



Historical perspective

Roadmap for 2D materials in biotribological/biomedical applications – A review

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ABSTRACT

The human body involves a large number of systems subjected to contact stresses and thus experiencing wear and degradation. The limited efficacy of existing solutions constantly puts a significant financial burden on the healthcare system, more importantly, patients are suffering due to the complications following a partial or total system failure. More effective strategies are highly dependent on the availability of advanced functional materials demonstrating excellent tribological response and good biocompatibility. In this article, we review the recent progress in implementing two-dimensional (2D) materials into bio-applications involving tribological contacts. We further summarize the current challenges for future progress in the field.

1. Introduction

Numerous medical devices are employed to treat or alleviate various diseases and injuries as well as anatomy and physiological process support. The global medical device market is expected to grow from nearly \$455 billion in 2021 to \$658 billion in 2028 due to an increasing number of healthcare facilities, the elderly population, healthcare expenditure, and technological advances [1]. Thereby, their safety and effectiveness represent major concerns critical for effective clinical application. All medical devices inevitably contact and interact with the human body, which is why their surfaces and interfaces are essential. The design of functional contacting surfaces comes into focus to fulfil the desired functions adequately. As illustrated in Fig. 1, this covers the interfaces between dental implants and bone or tissue [2], contact lenses and eye/eyelid [3], as well as surgical or cardiovascular devices and tissue [4]. Interactions also occur between components of biomedical devices, including metallic bone fracture fixations [5], the articulating surfaces of the femoral and tibial components in total knee replacements, or the femoral head and the acetabular cup in artificial hip joints [6,7].

Medical devices' failures or malfunctions often relate to processes/problems occurring in these interfaces, including excessive wear (prosthetic interfaces), tribo-corrosion and fretting (dental implants), loosening of bone fixation devices, or tissue damage due to high friction of invasive surgical or cardiovascular devices [5]. Therefore, the biotribological behavior of such systems plays a crucial role in prolonging their safe and reliable operation. The current state-of-the-art has been summarized in several reviews from different points of view and for various biomedical applications [5,8–10], including artificial joints [6,7,11,12], cardiovascular devices [4], dentistry [2], and the role of lubrication [13–15]. Apart from theoretical/numerical approaches to come up with optimized designs mainly devoted to *in-silico* trials for prostheses [16–22], current research is directed towards the development of low-friction, low-wear, and corrosion-resistant materials or surface modifications [7,23].

In this regard, the unique physical, chemical, structural, electrical, and optical properties as well as the large surface-to-volume ratio, tunable surface chemistry, and customizable functions make two-dimensional (2D) materials attractive for many biomedical/biotribological applications [24–27]. Since the first isolation from graphite

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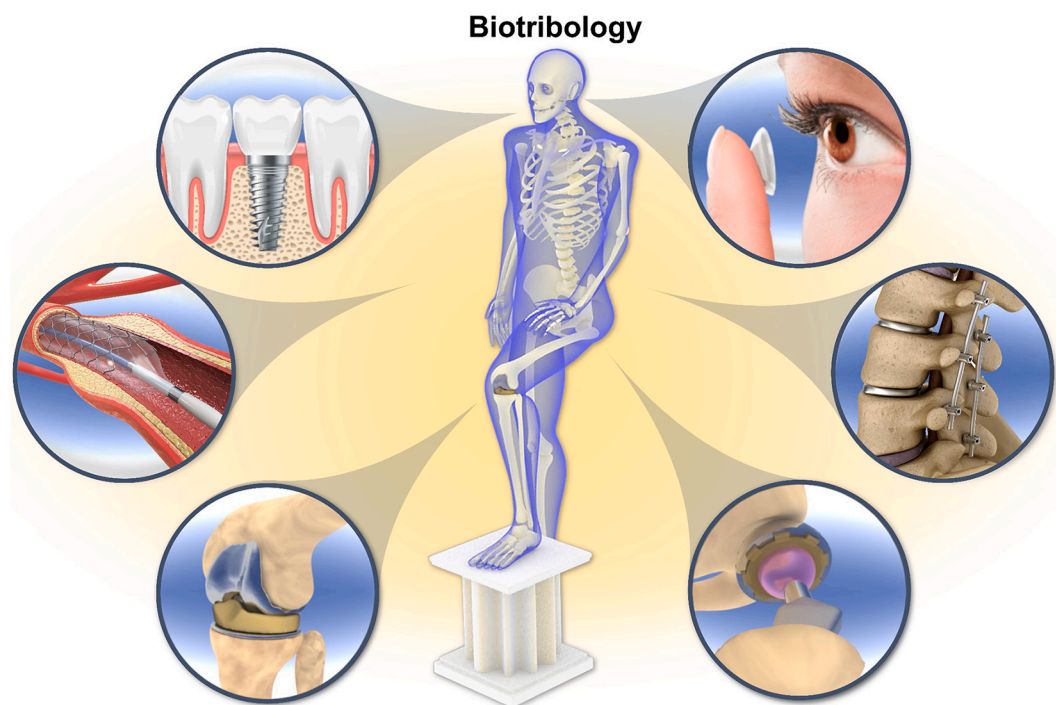


