Atomic-Scale Superlubricity in Ti$_2$CO$_2$@MoS$_2$ Layered Heterojunctions Interface: A First Principles Calculation Study

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ABSTRACT: The two dimensional (2D)-layered transition-metal carbides and nitrides (MXene) have been proved to be an excellent solid lubricant owing to their high mechanical strength, low shearing strength, and self-lubricating properties. However, the interfacial friction behavior between Ti$_{n+1}$C$_n$ (n = 1, 2) MXene and its heterogeneous system is not thoroughly exploited yet. Here, four types of van der Waals structures (Ti$_2$CO$_2$@Ti$_2$CO$_2$, Ti$_3$C$_2$O$_2$@Ti$_3$C$_2$O$_2$, MoS$_2$@MoS$_2$, and Ti$_2$CO$_2$@MoS$_2$) have been investigated by density functional theory (DFT) calculations. The results exhibit that Ti$_2$CO$_2$@MoS$_2$ possesses the lowest sliding energy barrier around 0.015 eV/oxygen(O) atom compared with the other three constructed models. Therefore, this work mainly focuses on the inner relation of Ti$_2$CO$_2$@MoS$_2$ interlayer friction behaviors and its attributing factors, including normal force and charge density. The DFT analysis shows that the roughness of the potential energy corrugated plane is positively correlated with normal force and predicted the ultralow friction coefficient ($\mu$) at 0.09 when sliding along the minimum energy potential route. Moreover, friction coefficient fluctuates at the normal force less than 10 nN determined by the combined effect of interfacial charge interlock and redistribution. This work reveals the intrinsic connection between the friction and charge interaction at heterogeneous interfaces.

INTRODUCTION

The two dimensional transition-metal carbides families, collectively referred to as MXenes, have garnered increasing attention in energy storage, catalysis, and mechanotribological aspects since its first discovery in 2011. The major of studies have shown that the introduced oxygen(O) terminated on the MXene surface during the etching process, which weaken interlayer coupling between adjacent layers. Recent works on the MXene family have proven it to be a new and promising candidate of the solid sliding friction lubricant in both theoretical calculations and experimental analysis, owing to its excellent characteristics. Such as weakly bonded multilayer structure, high mechanical strength, low shearing strength, and self-lubricating capability. Recent works on the good lubrication performance of Ti$_{n+1}$C$_n$ has been reported, such as a Ti$_2$C$_2$(OH)$_2$ nanometer sheet as an additive in base oil for effective improvement of the friction-reducing and antiwear ability.

In past years, studies have observed that heterostructure synthesis and fabrication together obviously provide us with more opportunities to change the interfacial properties and then enable better lubrication performance, such as fluorographene (FG)/molybdenum disulfide (MoS$_2$), graphene/...
MoS₂, and graphene/h-BN heterogeneous interface. The observation of the graphene/MoS₂ heterostructure reveals that the sliding energy barrier is much smaller than that of the homogeneous bilayer, which is in good agreement with the experimental results. Recent techniques based on atomic force microscopy in the lateral force mode have been used to experimentally explore the tribological properties between scanning probes and 2D nanosheets. However, since the reported frictional measurements were performed on only a homogeneous layer, and significant changes in probe shape or nanosheet surface chemistry cannot be ignored during the experiments. It is still a challenge to accurately simulate 2D materials interfacial friction behavior and investigate friction properties. In order to further explore the 2D Tiₙ₊₁Cₙ⁻MXene (n = 1, 2) interlayer friction mechanism and broaden its tribological applications, quantum mechanics-based first principles methods provide a powerful alternative to explore tribological properties of 2D materials at the atomic scale. Here, MoS₂ was selected as the heterogeneous nanosheet mainly based on the following two factors: (1) the superior mechanical properties, including large in-plane Young’s modulus and low bending rigidity; (2) good lattice matching with Tiₙ₊₁Cₙ; and (3) low intrinsic friction response.

