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Physical and chemical approaches to tailor 2D coating/substrate adhesion - Experimental and theoretical insights

Andreas Rosenkranz [a,b,*], Monica Soler [a,b], Guido Boidi [c], Bettina Ronai [c], Markus Varga [c], Fabiola Pineda [d], Maria Clelia Righi [e,**]

[a] Department of Chemical Engineering, Biotechnology and Materials, FCFM, University of Chile, Santiago 8370415, Chile
[b] ANID – Millennium Science Initiative Program, Millennium Nuclei of Advanced MXenes for Sustainable Applications (AMXSA), Santiago, Chile
[c] AC2T research GmbH, Viktor-Kaplan-Straße 2/C, Wiener Neustadt 2700, Austria
[d] Centro de Nanotecnología Aplicada, Facultad de Ciencias, Ingeniería y Tecnología Universidad Mayor, Santiago 8580745, Chile
[e] Department of Physics and Astronomy "Augusto Righi", University of Bologna, Bologna 40127, Italy

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ABSTRACT

Two-dimensional materials have been extensively studied due to their superior electrical, thermal, optical and mechanical properties, while the latter creates an enormous potential in tribology, especially when utilized as solid lubricant coatings. However, their usage as solid lubricant coatings under applied conditions remains scarce and challenging due to a limited quality of the coatings/substrate interface and lack of adhesion, thus inducing excessive wear as well as increasing friction and compromising durability/reliability. To enhance the performance of 2D coatings and bridge the gap between lab-tests and applied conditions (real-world applications), it is crucial to improve their substrate adhesion, facilitating the transition from a potential to tangible usage of 2D materials in tribology. Therefore, our perspective aims at summarizing the primary factors influencing the coating/substrate interface and adhesion, which can be subdivided into physical and chemical approaches. After critically summarizing the existing state-of-the-art related to experimental approaches, the current understanding based on numerical simulations, from density functional theory to machine learning-assisted molecular dynamics, is holistically analysed to provides atomistic insights to predict and design adhesive 2D interfaces. Our article closes with a perspective outlook on how to further boost coating/substrate adhesion thus guiding research activities with the overall goal to take full advantage of the outstanding properties of 2D materials in solid lubrication enabling the reliable implementation of 2D coatings in demanding, real-world tribological environments.

1. Introduction

Since the first mechanical scotch-tape exfoliation of graphene in 2004 [1], there has been an increasing interest in two-dimensional (2D) materials, particularly due to their unique physical and chemical properties, which outperform their three-dimensional (3D) counterparts [2]. Following the initial excitement connected with the excellent mechanical [3–5], thermal [6,7], electrical [8,9], and optical [10–12] properties of 2D materials, these materials have been extensively used in tribology [13–15]. Their strong in-plane and relatively weak interplanar bonds facilitate easy-shear motion between adjacent layers, thus reducing friction [16,17]. Moreover, these bond characteristics are essential to initiate the formation of stable and resistant tribofilms, which can simultaneously reduce friction while being mechanically robust and stable, thus enhancing durability and wear resistance [18–22]. Consequently, the design and development of 2D coatings applicable under lab- and applied conditions require an excellent chemical compatibility and mechanical adhesion of the coated material to the underlying substrate, thus making the coating/substrate interface the key influencer regarding the observed mechanical/tribological performance [23,24]. This is essential to guarantee that the 2D coatings are not removed by rubbing, thus withstanding rigorous testing conditions, i.e. severe temperature, humidity, atmosphere, and contact pressure as well as potentially inducing the formation of beneficial tribofilms.

* Corresponding author at: Department of Chemical Engineering, Biotechnology and Materials, FCFM, University of Chile, Santiago 8370415, Chile.
** Corresponding author.
E-mail address: arosenkranz@ing.uchile.cl (A. Rosenkranz).

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For tribological applications, graphene and MoS_2 are the most studied 2D materials until now [25–30]. However, the family of 2D materials constantly grows through the synthesis of new materials with improved properties including MXenes [19,31], hexagonal boron nitride (h-BN) [32,33] and black phosphorus (BP) [34–36]. Researchers also combine different 2D materials in heterostructures and/or composite structures (i.e., MXene/MoS_2 [37–39], MXene/graphene [40–42], graphene/MoS_2 [43–45], MXene/graphene oxide (GO) [20–22]) to induce synergic effects and overcome the drawbacks of the individual constituents. Apart from the usage of different 2D material systems, it becomes also evident that the choice of the underlying substrate and its surface chemistry notably affect the interfacial interactions and resulting adhesive strength for 2D coatings [46–48]. Such trends have been observed for graphene and its derivatives [46,47,49–51], MoS_2 [50], MXenes, among others. In case of graphene, it has been shown that graphene generally adheres well to semiconducting substrates with adhesion energies ranging between 73 and 188 mJ/m^2 [52]. A comparison of the resulting adhesion energies of graphene with metallic and ceramic substrates reveals that the resulting adhesional strength can be up to one order of magnitude increased for purely metallic substrates [50]. Regarding metallic substrates, graphene tends to adhere better to nickel (73.5 J/m^2) than copper (12.8 J/m^2) due to the establishment of more covalent bonds across the nickel-graphene interface. In contrast, copper generally forms ionic bonds with graphene [51]. Concerning metallic copper substrates, it has been shown that the oxidation state highly influences the resulting substrate adhesion. In this context, metallic copper or only lightly oxidized copper shows a more favourable adhesion behavior compared to highly oxidized copper substrates [46]. When moving from graphene-based materials to MoS2, MoS_2 has also shown an increased adhesional energy with metallic substrates compared to ceramic ones [50]. With respect to metallic substrates, it has been experimentally shown and numerically verified that MoS_2 adheres better to gold than copper substrates [53]. Moreover, it has been theoretically predicted that MoS_2 tends to show an enhanced adhesion with purely metallic substrates such as iron specimens (1.3 J/m^2) compared to the corresponding Fe_2O_3 substrates (0.35 J/m^2) [23]. Interestingly, the opposite trend holds true for $Ti_3C_2T_x$, which adhere better to Fe_2O_3 (0.87 J/m^2) compared to purely metallic Fe (0.40 J/m^2) [23]. When considering MXenes in particular, it has been demonstrated that the interfacial interactions and adhesion energies are less sensitive to the MXene stoichiometry and mainly governed by the existing surface terminations [54,55]. In case of silicon surfaces, Ti-based MXenes show similar adhesional energies than mono- and bi-layer graphene [55].

Screening the existing state-of-art literature, it becomes evident that 2D materials have moved towards the focus of fundamental research in tribology and are expected to play a pivotal role in future technological advancements due to their unique physical characteristics and potential applications. Remarkable tribological properties have verified in laboratory conditions, usually using less severe contact conditions, while the transition and application of 2D coatings to applied conditions and real-word components remain still scarce. Therefore, the gap between laboratory results and real-world applications can be considered as one of the remaining bottlenecks for many 2D materials. Despite the ongoing research effort, the main drawback and bottleneck of 2D coatings and their hybrids relate to poor coating/substrate interfaces and limited adhesion, thus compromising their friction and wear performance in lab-scale testing as well as applied field-tests. Therefore, we emphasize that improving the respective coating-substrate interface is one of the essential aspects, which will pave the way of 2D material coatings towards real applications. Consequently, our article sheds light on the available physical and chemical approaches (Fig. 1) to improve physical and chemical compatibility of the 2D coating with the underlying substrate thus creating strong and robust interfaces capable to withstand severe tribological testing conditions. Theoretical insights from computational modelling using density functional theory (DFT) and molecular dynamic (MD) simulations can further guide the research community on how to strengthen the involved interfaces.

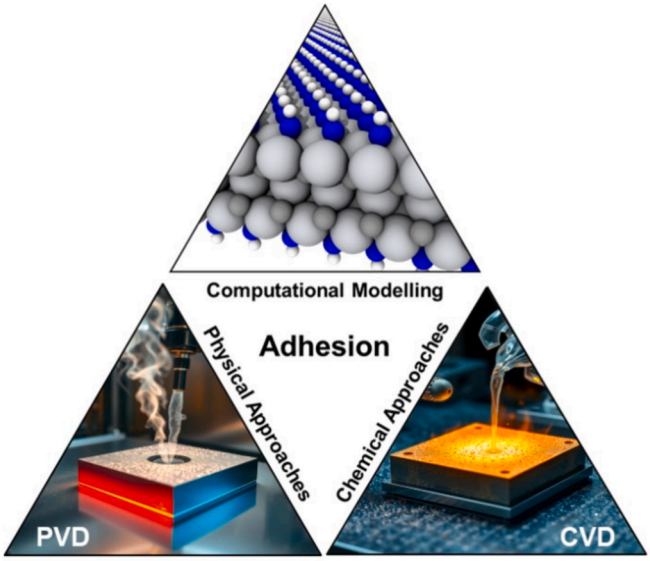

Fig. 1. Schematic illustration of physical and chemical approaches as well as input from theoretical modelling to improve the resulting coating-substrate interface and adhesion.

2. Physical approaches

Various physical approaches (Fig. 2) can be used to tailor the coating-substrate interface, thus improving the adhesion of 2D coatings. First, the deposition method chosen plays a pivotal role in the resulting interfacial strength and adhesion. Apart from that, other physical approaches that can precisely change and manipulate the coating-substrate

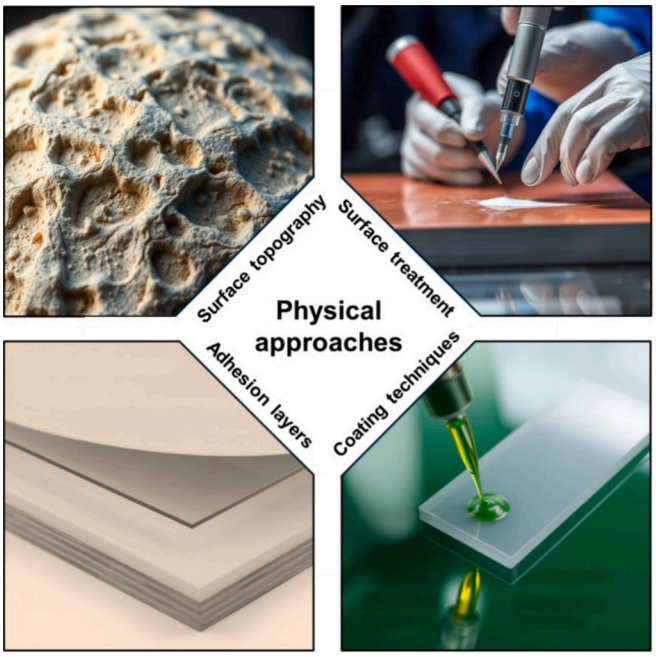

Fig. 2. Schematic illustration of examples, which resemble physical approaches inducing stronger, more robust substrate-coating interfaces and improving adhesion including a rough surface representing the effect of the underlying surface topography (top left); the manipulation of a surface, which can be achieved by surface treatment (top right); the usage of multi-layer architectures, which can involve the usage of an adhesion layer (bottom left) as well as a schematic representation of drop-casting for coating deposition (bottom left).

interface can be sub-divided into i) methods manipulating the underlying substrate topography/roughness. In this regard, the precise control and adjustment of the surface topography/roughness finds its inspiration in nature as evidenced by the multi-scale and hierarchical surfaces founds for the Gecko feet or butterfly wing thus beneficially influencing and favouring adhesion; ii) surface treatments to alter the surface energy or clearness without chemically compromising the substrate or iii) the deposition of thin films (adhesion layers) onto the underlying substrates to improve the chemical compatibility with the 2D coating.

