN — are considered to investigate the load-dependent frictional behavior. Loads above 143 nN were not used for COF evaluation because they initiate material removal in MoS_2, causing the measured friction to be influenced by wear rather than interfacial sliding. To ensure stability during sliding, the bottom portion of the MoS_2 model, consisting of approximately 900 atoms (gray colored balls in Fig. 3(b)) are constrained in all directions. The movement of the rigid body is controlled using the *fix rigid* command in LAMMPS. The system is maintained at a constant temperature of 300 K using the Nosé - Hoover thermostat (NVT ensemble). Each sliding simulation is performed for 1 ns with a timestep of 0.5 fs. To study wear mechanisms, a higher normal load of 358 nN is applied while keeping the horizontal sliding velocity unchanged. Normal loads higher than 500 nN were not considered in this study, as it can cause the rigid body to completely penetrate and disrupt the MoS_2 layers. Despite the nN load magnitude, the extremely small contact area produces GPa-scale contact pressures, comparable to those in practical applications.

3. Results and discussion

In this section, we present and discuss the key findings from our study, focusing on the influence of crystallinity on the frictional and wear behavior of MoS_2. First, the results from experimental studies are provided. Second, we investigate the underlying friction and wear mechanisms using MD simulations. Finally, a comparison between experiments and simulations are given in detail.

3.1. Experimental results

3.1.1. Nanofriction

The topologies and friction maps of MoS_2 with different crystallinity are shown in Fig. 4. A typical cluster like morphology is observed in MoS_2 with PC structures. We observed a root mean square (RMS) surface roughness of 16.2 nm for amorphous MoS_2. However, a higher RMS value of 60.3 nm is obtained for PC MoS_2, which is different from the results provided in the study by Krauss et al. [69]. This variation is caused due to the difference in the deposition method. On the other hand, crystalline MoS_2 shows a smooth surface with a low RMS value of 0.4 nm. The exfoliation process results in the formation of a few defects on the surface of MoS_2. However, these defects have a negligible influence on the friction (see friction map in Fig. 4).

The load dependent nanofriction results from our study are presented in Fig. 5, where normal load is plotted with respect to frictional load. The COF values were determined by performing a linear fit to the friction–load plots. It is observed that the COF is strongly influenced by the degree of crystallinity. We obtained a COF value of 0.035 ± 0.007, 0.52 ± 0.15, and 0.63 ± 0.02 for crystalline, PC, and amorphous MoS_2 respectively. It is to be noted that the trend in COF values observed in this study is different from the observations made by Hesam et al. [29]. This is due to the significant difference in sliding scale, normal load, sliding speed, and dimension of specimen. Further, Hesam et al. [29] investigated only sputtered amorphous and polycrystalline MoS_2, and their work focused on groove-forming scratching/plowing wear.

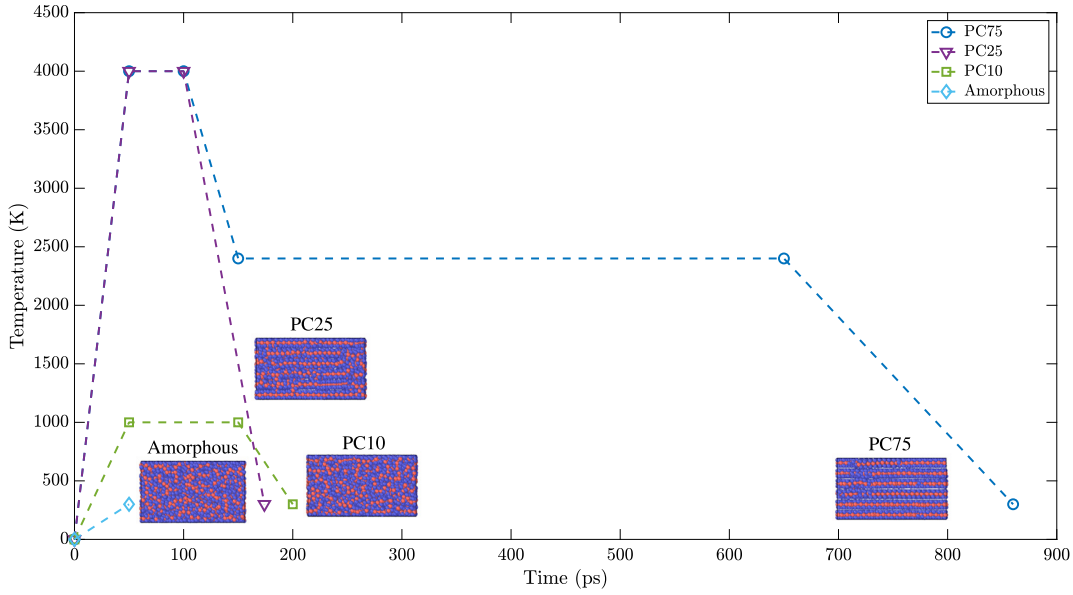
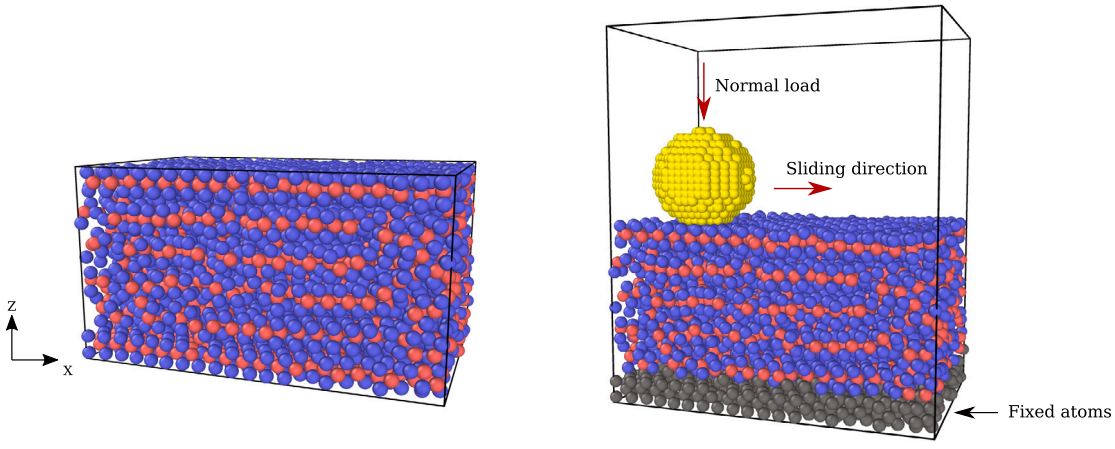