MoS₂ and graphene/h-BN heterogeneous interface. The observation of the graphene/MoS₂ heterostructure reveals that the sliding energy barrier is much smaller than that of the homogeneous bilayer, which is in good agreement with the experimental results. Recent techniques based on atomic force microscopy in the lateral force mode have been used to experimentally explore the tribological properties between scanning probes and 2D nanosheets. However, since the reported frictional measurements were performed on only a homogeneous layer, and significant changes in probe shape or nanosheet surface chemistry cannot be ignored during the experiments. It is still a challenge to accurately simulate 2D materials interfacial friction behavior and investigate friction properties. In order to further explore the 2D Tiₙ₊₁Cₙ⁻MXene (n = 1, 2) interlayer friction mechanism and broaden its tribological applications, quantum mechanics-based first principles methods provide a powerful alternative to explore tribological properties of 2D materials at the atomic scale. Here, MoS₂ was selected as the heterogeneous nanosheet mainly based on the following two factors: (1) the superior mechanical properties, including large in-plane Young’s modulus and low bending rigidity; (2) good lattice matching with Tiₙ₊₁Cₙ; and (3) low intrinsic friction response.

Therefore, the well-optimized structural models in this paper are decorated with oxygen atoms. This work aims to explore sliding energy barriers between adjacent layers using density functional theory (DFT) methods and predicts the lowest friction coefficient (μ) of the Ti₂CO₂@MoS₂ heterogeneous interface when sliding along the minimum energy potential (MEP) route. The Ti₂CO₂@MoS₂ heterogeneous interfacial friction mechanism and its attributing factors, including normal force and charge density, have been carefully studied as well.

### RESULTS AND DISCUSSION

In order to exploit the atomic level interfacial friction behavior of 2D-dimensional Tiₙ₊₁Cₙ⁻ (n = 1, 2) and its heterostructure, we performed DFT calculation on the Ti₂CO₂, Ti₁C₁O₂, and MoS₂ bilayers, as well as the Ti₂CO₂@MoS₂ heterostructure. The demonstration of interfacial sliding friction simulation is shown in Figure 1a referring to the reported studies. Different stacking sequences can be fabricated by moving the relative atoms positions of adjacent layers, respectively. Figure 1b,c shows top views of sliding routes with different underlying layers. Because Ti₂CO₂@Ti₂CO₂, Ti₁C₁O₂@Ti₂CO₂, and Ti₂CO₂@MoS₂ models have the same top view of underlying layer, in other words they have the same stacking sequences. Three typical stacking sequences are illustrated in Figure 1d–f. Additionally, three typical stacking sequences of MoS₂@MoS₂ are shown in Figure 1g–i.
In this paper, the sliding energy barrier $\Delta E$ is a descriptor that measures sliding difficulty. $\text{MoS}_2@\text{MoS}_2$, $\text{Ti}_2\text{CO}_2@\text{Ti}_2\text{CO}_2$, bilayer and $\text{Ti}_2\text{CO}_2@\text{MoS}_2$ heterogeneous interlayer without considering the normal force have been calculated, as shown in Figure 2. The detailed results of $\Delta E$ are listed in Table 1.

### Table 1. Maximum Energy Corrugation along the $x$ Direction in Different Heterogeneous Interfaces

| bilayer/heterogeneous interface | $\Delta E$ (eV/atom) |
|---------------------------------|---------------------|
| $\text{Ti}_2\text{CO}_2@\text{Ti}_2\text{CO}_2$ | 0.08                |
| $\text{Ti}_2\text{CO}_2@\text{Ti}_2\text{CO}_2$ | 0.13                |
| $\text{MoS}_2@\text{MoS}_2$        | 0.09                |
| path1-$\text{Ti}_2\text{CO}_2@\text{MoS}_2$ | 0.02                |
| path2-$\text{Ti}_2\text{CO}_2@\text{MoS}_2$ | 0.015               |