2.1. Coating deposition techniques

The choice of the most adequate approach to deposit 2D materials onto the underlying substrates is crucial for the resulting quality and homogeneity of the coating as well as the establishment of strong and robust substrate/coating interfaces. The existing deposition techniques range from rather simple approaches such as drop-casting, spin-coating and spray-coating (Fig. 2 for an illustration of a common application of spray painting) to more elaborated ones, including electrophoretic deposition (EPD) ending up in scalable, more sophisticated methods such as physical and chemical vapor deposition (PVD and CVD), atomic layer deposition (ALD) and matrix-assisted pulsed laser evaporation (MAPLE). In this regard, it is essential to consider that the resulting coating quality relies on the possibility to deposit compact, densified coatings. The coating-substrate adhesion generally correlates with the energy input on the underlying substrate during the deposition of the 2D material. The involved energy input can help to mechanically intermix the deposited materials with the underlying substrate and chemically establish new bonds across the interface thus boosting adhesion.

Related to rather simple techniques, drop-casting spreads previously dispersed nanomaterials in a suitable solvent on the substrate to be dried in a controlled atmosphere. The overall approach is simple and less costly due to the involved lab-equipment and the fact that the film thickness can be adjusted by simply changing the volume deposited and the concentration of the nanomaterial. Moreover, drop-casting is known to be a fast approach, which can deposit (nano)-particles onto surface in a reasonable amount of time. However, the main drawback of this technique is that it creates rather inhomogeneous and disrupted coatings. The resulting morphology and quality of drop-casted films largely depend on the wettability of the used solvent with the underlying substrate, the evaporation rate of the solvent in the subsequent drying process as well as capillary forces associated with drying, which may induce undesired concentration profiles. The major bottleneck of this approach relates to the limited coating quality causing a poor substrate-coating adhesion and leading to a downgraded friction and wear performance. The latter results from the fact that 2D materials are only dispersed and subsequently dried on the respective substrate without any further energy input. While drop-casting is a simple and fast approach with up-scale potential, it is not the preferred choice for coating deposition due to the limited coating quality.

Another simple approach, which creates improved coatings compared to drop-casting, is spray-coating (simplified scheme in Fig. 3). This approach is easily implementable, effective and straight-forward to create coatings on various substrates. It involves forcing a pre-formed dispersion through a nozzle, thus forming a fine aerosol to be sprayed onto the desired substrate. The quality and homogeneity of these coatings is affected by the distance between the substrate and the nozzle, the flow rate, the pressure used for deposition, the substrate temperature, the concentration of the dispersion, and the number of spray repetitions. Spray-coating is up-scalable, since the underlying size of the substrate is not a limiting factor, while also curved surfaces can be coated with this approach. Its main challenge connects with the coating morphology, which frequently turns out to be rather irregular with remaining porosity. While the coating quality is improved compared to drop-casting, the coating-substrate adhesion is a major concern due to the limited energy input of the method, which needs further optimisation.

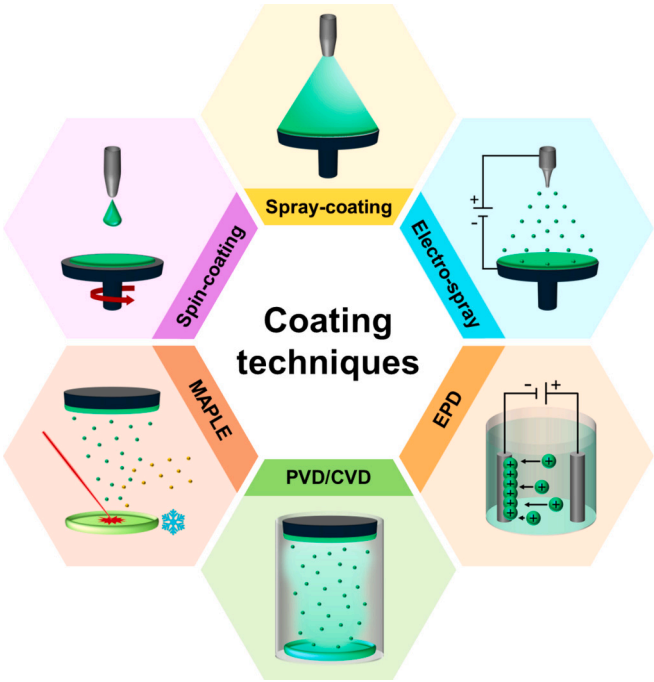

Fig. 3. Schematic overview of coating techniques applicable to 2D materials including spin-coating, spray-coating, electro-spray, electrophoretic deposition (EPD), physical/chemical vapor deposition (PVD/CVD), and matrix-assisted pulsed laser evaporation (MAPLE).

To create uniform thin coatings in a reproducible manner, spin-coating is often used. In this technique, a solution or a colloidal suspension is deposited in the centre of a flat substrate, which is then rotated to uniformly spread the solution through centrifugal forces, as shown schematically in Fig. 3 [56]. Similar to drop-casting and spray-coating, the solvent used evaporates, leaving the deposited particles on the substrate. The parameters influencing coating thickness are the solution concentration, volume, viscosity and evaporation, and rotational speed of the substrate [57]. The great advantage of this technique is the repeatability and homogeneity of the coating thickness. However, the adjustment of the coating thickness with low-viscosity solution is challenging. Moreover, this technique does not bring any benefits in terms of adhesion compared to spray-coating.

When moving to more elaborated deposition techniques, EPD (Fig. 3) comes into play. EPD resembles a highly versatile technique to generate uniform, compacted and more homogeneous 2D coatings, which can be deposited on various substrates having different geometries. The overall principle connects with the motion of charged particles in dispersion under the influence of an applied electric field towards the counter electrode with an opposite potential. The resulting thickness and film architecture as well as morphology are adjustable by the applied voltage, deposition time, distance between the involved electrodes and concentration of the 2D material in the deposition medium. When comparing EPD with drop-casting and spray-coating, EPD generates coatings with greater homogeneity and uniformity as well as improved substrate-coating interfaces. Considering the quality of the deposited coatings, EPD outperforms the more simplified techniques in terms of processing time, simple apparatus, substrate selection and geometry flexibility. A potential drawback of EPD relates to the fact that a homogeneous and stable dispersion is necessary, while the used particles require some positive or negative surface charge to be deposited on the respective counter-electrode. Moreover, the electrodes need to be electrically conductive, which somehow limits the respective material choice of the electrodes.

An additional technique that employs an electric field is electro-

spraying (simplified illustration in Fig. 3) [58,59]. A high-voltage source is applied to a metal nozzle through which the solution flows. The substrate is connected to the ground electrode and is typically heated to facilitate the solvent evaporation. The high electric field at the nozzle tip is responsible for the formation of fine drops and a thin jet cone of the solution, which is directed onto the substrate [60]. The parameters affecting the electro-spraying process include the voltage used, the geometry and dimensions of the nozzle, the distance between the nozzle and the substrate, the concentration of the solution, and the material utilized. This method can be performed at ambient temperature and atmospheric pressure, which makes it a low-cost, and low-energy method for the deposition of 2D materials.

PVD and CVD are two of the most prevalent techniques employed for the deposition of materials in both research and industrial contexts [61]. In both deposition techniques, the coating is formed by condensing a gas onto the substrate in a reaction chamber under vacuum, plasma, or low-pressure conditions (Fig. 3) [61–63]. In this context, PVD entails a physical reaction, whereby a solid precursor material is transformed into a vapor. In contrast, CVD involves a chemical reaction that produces the vapor (necessity of volatile precursor). Additionally, the temperature employed in PVD is lower (up to 450 °C) than that used in CVD, which typically exceeds 900 °C [61–63]. Both techniques are more sophisticated, costly and time-consuming than the aforementioned methods (i.e., drop-casting, spray-coating, spin-coating, EPD, electro-spraying). Furthermore, for CVD, hazardous chemicals are often used, and it is difficult to deposit different materials simultaneously due to the varying evaporation rates. Despite this, CVD and PVD are widely applied in industry, including in the aerospace, automotive, health and electronic sectors, due to their excellent reproducibility and coating adhesion to the substrate [64,65].

MAPLE has been employed for the deposition of a wide range of organic and polymeric materials, particularly in the domains of sensing, bioengineering, electronics, and optics [66,67]. Concerning this approach, the energy of a pulsed laser beam is applied to evaporate a frozen target within a vacuum chamber (as shown schematically in Fig. 3). The target is a solution consisting of a polymer or organic compound (typically <5 wt.-%) dissolved in a solvent [66]. The volatile solvent is extracted from the chamber by means of a pump, while the solute is deposited on the substrate. MAPLE allows for precise control of coating thickness (< 0.1 nm/ laser pulse, from nm to μm [66]) and it enables the formation of uniform and homogeneous thin coatings [68]. Moreover, this technique allows for the control of coating roughness, is highly reproducible and has been shown to enhance the adhesion of coatings to substrates [69]. It is important to note, however, that this technique is relatively complex, and the laser parameters must be carefully tuned to the properties of the target material and solvent in question.

2.2. Surface topography/roughness

When aiming at enhancing the resulting coating-substrate interface and improving the adhesional strength of 2D coatings to the underlying substrate, special attention needs to be paid to the role of the surface topography/roughness. As underlying substrates, mostly metallic surfaces are commonly polished or technically rough, which are then brought in contact with another rough counterpart (ball, pin or flat surface, among others). Depending on the used deposition technique, the surface topography/roughness can be replicated, which holds true for CVD and PVD, or ALD, among others [70]. When the substrate contains valleys on the surface, the deposition of nanoparticles can fill topographic defects, thus smoothing the resulting profile and reducing the surface roughness. This effect can be observed for drop-casting, spray-coating, and EPD, among others [70]. Irrespective of the deposition technique, the quality of the coating-substrate interface and the resulting adhesional strength are mainly governed by the established real area of contact between the coating and the substrate as well as the kinetic impact of the 2D materials upon deposition (for instance, spray-coating versus PVD techniques).