Fig. 2. Sequence of annealing and quenching temperatures used to create polycrystalline models with different percentages of crystals.



(a) PC25 after equilibration (b) PC25 with rigid body

Fig. 3. Atomic configuration of PC25, rigid body and imposed boundary conditions for sliding simulation.

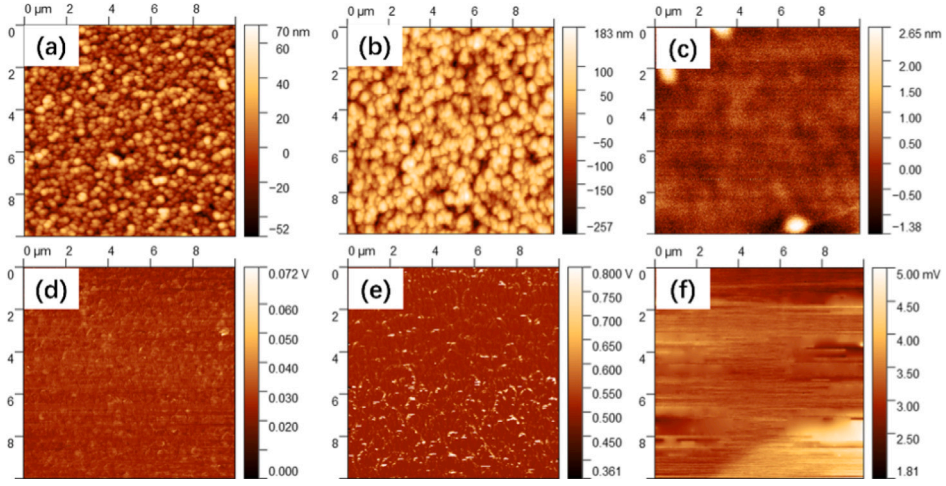
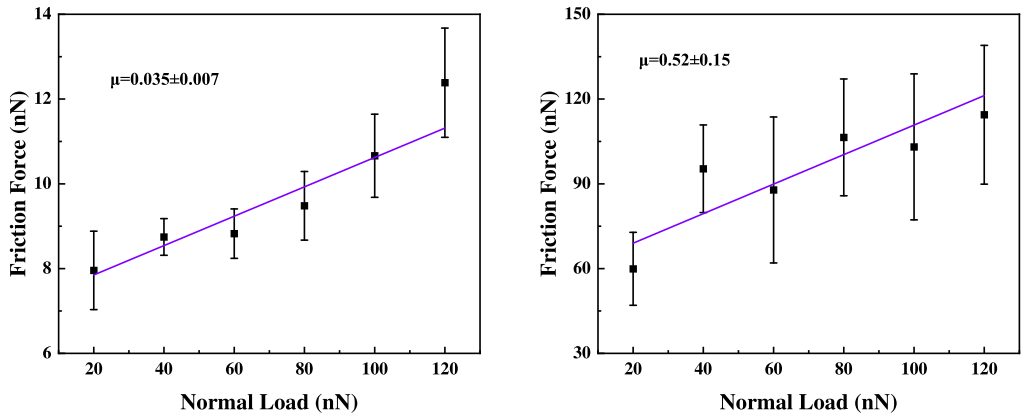
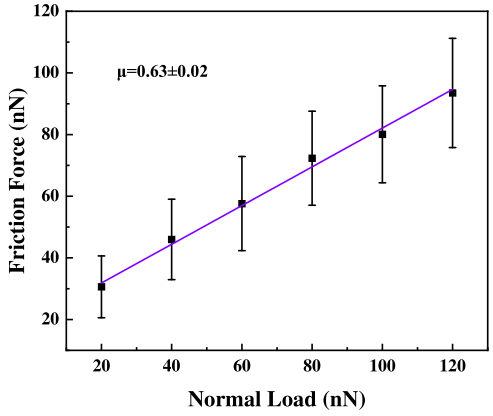


Fig. 4. Topologies of (a) amorphous, (b) polycrystalline, and (c) crystalline MoS_2. The images from (d)–(e) corresponds to friction map.



(a) Crystalline (b) Polycrystalline



(c) Amorphous

Fig. 5. Load dependent nanofriction results for MoS_2 with different crystallinity.

3.1.2. Nanowear

Nano-scale wear tests on MoS_2 were performed by repeatedly scanning a 2 μm × 2 μm area under a normal load of 600 nN for five cycles. As shown in Fig. 6, both amorphous and PC films exhibited a clear tribo-polishing effect. The surface roughness progressively decreased with cycle number, indicating material transfer and removal. Consistent with this polishing, the friction force decreased during successive sliding cycles for both films. Friction maps acquired before and after testing (Figs. B.2 & B.3) in a 4 μm × 4 μm region support this interpretation. In contrast to previous studies reporting low-friction flakes embedded within wear scars [58], no such features were observed in our experiments. The crystalline MoS_2 sample showed no discernible change within the wear track throughout the nano-wear test. The RMS roughness remained on the order of 0.1 nm at all scan cycles (Fig. 6), and no recognizable wear within the test zone was observed. The friction force on crystalline MoS_2 remained low during scanning, below 10 nN. The suppressed wear and low friction at this load are consistent with MoS_2 interlayer sliding during the test, as shown in Fig. 7. A post-test defect was occasionally observed out of the test zone, which is originated from interlayer shearing. Even though the AFM results shown in Figs. B.2 & B.3 do not allow for quantifying the wear resistance, it can be concluded that crystalline MoS_2 provides a better resistance to wear. Quantitative wear depth could not be reliably measured because the intrinsic surface roughness of the amorphous and polycrystalline films introduces height variations comparable to the wear-induced changes, making depth extraction experimentally unreliable.