Fig. 1. Biotribological areas for application of 2D materials to tailor friction and wear: Dental implants, contact lenses, cardiovascular devices, bone fracture fixations, total knee and hip replacements.

using scotch tape [28], graphene is by far the most explored 2D material with applications in tissue engineering, biosensing, bioimaging, and gene and medication delivery [29–32]. Graphene and its derivatives, graphene oxide (GO) and reduced graphene oxide (rGO), have their own set of advantages and uses. Compared to graphene [16], GO is the oxidized form of graphene with increased hydrophilicity, water dispersibility, non-toxicity, and biocompatibility [33]. rGO has many of the same features as pure graphene but its fabrication is less expensive. Transition metal dichalcogenides (TMDs) are a family of 2D materials consisting of a layer of transition metal atoms sandwiched between two layers of chalcogen atoms [34]. Thereby, molybdenum disulfide (MoS_2) and tungsten disulfide (WS_2) represent the most prominent TMDs [35]. Because of their outstanding photoluminescence and adjustable bandgap as well as good mechanical characteristics, TMDs have been used in optical, electronic, and biological applications [25]. Despite their exceptional electrical, optical, thermal, and mechanical features, transition metal oxides (TMOs) have received less attention than other 2D materials [36]. Hexagonal boron nitride (h-BN) has structural similarities to graphene, with boron and nitrogen atoms forming covalent bonds within a hexagonal structure. Functionalized h-BN is a promising candidate for medication delivery as well as tissue engineering and live-cell imaging due to its good mechanical and chemical properties [26]. Black phosphorous (BP), for which each phosphorous atom is tetrahedrally bonded to three adjacent phosphorous atoms creating the honeycomb structure, finds its application in electronics and optical applications due to its anisotropic properties and tunable bandgap [37]. Due to its biocompatibility, BP is explored for various biomedical applications, mainly drug delivery. MXene nano-sheets cover a recently discovered new family of layered transition metal carbides, nitrides, or carbonitrides, with their most explored member being $\text{Ti}_3\text{C}_2\text{T}_x$. MXenes exhibit outstanding electrical conductivity, excellent mechanical properties, tunable surface chemistry, and inherent antibacterial/antiviral properties, making them very promising to be applied in a biomedical context [38–41].

Regarding tribology, 2D materials have shown great potential to tailor friction and wear of various dry or lubricated systems when

employed as lubricant additives in base oils, as reinforcement phase in composites, or as solid lubricant coatings [42]. Graphene and its derivatives [43–46], TMDs [47–50], h-BN [51,52], BP [53,54], and MXenes [55–60] have been investigated in this context. Thereby, the layered nanomaterials may act as easy-to-shear films or nano-roller bearings, thus reducing friction [61]. Depending on the operational and environmental conditions, 2D materials form beneficial tribo-films through tribo-chemical reactions [62,63], which feature low shear resistance and can be transferred to the counter-body [64,65]. The newly formed tribolayer/tribolayer interfaces can substantially decrease friction and wear thus extending service life.

Excellent reviews focusing on various biomedical [24–27,66–70] or tribological [42,65,71–80] applications of 2D materials can be found in the literature. However, no article summarizing the tribological features of 2D materials in biomedical applications has been published to date. Therefore, the scope of this review lies in the progress and developments of 2D materials in biomedical tribology. In terms of biotribology, this article discusses the essential characteristics of 2D materials highlighting their importance and influence of being used in biotribological applications (Section 2) as well as the advancement and improvement in targeted applications, namely load-bearing implants (Section 3.1), dental implants (3.2), bone fracture fixation (3.3), invasive surgical devices (3.4), cardiovascular devices (3.5), contact lenses (3.6), and bio-sensing (3.7). Finally, the key findings, current challenges, and future research directions are presented in Section 4.