The general concept (the bigger the surface area, the greater the adhesional strength) finds its inspiration in animals such as insects, spiders, and lizards [71–73]. Their multi-scale textures induce a remarkable adhesional strength due to the involved contact splitting, thus establishing many small-sized contact spots instead of a few large-sized contact areas [74,75]. The most famous example for this outstanding adhesion relates to geckos, which results from the hierarchical, highly anisotropic and mechanically rather hard and compliant surface topography of their toes (top left image in Fig. 2) [76,77]. These optimized structures maximise the contact area between the gecko feet and the contacting surface thus maximising van-der-Waals forces and, therefore, adhesion. Similar effects have also been verified for other animals and insects [76,77]. Surface engineering strategies, such as laser surface texturing (LST) and substrate roughness control, play a critical role in modulating the tribological performance of coatings and lubricants. In laser-textured stainless steel and GCr15 substrates, the introduction of micro-dimples combined with h-BN induced a notable reduction of friction and wear under both dry and lubricated conditions. Separately, studies on MoS_2 coatings showed that the substrate roughness affects the friction behavior depending on the involved testing scale. For micro-scale tribological testing, rougher surfaces increased friction and wear, while at the macro-scale, they improved performance by enhancing load support and film stability. These findings underscore the need to tailor surface morphology according to the scale and operating conditions of tribological systems [48,78].

When bearing in mind that the optimisation of the contact area is one of the key factors to optimize and boost adhesional strength, surface texturing/patterning to manipulate the resulting surface topography/roughness plays an key role in creating high-adhesion biomimetically-inspired surfaces [79]. In this regard, micro-patterned elastomers with micropillars demonstrated an adhesional strength, which was 30 times greater than the respective reference surface without texturing [80]. Similar effects were verified for polydimethylsiloxane (PDMS) replica, which demonstrated a 3-fold increased adhesion for rigid, micro-textured aluminium surfaces [81].

Although the overall concept seems to be rather simple and straightforward, there is an important aspect to consider. The strength of van-der-Waals forces tends to decrease with increasing distance between the contacting surfaces. In this regard, surface texturing is known to increase the surface area (beneficial for improved adhesion) and the surface distance (detrimental for adhesion). Consequently, to achieve the greatest adhesional strength, special attention and care must be dedicated to this fact, thus ensuring the establishment of a conform contact between the deposited material and the surface texture underneath, which generally increases the elastic strain energy needed. When having a rigid and hard substrate material, the conform contact with the surface texture can only be well established using a fairly compliant nanomaterial with a balanced trade-off between elasticity, stiffness and bending rigidity [82]. For 2D nanomaterials, the texture dimensions such as depth and width must be adjusted based on the x- and y-dimensions of the 2D material to be deposited. If the texture parameters are not properly adjusted with respect to the material dimensions, the contact area between the coating material and the underlying texture may be only partially established. This may even lead to a downgraded adhesional strength and an easier removal of the deposited material.

Recent advances in scalable surface texturing techniques – such as LST, sandblasting, CNC machining, and nanoimprint lithography (NIL) - have opened promising pathways to engineer surface roughnesses and topographies at multiple length scales. These methods allow for the precise tuning of topographic features while maintaining scalability and cost-efficiency, thus bridging the gap between fundamental research and real-world implementation of robust 2D coatings.

2.3. Surface treatments

The surface energy is a crucial characteristic influencing wettability (as illustrated in the top right image in Fig. 2) and adhesion. Wettability is favoured when the substrate presents a higher surface energy compared to the coated material. Therefore, surface treatments aiming at increasing substrate adhesion affect the respective wettability and the underlying surface energy. In this context, the presence of surface contaminants can negatively influence the resulting substrate/coating adhesion.

There are several techniques to alter the surface energy and cleanness. Plasma approaches represent the most frequently utilized ones due to their effectiveness, short processing times (within minutes) and low consumption of chemicals [83]. Although plasma treatments are widely adopted, ultraviolet/ozone (UV/O_3) exposure represents a non-invasive alternative to enhance the surface energy of the substrates by removing organic contaminants and introducing oxygen-containing polar groups. This method is particularly suitable for sensitive surfaces, for which plasma etching is too aggressive. The plasma is formed by placing a gas or gas mixture (i.e., H_2, O_2, Argon, CF_4) under vacuum and applying an electromagnetic field. The plasma contains reactive species (i.e., ions, electrons and radicals) that can potentially chemically react with the material by etching the surface, thus removing surface contaminants, increasing surface energy and wettability of substrates as well as activating the underlying surfaces by creating reactive functional groups, potentially enhancing adhesion of 2D materials.

The main parameters governing plasma processes are the gas utilized, gas pressure, substrate temperature and exposure time [83]. By carefully controlling the plasma parameters, this technique can notably enhance the quality and adhesion of 2D coatings for various applications in electronics, optics, among others. For instance, Kim et al. [84] used atomistic simulations (i.e., DFT and MD simulations) to study the interaction between the acting plasma and the surface (Si wafer or glass), thus optimizing the gas used for specific 2D materials in electronic applications. The usage of oxygen plasma can greatly increase adhesion by enhancing the resulting surface energy, and creating reactive sites [85,86]. For graphene on Si substrates, the substrate treatment with an oxygen plasma before material deposition has shown to induce increased contact angles and adhesion [87]. The application of oxygen plasma facilitated the elimination of C-contaminants on the Si substrate, thereby enhancing surface hydrophilicity. This resulted in the homogeneous and reproducible deposition of graphene flakes on the Si substrate on a large scale. Additionally, fluorine-containing plasmas (CF_4/H_2) were more effective in terms of improving adhesion than pure H_2 plasma for graphene of Si substrates [88]. In general, CVD of graphene on copper substrates results in the formation of contaminants, including poly(methyl methacrylate) (PMMA) and silicon nanoparticles. The application of pure H_2 plasma treatment in conjunction with annealing at 400 °C proved ineffective to completely remove these contaminants and additionally introduced damages in the graphene structure. The use of CF_4/H_2 plasma was more effective in terms of contamination removal and avoided the alterations to the graphene structure [88].

Another method to reduce the amount of contaminations present (i.e., polymers [89,90] and water residues [91]) and improve the substrate adhesion connects with thermal annealing [92,93]. This post-processing treatment increases the contacting area between 2D materials and the substrate (increased substrate surface energy), thus enhancing van-der-Waals interactions and improving adhesion [86,93]. It is important to emphasize that the used annealing temperature must be optimized for each combination of 2D material and underlying substrate.

2.4. Adhesion layers

The layered nature of 2D materials is characterized by strong in-plane bonds and weak van-der-Waals forces between adjacent layers. While being beneficial for tribology in terms of easy-shear motion and tribofilm forming capabilities, the acting van-der-Waals forces are often insufficient to provide adequate adhesion on the underlying substrate, which can promote wear and coating delamination. The utilisation of adhesion layers (a real life example in the form of adhesive tape is pictured in Fig. 2) has been proven to be an effective approach to enhance the corresponding substrate/coating adhesion [94]. In this regard, adhesion layers resemble thin coatings that exhibit a high chemical affinity and reactivity with both substrate and 2D coatings, as well as a high surface energy (wettability). This property combination fosters the establishment of a robust coating-substrate interface, thus enhancing adhesion.

Metallic adhesion layers (e.g., Ti, Cr, Ta and W) can be employed for 2D coatings to boost adhesion with the substrate [94–96], which are typically deposited by PVD and CVD. It is imperative to consider the chemical affinities of the material involved as well as the thickness of the adhesion layer, as it is essential to avoid any mechanical damping effect as well as interference with the properties of the 2D materials.

Apart from single-element metallic adhesion layers, notable research activities have been dedicated towards the design of multi-layer architectures. The individual layers in multi-layer architectures can serve as ductile interlayers to reduce or even inhibit the propagation of cracks. This helps to improve the resulting adhesion and reduces the respective wear rates [97]. Intermediate layers can be used as gradient layers thus ensuring a smoother transition regarding the chemical and mechanical properties from the underlying substrate to the top 2D layer [98]. Hierarchical, multi-layer architectures can also combine harder and softer layers with the overall purpose of balancing internal and interface stresses. This overall approach must be particularly optimized regarding the adjusted thicknesses and mechanical properties of the individual layers [99].

Among the discussed strategies, multi-layer architectures and plasma-assisted adhesion layers show the highest potential for enhancing the coating performance for tribological applications. Their ability to combine mechanical robustness with tailored interfacial chemistry makes them promising to improve coating durability and adhesion in real-world environments.

2.5. Concluding remarks

In this section, we have underlined the essential role of the quality of the underlying substrate to create well-adherent coatings. In this regard, the surface roughness, cleanness and surface chemistry can be precisely manipulated by surface treatments, while all variables must be matched with the material to be deposited and coating deposition approach. Once having optimized the procedures for surface roughness and surface chemistry, thin metallic adhesion layers deposited by PVD can be utilized to further enhance the resulting interfacial strength and adhesion. When aiming at creating successful adhesion layer, the chemical affinity and compatibility with both the underlying substrate and 2D material are key influencers. After optimizing the substrate properties and potential adhesion layers, the deposition approach has to be appropriately chosen in terms of adjustable thickness, coating morphology, coating quality, processing temperature, among others. Moreover, it is pivotal to achieve a balance between the resulting coating quality and processing aspects such as deposition time, involved costs, substrate geometries, up-scalability and integration in industrial lines.

To facilitate a more intuitive understanding of the main differences between the discussed deposition techniques, Table 1 presents a qualitative comparison in terms of advantages, disadvantages, limitations, and adhesion level. This comparison highlights how more sophisticated techniques like PVD, CVD, and MAPLE generally offer better adhesion and coating quality due to higher energy input and precise process control, but at the expense of complexity and cost. In contrast, simpler methods, such as drop-casting and spray-coating, while scalable and cost-effective, suffer from a poor coating uniformity and limited adhesion. EPD and electro-spraying offer intermediate solutions, balancing

Table 1
Qualitative comparison of physical approaches used to deposit 2D coatings and improve the adhesion of these coatings to solid substrates. The table summarizes the main advantages, disadvantages, practical limitations, and relative adhesion performance of each technique.

Technique	Advantages	Disadvantages	Limitations	Adhesion level
Drop-casting	Simple, past. Low-cast	Inhomogeneous coatings, poor adhesion	Limited coating quality and performance	Low
Spray-coating	Scalable, applicable to various surfaces	Irregular morphology, porosity, limited energy input	Limited adhesion, needs optimization	Low to medium
Spin-coating	Uniform thickness, good reproducibility	Limited thickness control with low-viscosity solutions	No significant improvement in adhesion	Low
EPD	Compact, homogeneous coatings on various geometries	Requires charged, stable dispersions and conductive substrates	Material choices limited by charge and conductivity	Medium
Electro-spraying	Low-cost, ambient conditions	Needs fine control of parameters	Sensitive to setup geometry and parameters	Low to medium
PVD/CVD	High-quality, reproducible coatings, strong adhesion	Expensive complex, hazardous chemicals (CVD)	Cost, complexity, and chemical handling	High
MAPLE	Precise thickness control, homogeneous coatings	Complex, requires optimization of laser parameters	Complex setup and solvent dependence	High
Surface topography	Enhances real contact area, inspired by biological systems	May increase surface distance, decreasing Van der Waals forces	Requires careful adjustments of texture dimensions	Medium to high
Surface treatments (e.g., plasma)	Increases surface energy, removes contaminants	Plasma may damage sensitive surfaces	Required parameters control, can be surface-specific	Medium to high
Adhesion layers	Improves chemical compatibility and interfacial strength	May interfere with 2D material properties if not optimized	Needs matching chemical/mechanical properties	High

accessibility with improved coating compactness and substrate interaction. The table underscores the importance of matching the deposition technique to the application-specific demands regarding adhesional strength, substrate geometry, and process feasibility.