Table 1
Coefficient of friction for MoS_2 with different degrees of crystallinity.

System	Coefficient of friction
Crystalline	0.00014 ± 0.00005
PC75	0.0505 ± 0.0266
PC25	0.1048 ± 0.0235
PC10	0.1691 ± 0.0450
Amorphous	0.2736 ± 0.0911

3.2. MD results

Molecular dynamics simulations are performed on MoS_2 with different crystallinity to determine its frictional and wear performance.

3.2.1. Coefficient of friction

The COF is determined from the sliding simulations conducted at varying normal loads. For each applied normal load, the corresponding frictional forces are measured. To ensure the reliability of results, three independent MD runs are performed and the frictional forces are calculated as the average of these runs. The results are presented in Fig. 8, where the frictional force is plotted with respect to the normal load. The error bars in the plots indicate the standard deviation across the three runs. The COF is then computed from the slope of the linear fit to the data, as illustrated in Fig. 8. The calculated values of COF with corresponding error bounds are presented in Table 1.

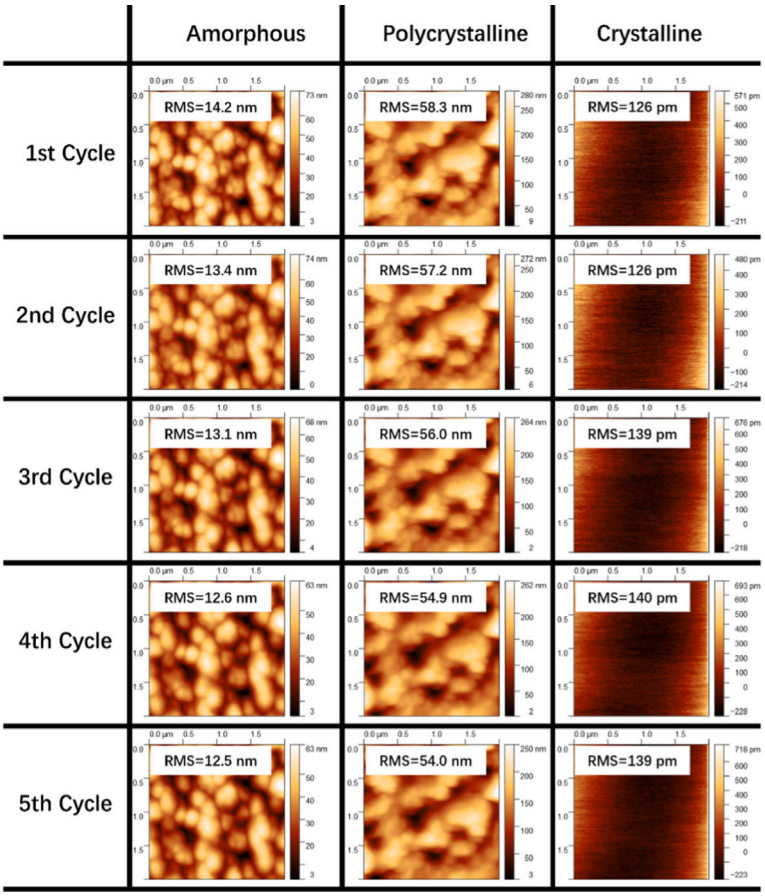


Fig. 6. Surface evolution of MoS_2 samples with different crystallinity in nanowear tests.

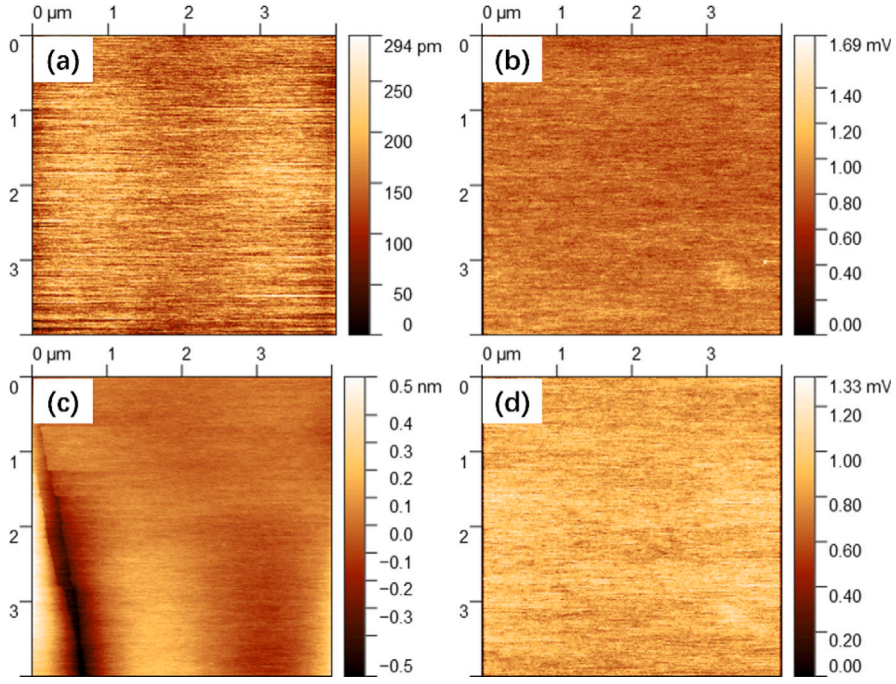


Fig. 7. Topographies and friction map of crystalline MoS_2, (a & b) before nanowear test, (c & d) after nanowear test.