The calculated results show that the $\text{Ti}_2\text{CO}_2@\text{MoS}_2$ heterogeneous interface shows the lowest energy barrier compared with $\text{MoS}_2@\text{MoS}_2$, $\text{Ti}_2\text{CO}_2@\text{Ti}_2\text{CO}_2$, and $\text{Ti}_2\text{CO}_2@\text{Ti}_2\text{CO}_2$ when sliding along the $x$-axis. The maximum energy barriers of $\text{MoS}_2$, $\text{Ti}_2\text{CO}_2$, and $\text{Ti}_2\text{CO}_2$ are 0.09, 0.08, and 0.13 eV/atom, respectively, which is significantly larger than 0.02 eV/atom calculated from $\text{Ti}_2\text{CO}_2@\text{MoS}_2$. At the same time, as shown in Figure 2a, a saddle point corresponds to the $T_3$ stacking appears on the curve of $\text{Ti}_2\text{CO}_2@\text{MoS}_2$ and $\text{Ti}_2\text{CO}_2@\text{Ti}_2\text{CO}_2$. The maximum $\Delta E$ corresponds to the $T_2$ stacking sequence, which the bottom oxygen atoms of the upper layer and top oxygen atoms of the underlying layer are in a high symmetry. The sliding energy barrier $\Delta E$ increases because of the strong Coulomb force between adjacent oxygen atom layers.

Furthermore, the interfacial friction behavior of $\text{Ti}_2\text{CO}_2@\text{MoS}_2$ when sliding along different directions have been explored, as illustrated in Figure 2b. The results show that $\text{Ti}_2\text{CO}_2@\text{MoS}_2$ has the property of energy surface corrugation anisotropy which is obviously dependent on the stacking configurations. The maximum, minimum, and saddle point correspond to different stacking configurations. When sliding along the $x$-axis, the maximum sliding energy barrier is 0.02 eV/atom and the saddle point corresponds to the $T_3$ stacking configuration. However, when sliding along the $y$-axis, there is no saddle point and $\text{Ti}_2\text{CO}_2@\text{MoS}_2$ shows a lower sliding energy barrier (0.015 eV/atom). This phenomenon can be attributed to the broken highly symmetrical interface stacking models when sliding along the $y$ axis, which weakened the interfacial Coulomb force. The results show that lower $\Delta E$ was achieved by assembling heterogeneous nanosheets together.

In theoretical explorations, the potential energy configuration has been universally accepted for nanotribology even superlubricity performance. To understand the energy surface corrugation of different stacking sequences, we sampled the potential energy surface (PES) using DFT calculations, by considering all configurations across a 7 × 8 grid above an identical underlying layer, with its interlayer distance fixed under 1 GPa for $\text{Ti}_2\text{CO}_2@\text{MoS}_2$.

Figure 3a–c represents three typical stacking sequences. The PES landscape calculated under 1 GPa (Figure 3d) presents an exceptionally mild corrugation of 0.015 eV oxygen(O) atom, which is lower than 0.02 eV/O atom along the same path 2 (0.2671 eV), as listed in Table 1. In other words, a smaller $\Delta E$ was achieved when a low normal force applied on $\text{Ti}_2\text{CO}_2@\text{MoS}_2$. The minimum energy path of $\text{Ti}_2\text{CO}_2@\text{MoS}_2$ has been determined and indicated by the red arrow in Figure 3d. When sliding along the minimum energy path, and the lowest energy barrier is 0.1429 eV (0.19 eV/nm²), which is much lower than the reported 0.38 eV/nm² for $\text{Ti}_2\text{CO}_2$. Additionally, the friction coefficient along the minimum energy path, as demonstrated in Figure 3e, is predicted at 0.09 through an instantaneous slope of the spline that fitted to the results of energy versus sliding distance, showing superlubricity of $\text{Ti}_2\text{CO}_2@\text{MoS}_2$. From the PES landscape, we found an interesting phenomenon that a low normal force prompts easier interlayer sliding, which can be attributed to the surface charge redistribution.