3. Chemical approaches

From the tribological point of view, another approach to develop sustainable and durable 2D coatings is to chemically modify or functionalize the substrate or the 2D material to be deposited, with the overall goal to increase their chemical compatibility/affinity and hence adhesion. To pursue this overall goal, a detailed understanding of the chemical affinities between the 2D coating and the substrate is imperative to design high-quality coating interfaces with good adhesion, thus enhancing wear/durability and reducing friction. To prepare these high-quality interfaces, the choice of the most adequate approach to deposit 2D materials onto the underlying substrates is again crucial for the quality and homogeneity of the coating.

The common interactions across the interface between 2D coatings and substrates generally relate to van der Waals forces, for which the London dispersion force is the major contribution, which results from the interaction of two instantaneously induced dipoles present on both surfaces [50,100]. This force becomes stronger with an increasing atomic radius due to the increased polarizability of the electron clouds and an increased contact area, which results in a stronger intermolecular interactions [50]. In this regard, the enhancement of the adhesion of a 2D coating to its underlying substrate can be understood as an increase of the chemical interaction and contact area across the interface.

This interaction can be generated and amplified through the chemical alteration of the superficial atoms and/or functional groups of the 2D material or the substrate. If the surface of the 2D coating or the underlying substrate do not offer a good chemical affinity, it is also possible to chemically functionalize them to enhance the possibility to induce new bonds or interactions, which is a way to chemically tailor and fine-tune the substrate/coating interaction. Finally, the intercalation of molecules, atoms, or ions or other materials that can interact or react with the substrate, is another way to strengthen the interfacial adhesion. In this context, it has been already demonstrated that chemical functionalization of the substrate and/or the 2D material can improve compatibility and bonding [101].

3.1. Substrate/coating interaction

Several studies have been focused on the understanding of the chemical affinities across the interface to assess the resulting adhesional strength. Torres et al. [50] investigated the van der Waals interactions of graphene and MoS_2 with various substrates including SiO_2, Si_3N_4, Au, and Pt. They demonstrated that graphene was more strongly adhered to these substrates than MoS_2 due to stronger interactions and MoS_2' higher Young's modulus for a given thickness. Although higher adhesion energy suggests stronger interfacial bonding, its impact on tribological performance (e.g., wear or friction resistance) is not always direct. The relationship depends for instance on additional factors like pressure and humidity and should be evaluated case by case. However, it is important to mention that the drawback of an increased Young's modulus can be overcome by particle interactions, as seen with the higher-than-expected adhesion energy between MoS_2 and SiO_2 due to an increased positive charge on the sulphur atoms of MoS_2 interacting with the negative charge of the oxygen atoms in SiO_2. These higher adhesion energies with the intermolecular S···O interactions causing an increase of interfacial bonding between sulphur and oxygen atoms have been previously described by Zhang et al. [102]. Torres et al. [50] also concluded that the adhesion energy for both graphene and MoS_2 was strongest on gold substrate with adhesion energies of 7687.1 and 1207.3 mJm^{-2}, respectively. They demonstrated that the London dispersion force becomes much greater for gold substrates compared to the other substrates (SiO_2 or Si_3N_4), which was explained by the polarizability of gold thus boosting interaction.

The existence of a native oxide on the interface has been shown to cause an increasing, additional adhesional strength. Calis et al. measured the work adhesion of MoS_2 for metal, semiconductor, and graphite substrates (Au, Ti, Cr, and Ge) by blister testing [103]. They reported that the van der Waals interactions between the Au substrate and the MoS_2 layer induced an easier layer exfoliation, compared with the other surfaces. This, in turn, implied an enhanced interfacial adhesion of the MoS_2 coating on Ti, Cr, and Ge surfaces, which was explained by the presence of native oxides. These results suggested that both surface roughness and chemical interactions play a critical role in the resulting adhesion of 2D materials on the underlying substrate, for which the passivation of these substrates helped to generate new chemical bonding thus increasing adhesion. While native oxides have been shown to improve adhesion by enabling chemical bonding, their behavior under tribological conditions must be holistically studied.

Depending on the oxide type and environment, they can undergo tribochemical transformations that either reinforce interfacial strength (e.g., by forming protective tribofilms) or degrade and lead to delamination. This situation highlights that controlled surface oxide formation, i.e. corrosion, may be even beneficial for the adhesion of 2D coatings.

The presence of airborne contaminants has been shown to notably affect the interfacial adhesion of 2D materials on substrates. Chandran et al. [104] assessed the interfacial adhesion of WS_2 bilayers grown via CVD on sapphire, SiO_2/Si, and fused quartz substrates by nano-scratch testing (Fig. 4). Their overall goal was to develop more effective transfer techniques for flexible electronic devices, looking for less adhesion, emphasizing the critical role of the substrate selection for their specific application. They concluded that the primary parameters influencing the resulting adhesional strength were the substrate properties, such as hardness and roughness as well as the presence of interfacial oxides. These findings confirmed that the adhesional strength of WS_2 increases in the order of sapphire, quartz, and SiO_2/Si. The maximum adhesional force observed for SiO_2/Si substrates was traced back to the involved charge transfer between the Si to the S atoms of WS_2. The presence of WO_3, formed during preparation of WS_2 on sapphire, increased the effective substrate roughness, thereby reducing adhesion. Although sapphire is intrinsically smoother, the oxide layer compromised its surface properties, resulting in lower delamination loads. Consequently, sapphire emerged as an ideal substrate to facilitate the easy transfer of flakes for flexible electronics, while SiO_2/Si proved to be a suitable candidate for device fabrication, offering robust adhesion between the WS_2 flakes and substrate [104].

The addition of functional groups like hydroxyl groups on 2D materials or substrate surfaces can also help to improve the resulting compatibility and bonding [105]. Rokni et al. [106] used atomic force microscopy (AFM) to directly and precisely measure the interfacial adhesion energies between tips coated with 2D materials interacting with SiO_x/Si substrates exposed to ambient air. They quantitatively characterized the nano-scale interlayer interactions of the 2D materials, such as graphite (G) over SiO_x substrates exposed in air, using different temperatures to understand the effect of airborne contamination and humidity. They verified that thermal annealing can effectively remove water and hydrocarbons from the air-exposed SiO_x surface, leading to higher interfacial adhesion energies. The usage of a pre-cooling treatment at −15 °C prior to annealing resulted in weaker interfacial interactions. This was explained by the hydrophilic nature of SiO_x that adsorbed a homogeneous and flat-water film with a thickness of 2–3 nm on its silanol (Si–OH)-rich surface when stored at −15 °C. This causes that the 2D crystal/water interface interaction governed the resulting interfacial adhesion energies measured, rather than measuring the interaction between 2D materials/SiO_x interface. In contrast, larger interfacial adhesion energies were measured for 2D materials/SiO_x interfaces when kept at room temperature prior to annealing. This was attributed to the presence of a mixture of ice-like monolayer/bilayer water structure and fully H-bonded to the silanol groups thus forming a trapped liquid-like film, bringing the 2D flakes into closer proximity with the SiO_x surface and enhancing the resulting interfacial adhesion energies. Although controlled annealing and pre-conditioning can temporarily improve adhesion, the re-adsorption of water and airborne contaminants is often inevitable under ambient conditions. This raises several concerns about the long-term reproducibility and stability of adhesion in real-world applications, especially for components operating in humid or fluctuating environments.

The presence of superficial functional groups on 2D coatings (e.g., for MXenes (-OH, -O, -F)) offers the possibility to form electrostatic interactions via hydrogen bonding with polymers such as polystyrene (PS) thus enhancing their interfacial adhesion [107–109]. For PS-MXene composites having a low MXene content, the composite assembling occurred through electrostatic interactions of the positive PS and the negative MXenes. In contrast, for higher MXene contents, van der Waals forces and hydrogen bonds were the main driving forces for assembling.

3.2. Chemical intercalation approaches

Nowadays, the necessity to contact graphene with a metal has motivated numerical studies of metal/graphene interactions, trying to improve the resulting interactions centered mainly on the intercalation

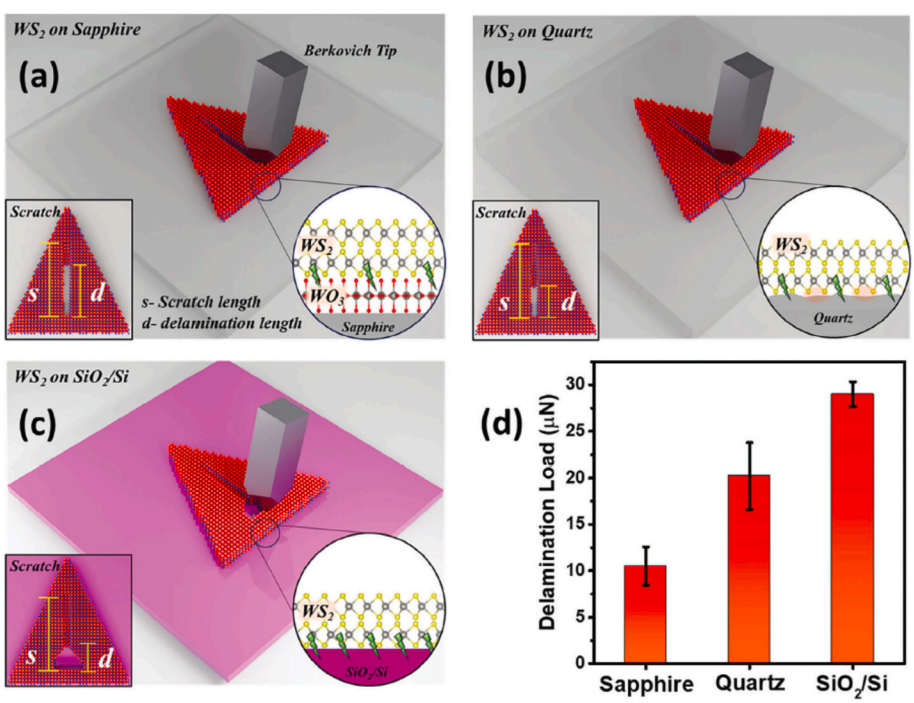

Fig. 4. Schematic representing the delamination process of WS_2 bilayers grown via CVD on three distinct substrates including (a) sapphire, (b) fused quartz and (c) SiO_2/Si, respectively, assessed by nano-scratch testing. (d) Summary of the substrate-dependent critical loads for WS_2 delamination. Reproduced with permission from [104].

process at the graphene/substrate interface. The possibility to intercalate molecules and atoms between 2D layers of stacked van der Waals heterostructures opens many research fields to create new materials with advanced properties [110]. In case of intercalating metal atoms, the deposited atoms do not stay at the graphene surface but can intercalate between the top layers and the substrate, which may happen at room temperature or can be induced by annealing, depending on the chemical nature of the metal [110]. Daukiya et al. focused on the functionalization of graphene by intercalating metal atoms and molecules between the layers, which created defects or changes in surface energy thus improving the resulting adhesion.