The computed values of COF summarized in Table 1 reveals a clear dependence on the degree of crystallinity in MoS_2. The lowest and highest values are observed for crystalline and amorphous MoS_2 respectively. We obtained a COF value of 0.00014 for crystalline MoS_2, which falls in the category of superlubricity [18]. This result aligns with previous experimental and computational studies that have reported

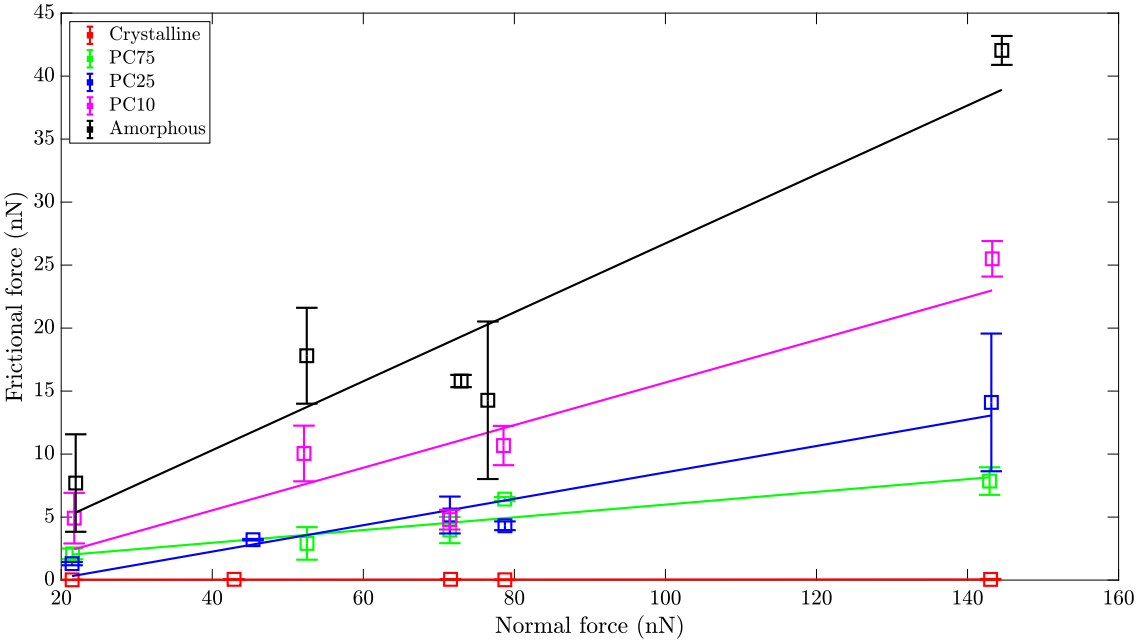


Fig. 8. Plots of normal force with respect to frictional force for MoS_2 with different degrees of crystallinity to measure the coefficient of friction.

superlubricity [6,18,52]. However, while most prior studies focused on sliding MoS_2 over itself, our work demonstrates superlubricity in crystalline MoS_2 during rigid body sliding simulations. The COF value increases with a reduction in crystallinity. Notably, the highest value (0.2736) is observed for amorphous MoS_2, which is consistent with the values reported in [51]. This trend underscores the significant role of crystallinity in determining the frictional behavior of MoS_2, with higher crystallinity favoring lower friction. Even though the COF values from our experiments and MD simulations do not match exactly, the overall trend remains the same. In the following section, we explore the nanoscale friction mechanisms responsible for the variation in COF with respect to the degree of crystallinity.

3.2.2. *Friction mechanism*

In this section, we explore the friction mechanisms responsible for the observed differences in COF with varying crystallinity. For crystalline MoS_2, the sliding surface remains flat throughout the simulation, even under the highest normal load of 143 nN. This flat surface minimizes resistance to the motion of rigid body, contributing to the exceptionally low COF value. Another major factor for the superlubricity observed in crystalline MoS_2 is the anisotropy in friction [59,70]. Even though we slide the rigid body in horizontal direction, the lateral movement is not restricted, allowing the rigid body to follow a zigzag path across the MoS_2 surface as shown in Fig. 9. The movement of the rigid body is captured at four different frames, each representing a window of 0.125 ns. In Fig. 9, the red arrow in the top two frames indicates the direction of sliding, while the black arrows depict the actual path followed by the rigid body. The movement of the rigid body over time is represented in a clockwise direction in Fig. 9. This zigzag trajectory represents the path of minimum energy, further reducing frictional resistance. The anisotropic friction observed in this work is consistent with the findings reported in prior literature [59]. However, this anisotropic friction is less prominent in PC MoS_2 and almost negligible in amorphous MoS_2, thereby validating their high values of COF.

To further understand the variation in COF values, we investigate the surface roughness of MoS_2 as a function of crystallinity. Using the Gaussian density method in OVITO [64,71], we construct a geometric surface mesh that describes the outer and inner boundaries of the atomic structure. This method generates an isosurface from a volumetric density field computed by superimposing 3D Gaussian functions centered at each atom in the top layer. The resulting mesh provides a quantitative estimate of surface roughness, which is visualized through color-coded gradients representing the distance from the bottom to the top of the surface. Here, we select atoms present at the top surface of MoS_2 and construct a surface mesh. The resulting surface meshing for MoS_2 with varying degrees of crystallinity are presented in Figs. 10(a)–10(d), illustrating the relationship between crystallinity and surface morphology. The red and blue colored regions in the figures represent the atoms at the highest and lowest positions among the group of selected atoms. A constant color in the plot implies that all the atoms are positioned at the same level. This is evident in Figs. 10(a) & 10(b) (crystalline and PC25), where there is negligible color variation. In contrast, PC10 (Fig. 10(c)) and amorphous MoS_2 (Fig. 10(d)) exhibits pronounced color variations, denoting a significant surface disorder or roughness. This progressive surface roughness with decreasing crystallinity along with the anisotropy in friction explains the observed trend in COF values. Next, we investigate the effect of crystallinity on the wear resistance of MoS_2.