In this work, $\text{Ti}_2\text{CO}_2@\text{MoS}_2$ possesses the lowest $\Delta E$ around 0.015 eV/O atom compared with the other three constructed models. Therefore, this work mainly focuses on $\text{Ti}_2\text{CO}_2@\text{MoS}_2$ interlayer friction behaviors. The calculated friction force is obtained through the instantaneous slope of the spline that fitted to the results of energy versus sliding distance, showing superlubricity of $\text{Ti}_2\text{CO}_2@\text{MoS}_2$. From the PES landscape, we found an interesting phenomenon that a low normal force prompts easier interlayer sliding, which can be attributed to the surface charge redistribution.
path 1 and 0.22 along path 2) compared with the 0.273 of Ti$_2$CO$_2@$Ti$_2$CO$_2$. Through the upper layer sliding in different directions, the correlation coefficient value of $R_1$ and $R_2$ are almost the same when sliding along different paths, indicating that the friction force positively correlates to the normal force.

The correlation coefficient $R_1^2$ and $R_2^2$ decreases when the normal force is less than 5 nN, as shown in Figure 4b. Despite the small deviation, the wild fluctuation of the friction coefficient cannot be ignored when the normal force is at less than 5 nN. The friction coefficient as a function of normal force is reported in Figure 4c. The friction coefficient irregularly fluctuates at a low normal force. The calculated friction coefficient is 0.195 along path 1 and 0.265 along path 2 when the normal force is at 0.9 nN.

In order to explain the reasons for irregular fluctuation, the sliding energy barriers under the incremental interlayer distance when sliding along path 2 have been calculated. The higher sliding energy barrier needs to be overcome when the interlayer spacing decreases, as illustrated in Figure 4d. Moreover, the increment of the sliding barrier increased and then became stable at an atomic scale level friction. The nature of the normal force at the atomic scale is the charge interaction between adjacent layers. The reasons for irregular fluctuation can be attributed to the charge environment.

In order to deeply exploit the relationship between the interface charge distribution and the surface energy corrugation, we calculated the charge difference maps under zero loads of Ti$_2$CO$_2@$Ti$_2$CO$_2$, Ti$_2$CO$_2@$MoS$_2$, and MoS$_2@$MoS$_2$ bilayers, as well as the Ti$_2$CO$_2@$MoS$_2$ heterogeneous interlayer under 20 nN, as shown in Figure 5.

We have carefully studied the interface charge density and its distribution under a different normal force. As demonstrated in Figure 5a, the electrons of the monolayer are accumulated around oxygen atoms, thus causing a rich electron environment. When Ti$_2$CO$_2@$MoS$_2$ is at the balanced interlayer distance (2.7 Å), the weak Coulomb force interaction between adjacent layers flatten potential energy corrugation. Once the normal force increases to 20 nN (shown in Figure 5b), the interlayer potential energy corrugation becomes rough. Additionally, because of the increasing charge interaction between adjacent oxygen atoms layers, the electrons enriched surrounding the oxygen atomic layers are transferred back to the sulfur atomic layer. The charge depletion causes lower potential corrugation, whereas charge accumulation gives rise to higher potential corrugation, and thus higher friction.

According to the reported studies, the geometric interlock of the bilayer has been used to explain the basic mechanism of the MoS$_2$ fracture. Because of the high symmetry and strong charge interlocking, the interface charge ripple of Ti$_2$CO$_2@$Ti$_2$CO$_2$ (Figure 5c) shows rough corrugation. Therefore, the reason for lower energy corrugation of Ti$_2$CO$_2@$MoS$_2$ is that the introduction of hetero-interface weakens the charge interlock and changes the charge redistribution. Then, the interface charge become more delocalized, resulting in a much smaller ΔE. The MoS$_2@$MoS$_2$ bilayer (Figure 5d) shows no obvious anisotropy in friction along different directions.