Intercalation does not always help to increase interfacial adhesion between a coating and a substrate. Yu et al. studied the deposition of a mica nanolayer (MNL) on a single layer graphene (SLG) supported by SiO_2 substrate to explore the effect of graphene layer on the adhesion between the MNLs and SiO_2 interface [111]. They demonstrated that the adhesion energy of MNLs/SLG/SiO_2 was considerably lower compared to the observed adhesion energy of MNLs/SiO_2 directly. They mentioned that the interfacial adhesion could theoretically derive from van der Waals, electrostatic and capillary forces, where the inclusion certainly affected the van der Waals force across the interface. They stated that screening of the electrostatic force from the potassium ions on the surface of MNL or the possible capillary effects of the water molecules at the interface contributed only to 40 % of the decrease, while the wrinkle effect of SLG caused about 60 % of the reduction. With a SLG in between MNL and SiO_2, the adhesion energy was decreased from 88.2 to 3.8 mJm^{-2}. This reveals that the wrinkles formed on the SLG considerably lowered adhesion across the interface, which can downgrade the observed friction and wear performance. Consequently, nano-scale adhesion can be tailored by controlling the wrinkle formation in graphene.

In the field of flexible electronics, 2D materials are deposited on elastic substrates to obtain 2D film/substrate with robust interfacial adhesion. Peng et al. [112] prepared two types of graphene/polymer samples with distinct interfacial adhesion, aiming to enhance the interfacial van der Waals interactions. On one hand, a graphene/PDMS (Gr/PDMS) sample was prepared, for which the graphene film was directly transferred to the top surface of the PDMS (a hydrophobic polymer made of methyl groups attached to a siloxane backbone). The methyl groups helped to shield the reactive siloxane backbone, giving PDMS its hydrophobic properties. On the other hand, a graphene/formvar-PDMS (Gr/Fo-PDMS) sample was prepared, for which an intermediate of formvar layer (thermoplastic resin composed of polyvinyl alcohol, formaldehyde, and polyvinyl acetate) was intercalated. Their experimental results showed two drastically different debonding rates, i.e., rapid snap-through debonding and more progressive crack propagation. The stronger adhesion, related to the more progressive crack propagation, was observed with formvar, which was explained because the polymer was added from a liquid phase and when it was fully coated between graphene and PDMS then it was cured, filling the vacancies and forming a more conformal contact between graphene and the substrate.

The usage of coupling agents such as aminosilane has been reported to enhance the adhesional strength in the interface of specific materials' combinations. Jia et al. [113] deposited $Ti_3C_2T_x$ or graphene oxide (GO) coatings on an aminosilane functionalized basalt fibre (BF) substrate to study the mechanical and interfacial properties in a BF poly(phthalazinone ether nitrile ketone) (BF/PPENK) composite. First, they deposited the 2D coatings onto the aminosilane functionalized BF surface by EPD. By assessing the dynamic contact angles of the different BFs, they verified an increase of the polar component (γ_p) of the surface energy for $Ti_3C_2T_x$ and GO-deposited BFs due to the functional groups of -OH and -F in $Ti_3C_2T_x$ as well as -COOH in GO, respectively. These functional groups increased roughness and wettability, which resulted in an improved interfacial adhesion between BF and PPENK.

Moreover, the incorporation of an adhesion layer, such as an interfacial hierarchical sandwich structure, has been reported on basalt fibre BFs-reinforced polymer composites. Yu et al. [114] reported the formation of an adhesion layer, such as a boron nitride/polyethyleneimine/MXene structure, using a facile layer-by-layer assembly, to strengthen and toughen the interfacial interaction. This approach helped to improve adhesion by enhancing the mechanical interlocking and chemical bonding of the composite. In this specific case, the bonding in the interfacial hierarchical structure was mainly through ionic and hydrogen bonds. They concluded that the interfacial sandwich structure improved the interface adhesion by augmenting the surface roughness through rigid boron nitride (BNOH) and $Ti_3C_2T_x$ as well as the chemical bonds through the bridge of the polyethyleneimine (PEI).

From a chemical point of view, the design of enhanced interfaces with better adhesion between a 2D material coating and its substrate should not only consider the respective interactions but also involve covalent bonding. Xiong et al. [115] grafted a GO coating onto CF using a fast and effectively thiolene click chemistry reaction using vinyl-terminated hyperbranched polyester (VHBP) as bridging agent. It has been demonstrated that the introduction of GO and VHBP greatly changed the surface morphology of the fibre thus improving the resulting wettability and surface energy. Additionally, the large number of functional groups of VHBP helped to enhance the mechanical interlocking and interfacial interactions between the fibre and EP resin (matrix) in the CF-VHBP-GO/EP composite [115].

3.3. Concluding remarks

Regarding **chemical approaches**, we have summarized examples that discuss the chemical affinity present in these interfaces, for which the nature of the atoms or functional groups used play the key role resulting in possible interactions and increasing adhesion. We also discussed the influence of airborne contamination or native oxides that induce changes in the interfacial interactions at the interface, which can partially improve the adhesion between surfaces due to the introduction of new bonds, such as electrostatic or hydrogen bonding. However, in other cases, the appearance of interfacial oxides resulted in a reduced adhesion owing to an increased effective substrate roughness, thereby reducing the area of contact. We have shed light on some examples of chemical functionalization using organosilane molecules, thus increasing adhesion by enhanced interfacial interactions. Some studies have been emphasized on how the intercalation of atoms, molecules adhesional layers, or the generation of heterostructures can be used to change the interfacial interactions.

4. Computational modelling

4.1. Substrate/coating adhesion

Computational analysis plays a critical role in understanding and elucidating the mechanisms underlying the adhesion of 2D materials on substrates and in designing application-specific solutions. A pivotal application concerns tribology, wherein 2D materials are employed as solid lubricant coatings. Experimental evidence, including AFM measurements, verify that the adsorption of even a single 2D flake (layer) on solid substrates can greatly reduce friction [116]. A preliminary attempt to explain this phenomenon was made by Klemenz et al. through classical MD simulations considering graphene on platinum [117]. They proposed that the ability of graphene to reduce the coefficient of friction (COF) originates from its high load-bearing capacity, which makes the coated surface more resistant to mechanical penetration by asperities. This characteristic remains stable until the layer exhibits signs of defect and/or rupture.

However, this explanation is incongruent with experimental findings, which demonstrated that graphene patches and even small flakes function effectively as lubricants [118,119]. This implies that accurately capturing the lubricating behavior of such nanoscale flakes remains a computational challenge. Most current models rely on idealized

geometries or periodic boundary conditions, which may not fully represent edge effects, flake-substrate misalignment, or dynamic detachment processes observed experimentally. An alternative interpretation was advanced by means of first-principles simulations of graphene on iron [120]. The authors demonstrated that the lubricating properties of graphene reside in its chemical stability. When graphene binds to a reactive surface, it reduces its surface energy, rendering the passivated surface almost inert and thereby minimizing its adhesion to any counter-surface. Fig. 5 a and b illustrate that by covering one of two mated iron surfaces with graphene, the separation between the surfaces increases by 119 %, while adhesion decreases by 78 %. A similar coating on the counter-surface results in a further increase in the interfacial separation (129 %) and adhesion reduction (11 %). This implies that the key lubricating effect relates to the reduction of the surface energy. Notably, Fig. 5 c shows that the interaction energy between two iron surfaces covered with graphene (black curve) closely resembles that of two isolated graphene layers (red curve), suggesting that the graphene coating completely screens the strong metal-metal interactions (blue curve). This explanation can be extended to account for the lubricating properties of other layered materials, such as boron nitride, and transition metal dichalcogenides (e.g., MoS_2), all of which exhibit low surface energies, though not sharing graphene's high load-bearing capacity.

In many applications, special attention has been dedicated towards graphene and its attachment to an underlying Si or SiO_2 substrate. Yang et al. [121] used MD simulations to investigate the interfacial behavior between graphene-based membranes with different distributions and concentrations of functional groups and protonated SiO_2 ($pSiO_2$) substrates. They found a positive correlation between the oxygen concentration of the membranes and the interfacial adhesion. In this regard, high oxygen concentrations and functional groups at the edges were shown to enhance the interfacial adhesion, where smaller sheets are more firmly adsorbed on the substrate.

The adhesion of 2D materials to solid substrates is crucial not only for tribology, but also for many other technological applications such as electronic devices, and catalysts. Therefore, it is essential to develop a comprehensive understanding of the factors that primarily influence the adhesion of 2D materials to substrates. A first principles high throughput study on solid-solid interfaces helped to identify these key factors by examining a database of adhesion energies for metallic heterointerfaces [122]. By applying the Sure Independence Screening and Sparsifying Operator (SISSO), a machine learning algorithm designed to extract effective material descriptors from potentially correlated features, and to identify an analytical equation that describes the relationship between these descriptors [123]. In this regard, the interfacial adhesion was found to be proportional to the geometric average of the surface energies of both constituents.

It is reasonable to assume that the surface energies of the constituents also govern adhesion in the case of layered-solid interfaces. For instance, an experimental study by Chang et al. found that the adhesion energy of as-grown graphene on nickel substrates is about 6.775 J/m^2, which is over seven times greater than that on copper. This difference can be attributed to Ni's higher surface energy [124]. In contrast, less reactive metals like Cu and Au interact with graphene mainly via van der Waals (vdW) forces, resulting in physical and not chemical adsorption [125,126]. Stronger chemical interactions may enhance adhesion and reduce wear, but can also increase tribochemical degradation under certain conditions, revealing a non-linear relation between stability and reactivity.