3.2.3. *Wear characteristics*

To study the wear characteristics of MoS_2 with different levels of crystallinity, we performed sliding simulation at an elevated normal load of 358 nN while maintaining a constant horizontal velocity of 10^{-4} Å/s. The path followed by the rigid body and the configurations of the top layer of atoms are investigated in detail and presented in Fig. 11. Further, we measured the depth of wear from the initial and final position of the rigid body. Fig. 11 reveals a strong dependence between crystallinity and wear resistance of MoS_2. For crystalline MoS_2 (Fig. 11(a)), a minimal wear depth of 5.32 Å with localized bond distortion at the topmost layer is observed. This wear depth increases progressively with decreasing crystallinity, culminating in complete structural disintegration for both for PC10 and amorphous MoS_2 (Figs. 11(d) & 11(e)). The sliding of top layer of crystal is also observed for crystalline MoS_2. Notably, the amorphous configuration shows a wear depth of 14.8 Å which is approximately three times greater than that of crystalline MoS_2. A significant deformation on the top layers of MoS_2 is observed during the wear simulations. This behavior

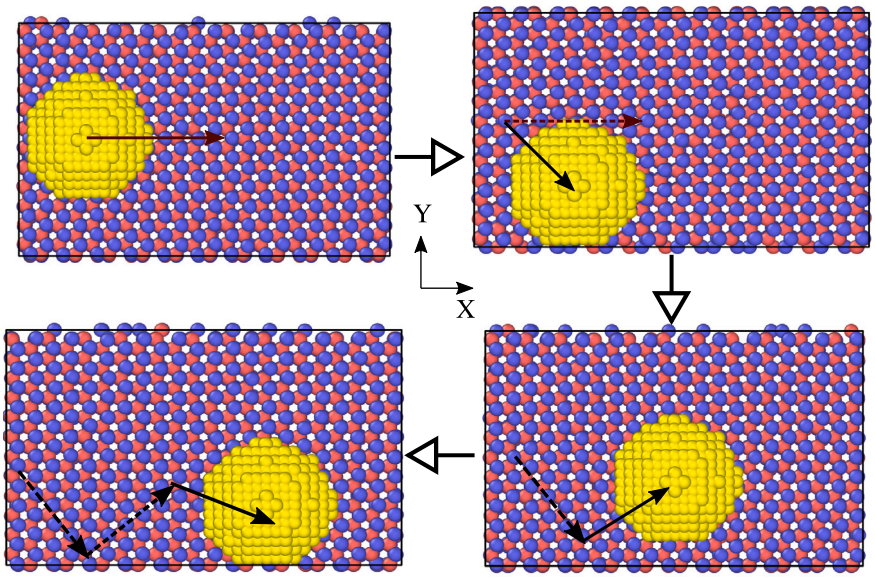
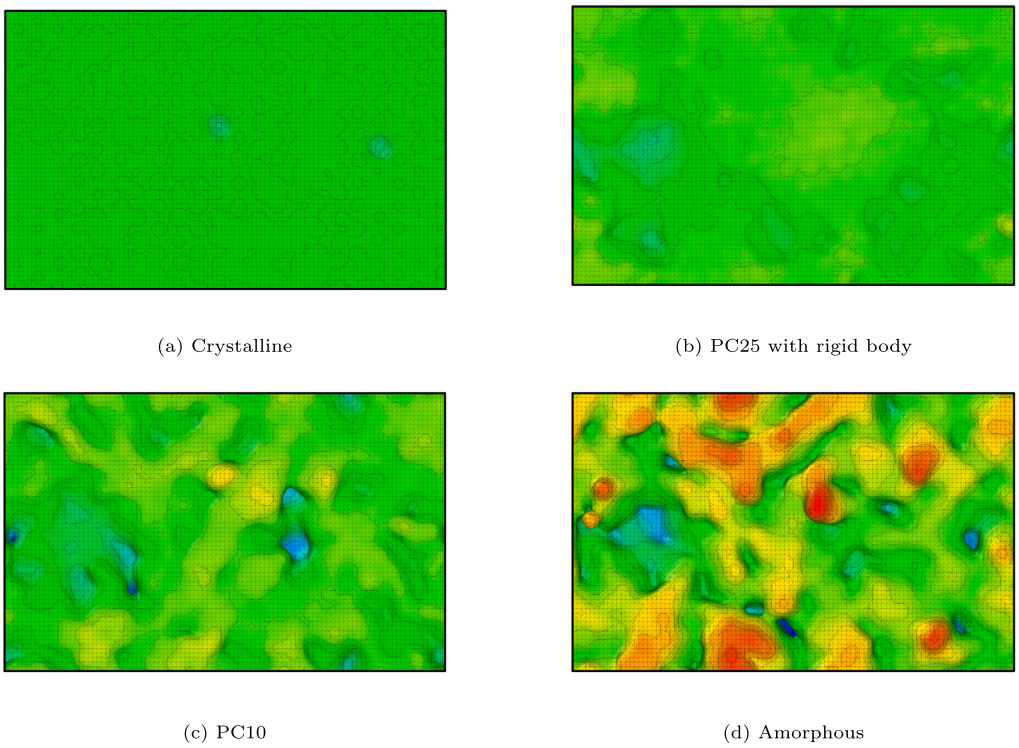


Fig. 9. Zig-zag movement of rigid body during sliding simulation in crystalline MoS_2.