Figure 4. (a) Effect of normal force load on Ti$_2$CO$_2@$MoS$_2$, and R1 and R2 represents linear fitting straight-line along path 1 and path 2, respectively, (b) enlargement of linear fitting straight-line in the shaded area, (c) friction coefficient as a function of the normal force when sliding along path 1 (x-axis) and path 2 (y-axis), and (d) potential energy profile of Ti$_2$CO$_2@$MoS$_2$ at different interlayer distances.

Figure 5. (a) Interface charge density and charge distribution under different normal force, (b) the calculated charge difference maps under zero loads of Ti$_2$CO$_2@$Ti$_2$CO$_2$, Ti$_2$CO$_2@$MoS$_2$, and MoS$_2@$MoS$_2$ bilayers, as well as the Ti$_2$CO$_2@$MoS$_2$ heterogeneous interlayer under 20 nN, and (c) potential energy profile of Ti$_2$CO$_2@$MoS$_2$ at different interlayer distances.
Despite lower potential corrugation, the accumulated charge layer strengthens Coulomb force causing large $\Delta E$.

In order to describe the inner relationship between the interlayer friction and the interfacial electronic properties quantitatively, the plane-averaged charge density difference $\Delta \rho$ in the $z$ axis has been calculated. Figure 5e shows the plane-averaged charge density difference of Ti$_2$CO$_2$@MoS$_2$ under a different normal force. The results show that the normal force promotes the accumulated interlayer charge of MoS$_2$ nanosheet transfer to the outer layer obviously but has little effect on the Ti$_2$CO$_2$ layer. The enlargement of the interfacial plane-averaged charge density, as shown in Figure 5f, provides more details for interfacial charge interaction. The charge accumulated in the oxygen atomic layer is transferred to the sulfur atomic layer. Moreover, the shape of the peak near the oxygen layer widens, indicating that interface charge distribution become more delocalized. Therefore, the interlayer charge interlock and the charge redistribution that the two factors affecting potential energy corrugation are competitive and jointly determine the sliding energy barrier, which correspond to the curve of friction coefficient $\mu$ versus normal force, as shown in Figure 4c. In other words, better lubrication would be achieved if the quantity of the electric charge could be bound to the interface and prevented from being transmitted to the subsurface.

**CONCLUSIONS**

In summary, atomic friction behaviors of Ti$_2$CO$_2$ and MoS$_2$ bilayers, as well as the Ti$_2$CO$_2$@MoS$_2$ heterogeneous interlayer have been exploited by using DFT calculations. The PES landscape of Ti$_2$CO$_2$@MoS$_2$ calculated at 1 GPa exhibits superlubricity along the minimum energy path with the friction coefficient at 0.09 and an exceptionally mild corrugation of $\sim$0.015 eV/O atom, proving the Ti$_2$CO$_2$@MoS$_2$ heterostructure as an excellent solid lubricant. The introduction of the heterogeneous interface breaks the interlayer charge symmetry and promotes the interface charge redistribution, causing mild energy corrugation of Ti$_2$CO$_2$@MoS$_2$. Furthermore, the frictional force is positively correlated with the normal force and increases with the normal force. The charge density difference and plane-averaged charge density difference reveals that the competitive relationship between the interlayer charge interlock and the charge redistribution results in the irregular fluctuation trend of the friction coefficient in the Ti$_2$CO$_2$@MoS$_2$ heterostructure.