First-principles calculations represent a valuable tool to estimate the adhesion of 2D layers on solid substrates. However, some uncertainties remain in situations, for which the attraction is governed by weak vdW interactions. Indeed, the adhesion energy values reported in literature for 2D materials on various substrates show a notable variability depending on the computational method employed. For instance, the graphene/Al(111) interface yields adhesion energies ranging from 0.18 to 0.41 J/m^2 (Table 2), highlighting how method-dependent these values are. This variability stems from the intrinsic difficulty in accurately describing non-local correlation effects within standard DFT frameworks like those based on the local density approximation (LDA) of the exchange correlation (xc) functional or gradient-corrected xc functionals, such as that proposed by Perdew, Burke, Ernzerhof (PBE) [127]. While LDA tends to overbind, and, in some cases, this tendency mimics dispersion-like behavior, yet this effect is system-dependent and not systematic, PBE yields too repulsive energy-distance curves. Consequently, it is often corrected by adding a parametric term to

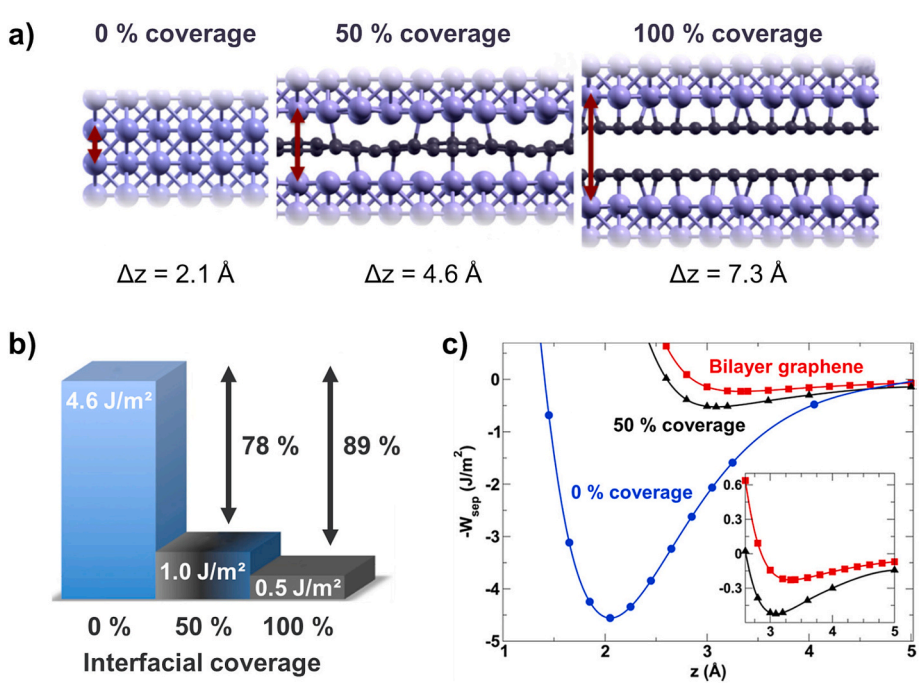

Fig. 5. Interaction of iron surfaces covered by graphene. Effects on (a) interfacial separation, (b) adhesion energy, and (c) surface-surface interaction energy as a function of interfacial coverage. Reproduced with permission from [120].

Table 2
Summary of adhesion energies of various 2D materials on different substrates calculated using RPA, and DFT with different approximations for the exchange correlation functional. The graphene (MoS_2) adhesion energies reported in eV/atom (eV/MoS_2-unit) in literature have been converted in J/m^2 assuming an area per atom (MoS_2-unit) equal to 2.62 10^{-20} m^2 (8.75 10^{-20} m^2).

2D material / Substrate	Adhesion (J/m^2)			
	RPA	PBE + vdW	vdW-DF	LDA
Graphene/Al(111)	0.32[a]	0.41[b]	0.21[c]	0.18[a]
MoS_2/Al(111)			0.55[f]	
Graphene/Ni(111)	0.43[a]	0.43[b]	0.27[c]	0.31[a]
MoS_2/ Ni(111)			0.93[f]	
Graphene/Co(0001)	0.40[a]	0.43[b]	0.18[c]	0.01[a]
MoS_2/Co(0001)			1.04[f]	
Graphene/Pd(111)	0.55[a]	0.51[b]	0.30[c]	0.26[a]
MoS_2/ Pd(111)			0.98[f]	
Graphene/Pt(111)	0.51[a]	0.50[b]	0.32[c]	0.22[a]
MoS_2/ Pt(111)			0.78 [f]	
Graphene/Cu(111)	0.42[a]	0.48[b]	0.27[c]	0.24[a]
MoS_2/ Cu(111)			0.73[f]	
Graphene/Ag(111)	0.48[a]	0.46[b]	0.26[c]	0.18 [a]
MoS_2/ Ag(111)			0.64[f]	
Graphene/Au(111)	0.58[a]	0.51[b]	0.29[c]	0.21[a]
MoS_2/ Au(111)		0.60[f]		
Graphene/Ti(0001)				2.53[c]
MoS_2/Ti(0001)		0.93[f]		
Graphene/TiO_2(110)				0.58[c]
Graphene/Al_2O_3(0001)				0.59[c]
Graphene/ SiO_2-OH-SiO_2		0.17–0.23[d]		0.09–0.115[d]
Graphene/Fe(110)		0.89[e]		
MoS_2/ Fe(110)		1.60[e]		
Phosphorene/Fe(110)		2.64[e]		
Ti_2CO_2/Fe(110)-Fe_2O_3		1.3–0.3[g]		
Ti_2COH_2/Fe(110) -Fe_2O_3		1.2–2.2[g]		
Ti_2CF_2/Fe(110) -Fe_2O_3		0.2–0.2[g]		
$Ti_2C(O,OH,F)_2$/ Fe(110)		0.4[g]		

[a] [131].
[b] [132].
[c] [133].
[d] [134].
[e] [135].
[f] [136].
[g] [54].

describe the long-range attraction, as in the dispersion-corrected DFT-D methods proposed by S. Grimme [128]. Even non-empirical methods, such as vdW-DF, sometimes fail to accurately predict weak layer-substrate interactions.

Higher-level methods such as the Random Phase Approximation (RPA) [129,130] offer a more accurate and parameter-free description of long-range correlation effects, including dispersion forces, and are thus considered more reliable. However, their computational cost remains prohibitively high for large or complex systems, limiting their widespread application. Furthermore, even among dispersion-inclusive methods, the choice of vdW functional can lead to notable discrepancies. For instance, for graphene/Pt(111), the vdW-DF value is 0.32 J/m^2, whereas the PBE + vdW value is 0.50 J/m^2 (Table 2). Overall, these inconsistencies highlight the need for a careful benchmarking of adhesion energies against high-level theoretical methods or experimental data whenever available. Nevertheless, despite these challenges, first-principles calculations are often capable to capture the adhesion trends reliably - for example, when comparing the adhesion of different layered materials, such as graphene and MoS_2, on the same substrates or the same layer on different substrates.

A comparison between adhesion energies for graphene and MoS_2 reveals distinct trends that reflect the different electronic and structural properties of both prototypical 2D materials. Graphene, being a purely sp^2-hybridized carbon sheet with delocalized π electrons, interacts weakly with most substrates via vdW forces. In contrast, MoS_2 exhibits notably stronger adhesion to a wide range of substrates (Table 2), due to its higher polarizability and the potential for more complex interactions, such as a partial charge transfer and d-orbital hybridization with metal surfaces. These effects are particularly evident on reactive substrates like Pd(111), Pt(111), and Fe(110), respectively, for which chemisorption may play a role.

The adhesion energies calculated for graphene on different metals have shown that transition metals with low d-shell occupancy (e.g., Ti, V, Cr, Fe, and W) exhibit higher adhesion due to orbital hybridization with graphene's delocalized π-electrons. In contrast noble metals (e.g., Pt, Au, and Ag) show a greatly reduced adhesion, typically below 1 J/m^2, due to their weaker interaction with graphene. Post-transition and alkaline earth metals exhibit even lower adhesion, for which d-electron-free metals (d = 0) tend to show the weakest interactions. For transition metals interacting with MoS_2, two distinct groups based on d-shell occupancy have been identified. On the one hand, metals with less than 50 % d-shell occupancy exhibit adhesion energies greater than 2 J/m^2, while, on the other hand, those with higher d-shell occupancy show values around 1.5–1.8 J/m^2 at similar interface distances (Table 2 and unpublished, ongoing research work by Righi et al.).

MXenes tend to show a markedly stronger adhesion to substrates compared to more weakly interacting 2D materials like graphene (Table 2). Their higher adhesion energies are governed by the surface terminations. Ti_2CF_2 shows weak physisorption, dominated by vdW forces, with minimal charge transfer and large interfacial distances. In contrast, Ti_2CO_2 and $Ti_2C(OH)_2$ chemisorb on iron, leading to stronger adhesion due to charge transfer: Ti_2CO_2 oxidizes the surface, while $Ti_2C(OH)_2$ reduces it. A similar behavior has been observed for hematite, with $Ti_2C(OH)_2$ forming short, strong hydrogen bonds and transferring both charge and mass (H atoms).

As the adhesion of a 2D material to a substrate is highly affected by the latter's surface energy, the chemical modification of the substrate represents a potential strategy to control the resulting adhesional strength. As demonstrated by high-throughput first-principles calculations, surface chemistry modifications have been shown to be highly effective in altering metal-on-metal adhesion. For instance, covering the surface of highly reactive transition metals (such as W, Cr, Mo, Fe) with a ¼ monolayer (ML) of sulphur atoms reduces their adhesion to a homogeneous counter-surface by 46 %. When using fluorine, this reduction reaches 63 %. This observed tendency is particularly interesting and important when considering that S and F are key components of lubricant additives and can relate to the friction reduction under boundary lubrication.

Conversely, other elements can enhance interfacial adhesion. For instance, ¼ ML of boron on a low-energy surface like Au (111) tends to increase its adhesive strength by 24 %, while this enhancement can reach 94 or even 175 % with 50 and 100 % surface coverages, respectively [137]. The effects of certain chemical modifications, such as those involving F and B, appear to be universal, as they also influence the adhesion of metal-semiconductor interfaces in the same manner [138]. Consequently, these strategies can be applied to modulate the adhesion of 2D materials to substrates, with potential benefits for various applications. This holds particularly true for tribology, for which one key challenge hindering the widespread use of 2D solid lubricants in macroscopic applications, is the low resistance to peel-off/wear-off during rubbing under elevated loads and contact pressures. In this context, chemical modifications of the underlying substrate can help to enhance the adhesion between 2D layers and the substrate without compromising the layer slipperiness.

Considering this aspect from the nanomaterial perspective reveals that chemical modifications of the 2D layered materials to enhance their adhesion to solid substrates also increase the interlayer adhesion. For instance, the adhesion of a titanium carbide layer on iron is shown to increase by 644 % (719 %) when replacing fluorine with hydroxyl (oxygen) terminations (Fig. 6) [54]. However, as discussed in the next section, an increased content of O and OH atoms for MXene terminations causes an increase of the interlayer binding energy and shear strength.

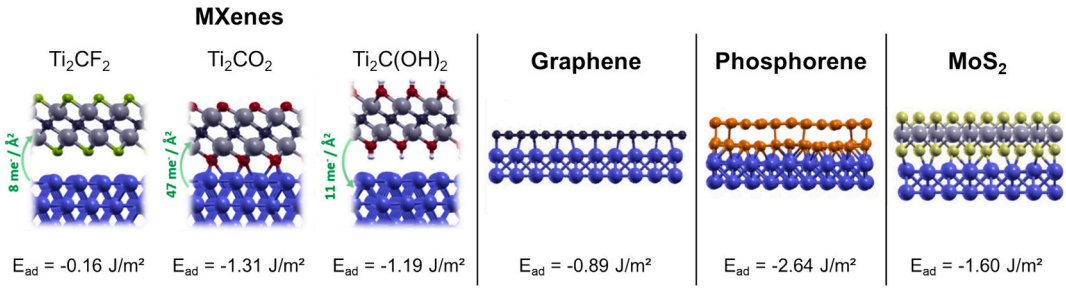

Fig. 6. Optimized configurations and adhesion energies of different 2D materials on the (110) surface of iron. Reproduced with permission from [54,135].