(a) Crystalline

(b) PC25 with rigid body

(c) PC10

(d) Amorphous

Fig. 10. Geometric surface meshing for MoS_2 with different levels of crystallinity. Color gradients are used to represent the surface roughness with red and blue colored regions indicating atoms at the bottom at top surface.

arises from the finite thickness of the simulated structure, where the applied load propagates through the entire film and induces lateral deformation of the top layers due to bond breakage. Such deformation spreading beyond the immediate contact region is typical in atomistic wear simulations of thin slabs [72].

To further reinforce the dependence between crystallinity and wear resistance, we quantitatively analyzed the displacement of surface atoms in MoS_2. The displacement vectors in the Z direction are calculated from the atomic positions before and after sliding. The displacements are color coded and presented in Fig. 12. The red and blue colors represent maximum and minimum displacements. Crystalline, PC75, and PC25 configurations (Figs. 12(a)–12(c)) exhibit uniform atomic displacement patterns, indicating homogeneous deformation. In contrast, PC10 and amorphous MoS_2 (Figs. 12(d) & 12(e)), demonstrate significant displacement, with the amorphous structure showing distinct void formation at the surface. The pronounced displacement of atoms at lower degrees of crystallinity implies a reduction in wear resistance. These atomic-scale observations — combining both structural configurations and displacement patterns — establish a direct correlation between crystallinity and wear resistance. Thus, a higher degree of crystallinity provides superior wear resistance compared to amorphous MoS_2. It is to be noted that although grain boundaries and vacancies

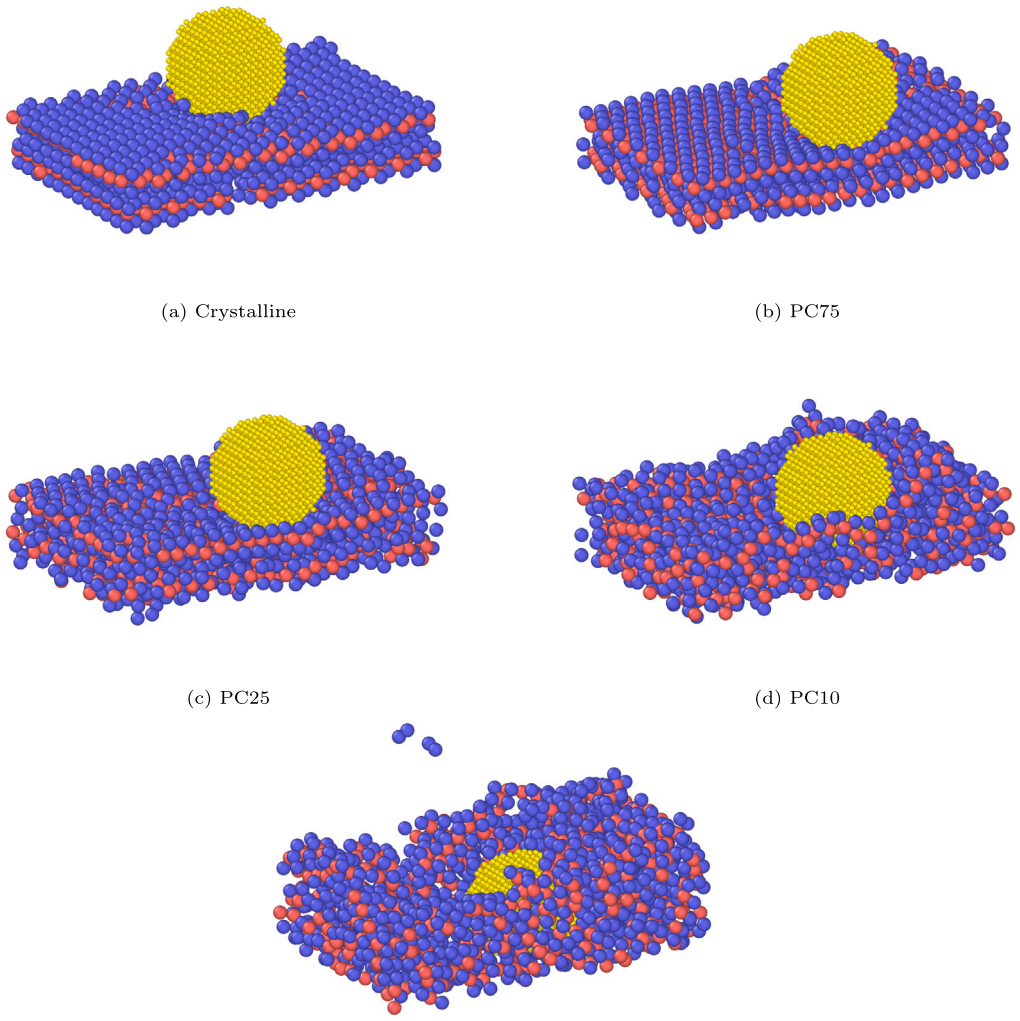


Fig. 11. Atomic representation of wear in MoS_2 with different levels of crystallinity.

affect tribological behavior, their numerous possible configurations at a given crystallinity would introduce additional variables and obscure the isolated effect of crystallinity, and hence not considered in this study.