**COMPUTATIONAL DETAILS**

The theoretical calculations were performed by using the Vienna Ab initio Simulation Package (VASP 5.3.5) under the framework of the well-defined DFT simulation.$^{29,30}$ The projector augmented-wave method and Perdew–Burke–Ernzerhof functionals in the generalized gradient approximation method were used to describe the interaction between the core–valence electrons and electron exchange–correlation interaction, respectively. The cutoff energy was set to be 450 eV. The tolerance of energy precision of $10^{-5}$ eV and force convergence of 0.02 eV Å$^{-1}$ were employed in the structure geometric optimization. Monkhorst–Pack k-points meshes of $7 \times 7 \times 1$ was used for all the structure geometric optimization and DFT-D3 method for correcting dispersion in the system.$^{31}$
The supercells of $3 \times 3 \times 1$ have been fabricated for theoretical calculations at the atomic scale. In addition, 15 Å vacuum in the z-axis direction was employed to the upper layer to prevent interaction with the periodic layer. Because an induced lattice mismatch of 3.2% is within a reasonable range when the heterogeneous interlayer is fabricated, the construction of the heterogeneous interlayer model is reliable.

The adhesion energy $E_{\text{ads}}$ is used to describe the bonding strength between adjacent layers without considering the normal force, which can be acquired by subtracting the corresponding two monolayer energies from the bilayers calculated and defined as

$$E_{\text{ads}} = E_{\text{total}} - E_u - E_t$$  \hspace{1cm} (1)

where $E_{\text{total}}$ is the total energy of the heterogeneous or homogeneous models, $E_u$ and $E_t$ represent the total geometry optimized energy of the underlying layer and top layer, respectively.

Single-point self-consistent field calculations were employed to calculate sliding energy barriers under different normal forces when sliding along the x-axis (path 1) and y-axis (path 2). We scanned single-point energy with the interlayer distance ranging from 1.9 to 2.7 Å in steps of 0.1 Å. The calculated normal force is obtained through the instantaneous slope of the spline that fitted to the curves of energy versus interlayer distance. The average normal force can be expressed as

$$F_{\text{N}} = \frac{1}{N} \sum_{i=1}^{N} F_{\text{N}}(\tau_i)$$  \hspace{1cm} (2)

Lateral force ($F_{\text{L}}$) can be calculated by the same method, and more details refer to previous literature studies. The static friction coefficient is obtained according to the standard definition.

$$\mu = \frac{F_{\text{L}}}{F_{\text{N}}}$$  \hspace{1cm} (3)

To quantify interfacial friction of adjacent layers, the PES of the Ti$_3$C$_2$O$_2$@MoS$_2$ heterogeneous interlayer is mapped by calculating adhesion energy on a $7 \times 8$ grid. At each configuration of the grid, the upper layer was allowed to move along the plane of the underlying layer (the xy-plane) with a fixed interlayer distance in the direction of the z axis. Then, the PES landscape under 1 GPa can be obtained by adjusting the interlayer distance. The maximum energy corrugation is defined as

$$\Delta E = E_{\text{max}} - E_{\text{min}}$$  \hspace{1cm} (4)

where $E_{\text{max}}$ and $E_{\text{min}}$ represent the maximum and minimum energy of the PES when sliding along different directions.

The reported studies reveal that various functional groups, such as $-\text{O}$, $-\text{OH}$, and $-\text{F}$ groups are easily terminated on the surface of MXenes. However, the oxygen-decorated MXene model is universally accepted in theoretical calculations based on the following considerations: (1) in most cases, oxygen functional groups dominate during the etching process; (2) the high-temperature post-treatment or lithiation reactions will induce the conversion of $-\text{OH}$ groups to $-\text{O}$ groups; and (3) O-decorated MXenes show stability in thermodynamics and kinetics. Therefore, all structures in this work are fully covered oxygen. Additionally, the Ti$_3$C monolayer was obtained from the experimental synthesis by exfoliating the aluminum atomic layer. Then, the results of complete adsorption site tests show that the $-\text{O}$ termination is more favorable absorbed at the carbon (C) site at the surface of Ti$_{\text{h}}$C$_n$ ($n = 1, 2$) and sulfur (S) site at the surface of MoS$_2$, respectively.

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**Notes**

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