The presence of defects can increase the surface energy of a layer, thereby enhancing its adhesion to the underlying substrate. A typical example relates to graphene, which can feature highly reactive dangling bonds at its edges. For instance, first-principles calculations have estimated that unsaturated edges in graphene increase the adhesion of a graphene ribbon to iron by 63 % compared to a similar portion of defect-free graphene [120]. However, while defects enhance adhesion, they also increase the local reactivity and disrupt lattice continuity, which can lead to elevated friction and wear under sliding conditions. Consequently, it becomes evident that using unsaturated graphene flakes as lubricants is not a viable option to obtain better lubricating properties, as evidenced by the well-known issue of graphite's lubricity degrading in vacuum or dry conditions. In these environments, the absence of passivating species causes graphite/graphene to transform from a solid lubricant to a sticky medium [139]. Strategies, such as employing passivated graphene flakes or aromatic molecules that lose their lateral groups and expose dangling bonds only upon contact with a substrate, can help to effectively address the challenge of achieving strong substrate-molecule adhesion while maintaining weak molecule-molecule interactions [119].

Since layered materials can easily undergo deformations, their adhesion on substrates can be affected not only by the surface energy, but also by structural changes occurring upon adsorption. An example is offered by phosphorene, which almost splits into two atomic-thick layers upon deposition on iron due to the involved charge transfer to form new P-Fe bonds thus weakening the vertical P-P bonds (Fig. 6). Therefore, the resulting adhesion energy of phosphorene on iron is much higher than that of graphene and MoS_2. Based on the pure evaluation of the surface energies, an opposite trend would have been inferred, which underlines the important role of structural changes on the resulting adhesion behavior (the interlayer adhesion of phosphorene is merely 15 % higher than that of graphene and 20 % lower than that of MoS_2) [135].

Another important factor affecting the adhesion of 2D materials to substrates is their in-plane strain, which can influence the adhesional strength by modifying electronic interactions between the layer and the substrate [140]. For instance, DFT studies have shown that compressive strain enhances graphene adhesion by increasing the orbital overlap, whereas tensile strain tends to weaken adhesion by altering the electronic band structure of graphene [141]. In real-world applications, strain can originate from mismatched thermal expansion between the coating and substrate, while residual stresses can stem from deposition, or mechanical loading during operation. Consequently, the assessments and control of these sources of strain could offer new approaches to tailor interfacial adhesion dynamically or improve coating reliability under service conditions.

Out-of-plane deformations due to low bending stiffness of 2D materials often appear on weakly adherent substrates. A layer puckering was observed when sliding an AFM tip above different layered materials, which is sensitive to the number of layers and to the considered 2D materials. This effect becomes less pronounced as the number of layers increases, thus reducing friction. Different hypotheses were proposed to explain the higher friction measured for the puckered layers. Recent first principles calculations have demonstrated that graphene puckering is accompanied by a change in the hybridization of the carbon atoms from sp^2 to sp^3-like, which went hand in hand with a corresponding increase of reactivity due to the appearance of dangling bonds [142].

The relative orientation of a 2D layer with respect to the substrate is also an additional factor that influences adhesion, as it changes the local commensurability and relative position of the mated atoms. First-principles studies have demonstrated that specific rotation angles can lead to the formation of moiré superlattices, which alter the charge transfer mechanisms and adhesion properties [143]. For instance, graphene on Ru (0001) exhibits strong adhesion at specific moiré configurations, while graphene on Cu (111) shows weaker adhesion [144]. However, it is worth pointing out that a systematic study of the effect of the relative orientation is yet to be presented. In computational simulations, larger supercell size captures more realistic strain relaxation effects, leading to more accurate adhesion determination [145]. However, increasing the supercell size also increases the computational cost, which asks for an appropriate balance between accuracy and efficiency. The advent of machine learning potential offers a new opportunity for reaching this balance [146].

4.2. Interlayer adhesion (slipperiness)

The interlayer interaction energy is a key property of 2D materials, which can be accurately modeled ab-initio using DFT (quasi-static) calculations. Its value can be directly compared with exfoliation energies from experimental data. The prediction of this energy, along with a deeper understanding of what characterizes interlayer interactions and how they can be tuned, is crucial to better control delamination and multilayer deposition processes.

The interlayer adhesion or interlayer binding energy can be calculated based on its definition as the negative of the work of separation, which is the energy necessary to separate two adjacent layers: $E_{ad} = -W_{sep} = (E_{bilayer} - 2E_{mono})/A$, for which $E_{bilayer}$ is the energy of the two interacting layers at the equilibrium distance, E_{mono} stands for the energy of a single layer, and A resembles the contact area. The variation of the adhesion energy as a function of the relative lateral position of two layers, described by a potential energy surface (PES), determines the tribological properties of the bilayer such as the interlayer shear strength. This implies that the static friction force per unit area can be directly estimated from the derivative of the PES profile along a specific direction. Consequently, the PES corrugation represents the maximum energy barrier that needs to be overcome during sliding of two adjacent layers [147]. A decrease in corrugation corresponds to an increase in the slipperiness of the nanomaterial.

The magnitude of adhesion (PES corrugation) is determined by the electronic charge redistribution occurring, when two layers are brought into contact (laterally displaced). Wolloch et al. [148] directly connected the charge redistribution, summarized in a well-defined figure of merit, to the adhesion energy, maintaining a linear fit across different types of materials, including 2D materials.

One of the most challenging aspects of DFT calculations of interlayer adhesion is the absence of (strong) chemical bonding. Interlayer interactions are dominated by weaker forces, including hydrogen bonding, dipole-dipole interactions, and London dispersion forces. Capturing the elusive dispersion forces in materials with complex electronic structures using parameter-free methods remains computationally demanding. More affordable semi-empirical approaches can be successfully used after fine-tuning parameters through benchmarking against higher level theory methods, such as the Møller-Plesset perturbation theory and the random phase approximation. Within this approach relevant understanding of the nature of layer interactions can be obtained. For instance, Marquis et al. [54] demonstrated that MXenes' interlayer adhesion is completely governed by the layer termination: the innermost atomic planes do not notably contribute to the interaction. Consequently, they theoretically verified that the substitution of carbon with nitrogen does not change interlayer adhesion. The chemical composition of the layer terminations, which can be tuned by varying etching conditions, determines the adhesional strength. As can be seen in Fig. 7 a, F−, and O − terminated layers weakly interact through dipole-induced dipole interactions, while dipole-dipole interaction are present for -OH terminated layers, which increase the interlayer adhesion. The latter is further enhanced by the presence of -O and -OH groups on the faced layers, which form hydrogen bonds. An increase in the interlayer adhesion is usually accompanied by an increase of the PES corrugation, which reduces the layer slipperiness (Fig. 7 b). The PES corrugation also increases when a load is applied since the Pauli repulsion at short distances makes the layer stacking with on-top atoms energetically unfavorable [149].

Point defects can also lead to an increased interfacial adhesion due to enhanced local reactivity, which negatively affects the material's lubricity. Consequently, friction has been shown to increase monotonically with the vacancy density [150].

The interlayer adhesion and slipperiness are also affected by intercalated atoms and molecules. In case MoS_2, it was shown by ab-initio MD simulations that intercalated water molecules produce a viscous friction force between the layers, consistently with the observed degradation of the lubricating properties of MoS_2 by air humidity [151]. Interestingly, Stella et al. [152] correlated the friction increase with the tendency of water molecules to form ordered clusters that align with the substrate at low coverage, creating an interlocking effect between the layers. This feature is further enhanced with increasing load, which, by increasing PES corrugation, tends to trap molecules in the energy minima. This insight is particularly relevant for real-world applications, where residual humidity is difficult to eliminate and may critically affect the long-term tribological performance of 2D coatings. Interfacial incommensurability, and the corresponding improvement in material lubricity, can only be recovered when, at full coverage, water islands reach a critical size, for which intermolecular hydrogen bonding interactions can overcome the lattice mismatch with the potential corrugation. This highlights that promoting interfacial incommensurability is an interesting design strategy to enhance the lubricity of solid lubricants, particularly at the nano- and micro-scale.

4.3. Concluding remarks

Based on this section, it becomes evident that controlling the adhesion of 2D materials or thin films to solid substrate is a complex task that can benefit from the integration of experiments with computational tools. As outlined, the adhesion energy of a flat interface is proportional to the geometric mean of the surface energies of both contacting surfaces. Ab-initio calculations can provide accurate estimates of the surface and adhesion energies as well as predict how these energies change with chemical functionalization, or by introducing reactive defects such as dangling bonds. Moreover, first-principles high throughput calculations permit the automatized screening of many material pairings and modifications. However, the adhesion between two surfaces also depends on the structure of both contacting surfaces, particularly on the deformation that 2D films can experience due to mismatch with the substrate. For instance, it is known that 2D materials can exhibit large out-of-plane deformations, especially when adhesion to the underlying substrate is weak. Other effects related to the structure's influence on adhesion include the reciprocal orientation of the film and the substrate. By rotating a 2D material relative to the substrate, moiré patterns/configurations can be formed, which can result in different adhesion values. To account for these effects, more computationally demanding calculations needs to be conducted.