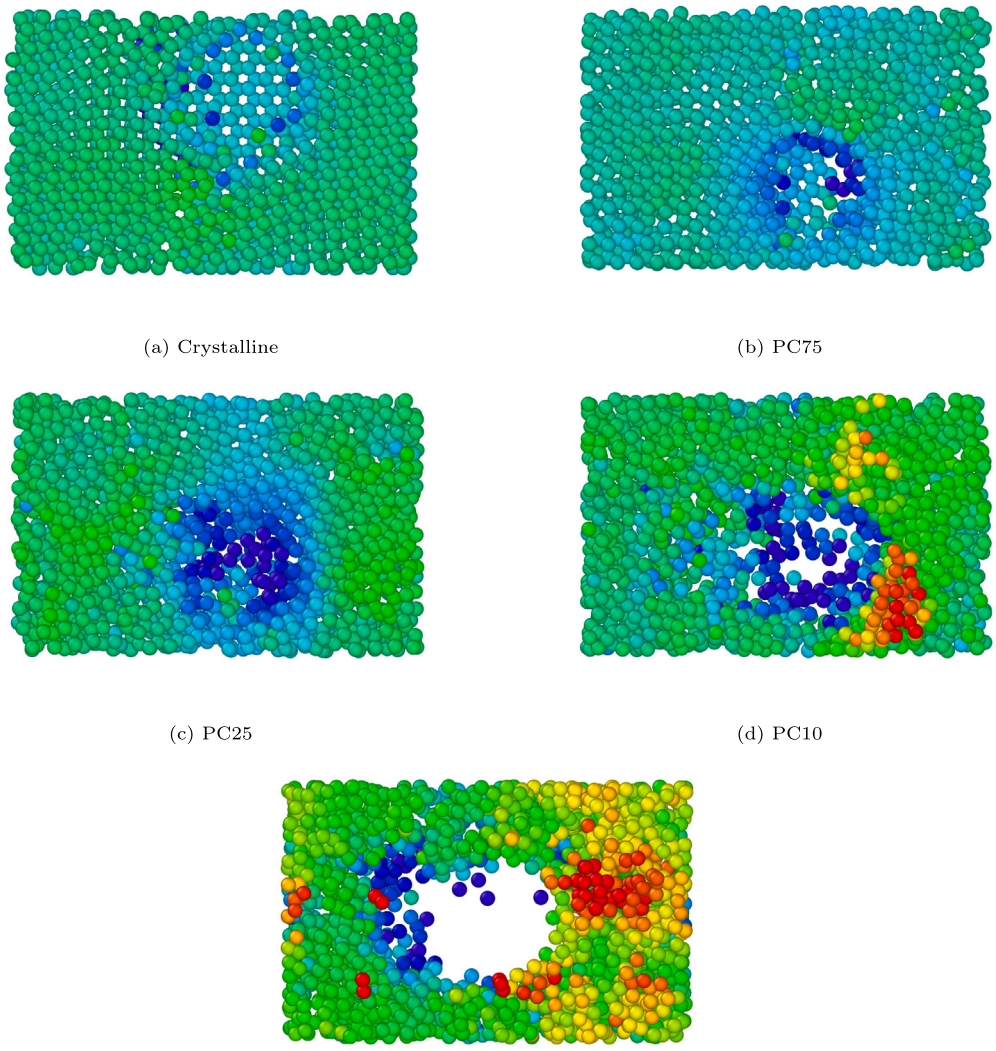
3.3. Comparison between experiments and MD simulations

The combined experimental and MD results reveal a consistent correlation between the degree of crystallinity and the tribological performance of MoS_2. Both approaches demonstrate that increasing crystallinity leads to a substantial reduction in COF and enhanced wear resistance. In the experiments, crystalline MoS_2 exhibited an ultralow COF (0.035) and negligible wear, whereas amorphous and PC films showed higher friction and pronounced tribo-polishing effects. Similarly, the MD simulations reproduced this monotonic trend, with the crystalline structure achieving superlubricity (0.00014) and the amorphous configuration showing a nearly threefold increase in wear depth. Although the absolute COF values differ due to differences in testing scale, surface roughness, and sliding conditions, the underlying dependence on crystallinity remains the same across both studies. Since the factors influencing MD simulations and experiments have a distinct physical origin, they cannot be isolated in a unified framework. Thus, a direct quantitative calibration between experimental and simulation COF is not feasible.

Mechanistically, the agreement between experiments and simulations stems from the same physical origins. The MD simulations reveal that the atomically ordered layers in crystalline MoS_2 maintain smooth sliding interfaces and enable anisotropic frictional response through interlayer shear. This matches the low-friction behavior and negligible wear observed experimentally. Conversely, amorphous and PC MoS_2 display significant surface roughness and atomic-scale disorder, promoting energy dissipation and surface damage; consistent with the experimentally observed tribo-polishing effect. Sliding of top layer of crystalline MoS_2 is observed during wear process in both experiments and MD simulations. Overall, the parallel trends in friction and wear from both experimental and atomistic scales confirm that crystallinity is the dominant factor governing the tribological response of MoS_2. The integrated insights from experiments and simulations not only validate each other but also provide a comprehensive understanding of how structural order controls frictional anisotropy, wear resistance, and ultimately, superlubricity in layered materials.

4. Conclusion

This study establishes a clear relationship between crystallinity and the tribological performance of MoS_2 through a combined experimental and molecular dynamics approach. Both experiments and



(a) Crystalline (b) PC75

(c) PC25 (d) PC10

(e) Amorphous

Fig. 12. Color coded displacement of atoms at the top surface of MoS_2. Red and blue colors denote atoms with maximum and minimum displacement.

simulations consistently demonstrate that increasing crystallinity leads to a significant reduction in friction and enhanced wear resistance. Crystalline MoS_2 exhibits ultralow friction and negligible wear due to interlayer sliding and frictional anisotropy, whereas amorphous and PC structures show higher friction and pronounced material removal driven by surface roughness and atomic-scale disorder. The strong agreement between experiments and simulations confirms that crystallinity is the dominant factor governing the frictional and wear behavior of MoS_2. These findings provide atomic-scale insights into the mechanisms governing friction and wear in layered materials, establishing a fundamental basis for the design of advanced solid lubricants and wear-resistant coatings. The results have direct implications for engineering applications demanding ultralow friction and high durability, including aerospace systems, microelectromechanical devices, and next-generation lubrication technologies. Beyond establishing this fundamental relationship, the present study also offer clear design guidance: crystallinity can be used as a tunable parameter to tailor MoS_2 coatings for specific operating conditions.