2. 2D materials in biotribology – how, where, and why?

2.1. Approaches for implementation

Depending on the application needs, the introduction of 2D materials can be divided into three main approaches: the deposition as protective coatings, the introduction as fillers in composites, or the usage as additives in fluids (Fig. 2a). All approaches are potentially applicable for biotribological applications and possess their advantages depending on the application requirements. Specifically, the use of protective coatings

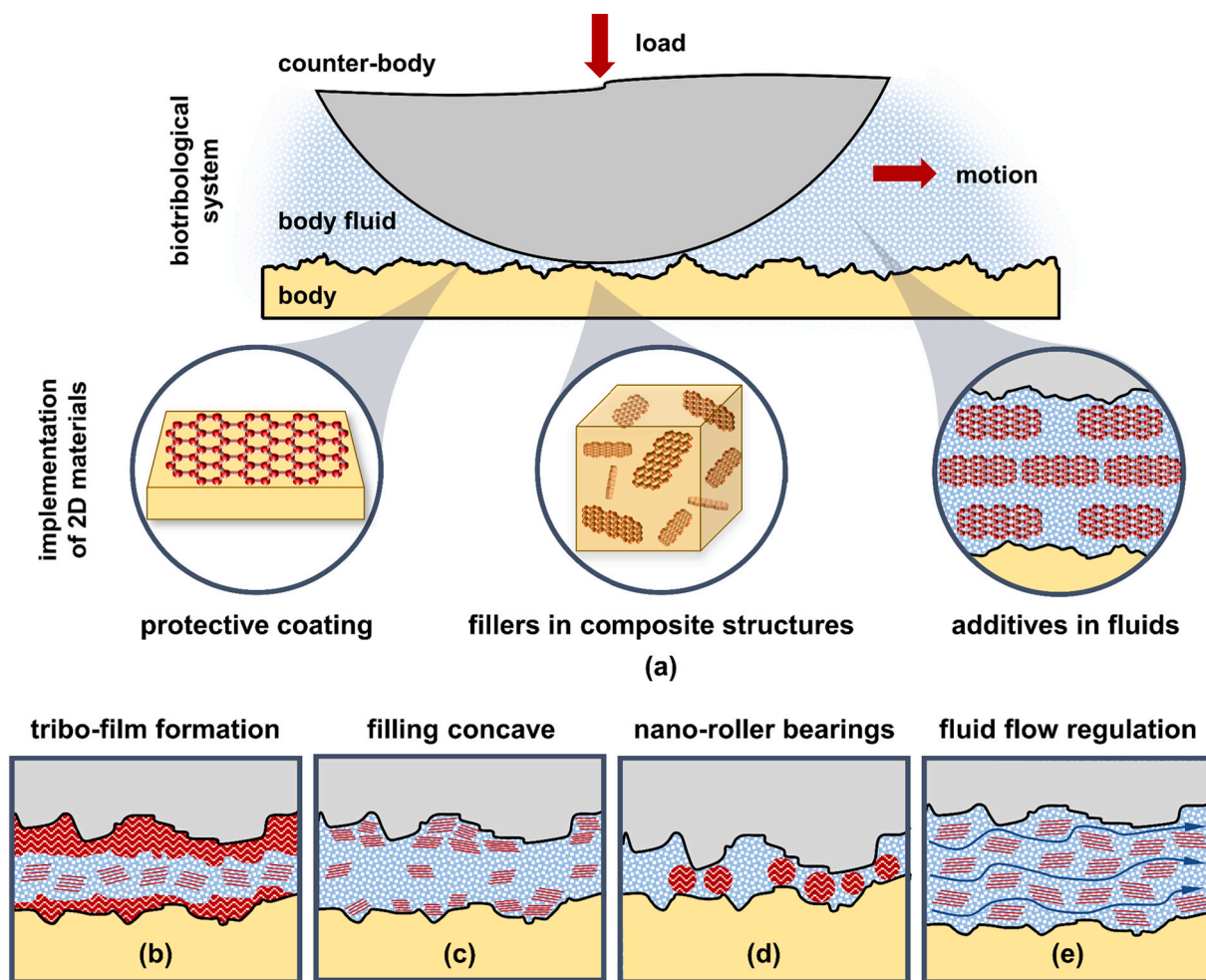


Fig. 2. (a) Implementation of 2D materials into biotribological systems. Lubrication mechanisms involving ultra-thin layered 2D materials: (b) Tribochemically-induced film formation, (c) lowering the surface roughness by filling concave valleys, (d) nano-rolling bearing effect, (e) fluid flow regulation. Redrawn and adapted from [75] with permission by CC BY 4.0 (MDPI, 2018).