5. Concluding remarks and scientific outlook

As holistically discussed and outlined, 2D coatings have shown an exceptional friction and wear performance, which has been mainly verified in basic laboratory experiments. In this regard, graphene-based coatings have gained prominence for their ultra-low friction states as experimentally demonstrated by [29,153]. Coatings made of MoS_2 have been shown to be excellent solid lubricants under dry and vacuum conditions thus making them well suited to be applied in aerospace

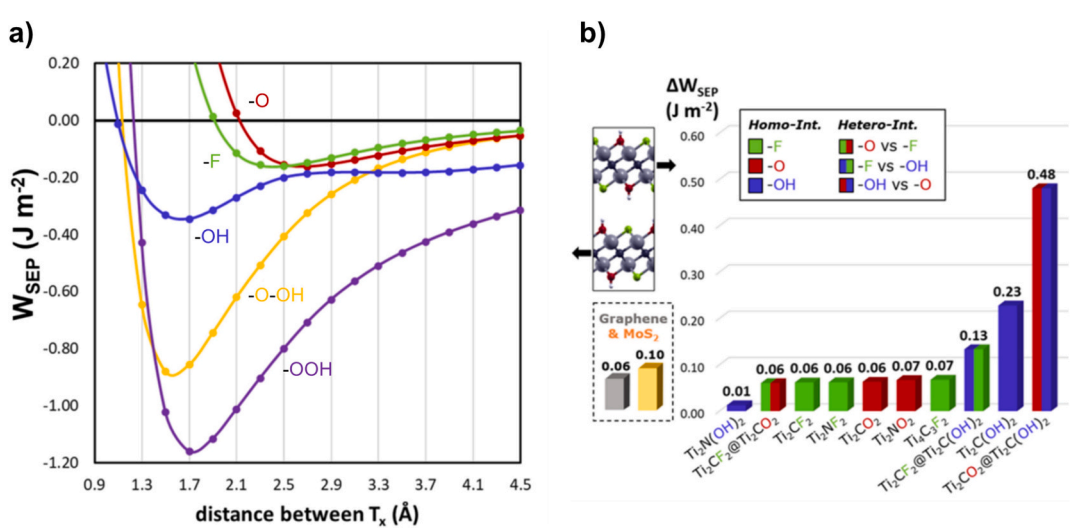

Fig. 7. Interlayer energy as a function of separation (a) and PES corrugation (b) for MXene interfaces with -F, -O, and -OH terminations. The interlayer adhesion energy is given by the minimum of the curves. Reproduced with permission from [54].

applications [25]. In contrast, MXene-based coatings (mainly $Ti_3C_2T_x$, among others) stand out for the outstanding wear-resistance and durability due to the involved tribo-film formation involved pressure-induced tribo-chemical reactions (mechanochemistry) [154,155]. Our literature review offers a concise overview about strategies aiming at enhancing the adhesion of 2D coatings to various substrates. It combines approaches from physical surface engineering, chemical functionalization, and theoretical modelling. Our key contributions include a systematic comparison of surface preparation techniques, a critical analysis of adhesion layers and their mechanical implications, and an exploration of how chemical methods can strengthen interfacial bonds. Additionally, the integration of DFT and MD simulations as well as the potential of ML are highlighted as powerful tools to predict and optimize the interactions between 2D coatings and the underlying substrates. Consequently, we can envision a clear roadmap to design robust and wear-resistant 2D coatings that are suitable for demanding tribological applications.

Although bearing tremendous potential for applied research and transfer to industrial scale, the usage of 2D coatings in mechanical components and the transfer of the gained fundamental knowledge to applied conditions remain rather scarce. One of the greatest hurdles to be overcome in this context relates to rather poor coating-substrate interfaces with limited adhesion, which may be acceptable in lab-experiments but are detrimental in mechanical components working under elevated contact pressures. Consequently, to make 2D material coatings are real success story, particular attention must be dedicated towards the design and tailoring of the involved coatings-substrate interface with a special emphasis on cleanness, mechanical integrity and chemical compatibility. In this regard, several pathways including physical and chemical approaches as well as input from theoretical modelling can help to design and further optimize well-adherent 2D coatings thus creating mechanically strong and robust interfaces.

In terms of **physical approaches**, we have outlined the critical role of surface quality, surface topography and cleanness to design mechanically robust, well-adherent coatings. These aspects must be precisely matched with the material to be deposited and deposition approach used. After having optimized roughness and chemistry aspects, surface treatments can be combined with the usage of thin metallic adhesion layers deposited by PVD to further boost the resulting adhesion/interfacial strength. It is important to consider that adhesion layers create additional interfaces, which can be weak points in the coating system thus inducing early delamination, crack initiation and propagation as well as catastrophic wear and coating failure. Therefore, special attention must be given to the mechanical and thermophysical properties of the adhesion layers used. Mismatches in mechanical properties (hardness, strength and ductility) and thermal properties (e.g., coefficient of thermal expansion) should be avoided to design strong and robust interfaces thus minimizing the risk for coating failure.

With respect to **chemical approaches** to improve the interfacial adhesion between 2D coatings and the underlying, we have discussed the critical role of the nature of atoms and functional groups as well as airborne contamination and native oxides regarding their effect to influence the resulting interfacial interaction. We have also outlined the potential of chemical functionalization using organosilane molecules and intercalation agents, thus increasing adhesion by enhanced interfacial interactions. In this regard, the appropriate selection of these atoms, molecules and adhesion layers is essential since they can generate new bonds increasing adhesion strength. A highly prospective but rather unexplored strategy relates to click chemistry reactions to increase mechanical interlocking and interfacial interactions thus boosting interfacial adhesion. From a chemical point of view, selected chemical functionalization based on click-chemistry is the key strategy to be followed when considering the interactions that can be induced by the tailor-made new functional groups. The overall concept of click-chemistry can be also combined with the usage of adhesion layers or the design of surface textures to increase the real are of contact thus combining physical and chemical approaches. We anticipate that exactly this combination can boost the resulting interfacial strength and adhesion thus creating mechanically strong and robust interfaces, which are highly suitable solid lubricant coatings to be applied under real-world conditions.

Regarding **numerical approaches**, ab-initio calculations can provide accurate estimates of the surface and adhesion energies as well as predict how these energies change with chemical functionalization, or by introducing reactive defects such as dangling bonds. Databases produced by high-throughput calculations can benefit from an analysis by means of Machine Learning (ML) algorithms to identify general trends and predict novel, more adhesive layers compositions. To account for effects such as deformation, mismatches or rotation, first-principles calculations are no longer sufficient because they involve larger scales than those accessible with DFT calculations. ML potentials (MLPs), which are essentially neural networks trained to reproduce the energies and forces computed from first principles, represent a possible solution to overcome this limitation. Their use and implementation in MD allow for a notable increase of the size- and time-scale of the simulated systems, while maintaining the accuracy of first-principles calculations. In this regard, it is important to emphasize that ML-based MD has been successfully applied in various fields of materials science. However, in tribology, its use is still in its infantries and limited to a few examples, which nonetheless demonstrate its promising potential in this area [146,156,157]. In the assessment of adhesion of 2D materials and thin films to solid substrates, the use of this technique could prove to be a turning point. It would enable the integration of the description of chemical interactions, which are crucial for determining interfacial adhesion, while also accounting for physical effects such as deformations, surface commensurability, stains, moiré patterns, which typically require the simulation of very large cells. Consequently, this would reconcile two currently incompatible aspects with the use of existing simulation techniques. ML-MD could also be used to conduct *in-silico* experiments, such as nano-scratch testing [158], which are performed in laboratories to measure the adhesion of thin films to substrates, providing initial hints on the performances of selected coatings in different operando conditions.

6. Applied perspective and outlook

The beneficial and exciting friction and wear properties of coatings made of 2D materials or their hybrids have already been partially transferred to mechanical components working under realistic conditions [70,159,160]. Due to the outlined interfacial and adhesion problems, the overall success of this approach is still limited. However, it is important to emphasize that some positive results have already been reported in literature for graphene- [161] [162], MoS_2- [163] [164] or MXene-based [165–167] coatings, while MoS_2 coatings deposited by PVD have demonstrated the most advanced progress in this field. Regarding the use of graphene and GO, these materials have been used as reinforcement agents to introduce the performance of polymeric bearings [168]. Concerning roller bearing applications, it has been shown that graphene nanoplatelets notably decrease the observed frictional torque under an applied contact pressure of about 1–1.5 GPa [161]. For thrust roller bearings, the synergetic effect of surface texturing and graphene has also been verified leading to an additional 30 and 35 % friction and wear reduction, respectively [162]. Related to the use of MoS_2 coatings, the first successful reports of the applications of MoS_2 in real machine components date back to the early 90s. In aerospace application, sputtered MoS_2 coatings were successfully tested in gimbal bearings, which requires low frictional torques and extended durability [163]. Similarly, MoS_2 coatings deposited on the bushes and shafts of journal bearings induced a beneficial friction and wear performance under air and vacuum conditions [164]. Beneficial effects were also verified for MoS_2 coatings used in thrust ball bearings [164], ball bearings and [169] and gears [170]. The first knowledge transfer for

MXene coatings to mechanical components was demonstrated by Marian et al., which demonstrated an enhanced performance for MXene-coated thrust ball bearings [165] and rolling bearings [166]. In this regard, the study assessing the performance of multi-layer $Ti_3C_2T_x$ coatings in rolling bearings is particularly worth mentioning. Compared to MoS_2 and diamond-like carbon (DLC) coatings, the MXene coatings reduced the wear of the bearing washers by 94 %. While showing a similar frictional performance compared to MoS_2 and DLC coatings, the MXene coatings extended by useful wear lifetime by 30 and 50 %, respectively. Hybrid $MoS_2/Ti_3C_2T_x$ have depicted promising results when applied in dry-running journal bearings [167], which underlines the effectiveness of hybrid solutions in applied scenarios. For a thorough and complete understanding of the use of 2D materials as coatings, lubricant additives, and fillers in mechanical components, interested readers are referred to [70,159,171].

Considering the potential applications and the challenges addressed in this article, we summarize and analyze several physical and chemical strategies aimed at enhancing the adhesion of 2D coatings. However, it must be emphasized that not all approaches are equally mature or readily applicable. Some methods have already shown consistent results under controlled conditions, while others remain highly promising but require further validation under more demanding, real-world environments. Advancing towards industrial implementation will, therefore, demand not only continued exploration of emerging strategies, but also a focused prioritization of those with greater scalability, mechanical robustness, and compatibility with conventional deposition methods. By integrating high-throughput screening, interface engineering, and interfacial chemistry within a predictive, experimentally validated framework, the design of adhesive and wear-resistant 2D coatings can finally move from fundamental research to industrial relevance, reshaping the future of solid lubrication technologies. Taking all these aspects together, it becomes evident that the design strategy and rationale to enhance the interfacial adhesion between 2D materials and the underlying substrate relate to the combined use of high-throughput screening (selection of the most suitable material pairings) as well as coupling physical and chemical approaches (surface engineering with chemical functionalization). This will help to establish mechanically robust interfaces to make 2D coatings a real success story in tribology and solid lubrication.

In terms of future prospects, we anticipate that the concepts discussed in this article can be applied practically by progressing from laboratory-scale experiments to industrial-scale processes. Our focus will be on prioritizing research regarding scalable deposition techniques, customized surface treatments, and predictive modelling tools that combine chemical and mechanical optimization to achieve promising results. Advances in these areas can accelerate the development of next-generation 2D coatings with enhanced adhesion, durability, and multifunctionality, ultimately redefining their role in tribology and solid lubrication technologies.

Notes

Authors approve ethics in publishing and consent to participate thereof.

The authors declare consent for publication.

CRediT authorship contribution statement

Andreas Rosenkranz: Writing – review & editing, Writing – original draft, Validation, Supervision, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization. **Monica Soler:** Writing – review & editing, Writing – original draft, Investigation, Funding acquisition, Formal analysis, Conceptualization. **Guido Boidi:** Writing – review & editing, Writing – original draft, Formal analysis, Conceptualization. **Bettina Ronai:** Writing – review & editing, Writing – original draft, Formal analysis. **Markus Varga:** Writing – review & editing, Funding acquisition, Formal analysis. **Fabiola Pineda:** Writing – review & editing, Writing – original draft, Investigation, Funding acquisition, Formal analysis. **Maria Clelia Righi:** Writing – review & editing, Writing – original draft, Investigation, Funding acquisition, Formal analysis, Conceptualization.

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Declaration of competing interest

The authors declare no competing interests.

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Data availability

No data was used for the research described in the article.

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