CRediT authorship contribution statement

Abhiram B.R.: Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Yue Wang:** Writing – original draft, Methodology, Formal analysis, Conceptualization. **Ilia Ponomarev:** Methodology, Formal analysis, Conceptualization. **Tomas Polcar:** Writing – review & editing, Supervision, Resources, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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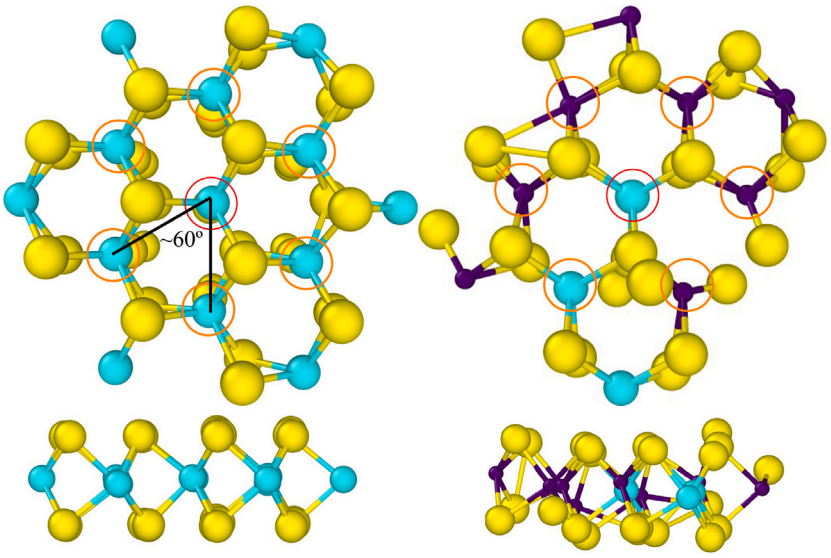

Fig. A.1. Representation of "crystalline Mo" atoms: the top images show views along *z*-axis, while bottom images present views perpendicular to it. Cyan spheres are "crystalline Mo" atoms, purple spheres - other Mo atoms, yellow spheres - sulfur atoms. The red circle highlights the Mo atom in focus, orange circles denote second-neighbor Mo atoms.

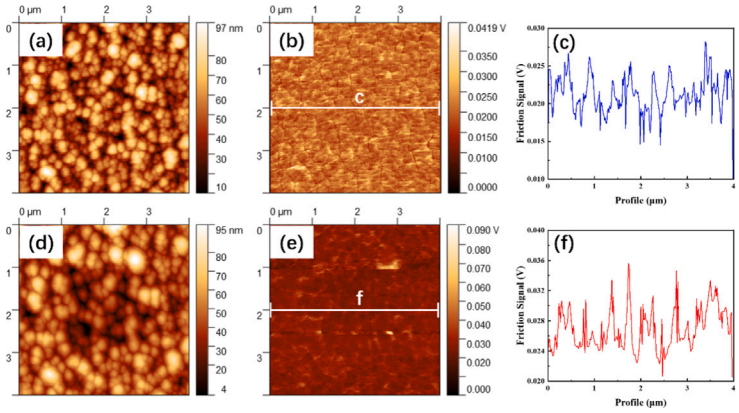

Fig. B.2. Topography, friction map and line profile from friction map (white line) of amorphous sample: (a–c) original surface before wear test, (d–f) after wear test.

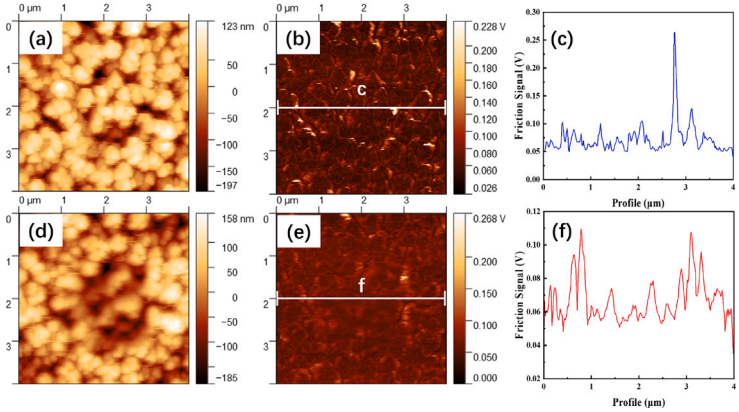

Fig. B.3. Topography, friction map and line profile from friction map (white line) of polycrystalline sample: (a–c) original surface before wear test, (d–f) after wear test.

Appendix A. Degree of crystallinity

The degree of crystallinity is quantified as the percentage of Mo atoms classified as "crystalline Mo" among all Mo atoms in the system. This classification is based on the characteristic structural arrangement of the 2H-MoS_2 crystal. To determine whether a given Mo atom is crystalline, we analyze its surrounding Mo neighbors and the angles formed by Mo–Mo–Mo triplets, where the atom in focus is the central

one. In a perfect crystal, each Mo atom is surrounded by six second-neighbor Mo atoms arranged in the corners of a perfect hexagon. This results in six Mo–Mo–Mo angles of 60°, six angles of 120°, and three angles of 180°. To identify crystalline Mo atoms in our analysis, we consider atoms meeting the following criteria: at least five Mo–Mo–Mo angles must fall within 50° to 70°, at least five more within 110° to 130°, and at least two angles exceeding 170°. This criterion minimizes false positive identifications. Fig. A.1 provides two examples of crystalline Mo atoms: one nearly ideal from a perfect crystal (left) and another highly distorted from an amorphous structure (right). It is to be noted that crystallinity can also be assessed using S atoms instead of Mo atoms. However, both methods provide equivalent degrees of crystallinity.

Appendix B. Topography and friction map

See Figs. B.2 and B.3.

Data availability

Data will be made available on request.

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