is of high interest for the design of artificial joints, where their tribological characteristics allow to suppress the wear debris generation and release during operation without affecting the bulk mechanical characteristics of the majority of metal alloy-based prosthetic devices. In this case, the application approach is to deposit the coatings directly on the prosthetic device surfaces before the joint arthroplasty surgery. Meanwhile, dental or ocular systems may benefit from improvements in the composite materials' mechanical characteristics implemented for their use. Consequently, most of the ongoing research focuses on designing 2D fillers into polymeric or ceramic matrices. Here, the 2D materials are premixed in various concentrations into the powders or melted solutions of the materials. The use of 2D materials as additives to improve lubrication is potentially viable for systems, which highly depend on bio-fluids with limited renewal capacity, such as in ocular and load-bearing designs when they can be introduced in the form of injectable solutions or drops. In contrast to coatings and composites, the efforts also should be dedicated to ensuring good dispersibility and colloidal stability of these solutions.

2.2. Tribological potential

The discovery of 2D materials encouraged the exploration of their use for different applications. The major reason is the wide range of specific characteristics making 2D materials attractive for efficient implementation inside the human body. Specific to tribology-related applications, 2D materials have already demonstrated excellent

mechanical strength, chemical inertness, wear resistance, and easy shearing (low friction) [81–83]. Wear induced by tribocorrosion of metals covered with passive oxide films has represented a research topic of considerable interest in recent years, allowing the possibility of obtaining knowledge on a micro- and macro-scale. Recent advances in nanotechnology have also boosted the understanding and modeling of the fundamental tribological and corrosive mechanisms of 2D materials [84]. In general, according to the conventional tribo-corrosion theory [85], beyond oxide tribofilms having a nanometer thickness, the wear rate tends to increase more quickly with increasing contact pressure in the presence of physiological solutions than under dry friction.

The idea of using 2D materials for biotribology has been explored for a while. Earlier studies showed that the 2D forms of carbon (graphitic layers) could naturally occur in the joints during sliding [86], as a consequence of tribocatalytically-generated films [87], thus demonstrating the great potential for using 2D materials in the bio-environment. Upon addressing the challenges associated with the biocompatibility of 2D materials, the use of layered films would create new opportunities for their bio-applications inside the human body and/or in contact with biological systems. Below, we overview the main characteristics of 2D materials and discuss the approaches for their implementation in a biotribological context.

2.2.1. Mechanical strength

The unique layered structure of 2D films notably reduces the size of

defects and, typical for bulk brittle materials [88], significantly impacts the ability of 2D materials to sustain stresses and high deformation strains [81,89]. Consequently, 2D films were considered to protect the bulk functional components from external damage [42].

Focusing on composites, 2D materials as a filler component (reinforcement phase) distributed in polymeric matrix increase the overall composite's stiffness, improving the load transfer and load redistribution through the softer matrix simultaneously [90,91]. Consequently, reduced damage of macromolecules exposed to stresses and an improvement of the overall composite's strength can be expected.

2.2.2. Chemical inertness

One of the essential parameters relates to the ability of the 2D materials to sustain exposure to the corrosive environment in the human body [23,92,93]. Inherent resistance to degradation in aggressive environments also connects to the limited number of defects and dangling bonds in these materials. These aspects help to prevent their chemical bonding with surrounding compounds. Consequently, 2D materials were shown to provide excellent protection of the underlying surface from corrosion, that is a critical aspect when considering new material systems for biotribological applications [94,95]. Specifically, the human body is a complex environment that promotes corrosion and degradation of materials used in implants, which can further accelerate the failure of the materials under loading and shearing stresses. Upon sliding, the release of the corrosion components and wear debris to the body in turn may cause an osteolytic cascade reaction of the immune system. Because of severe corrosion damage, promoted by contact stresses, the pool of the material candidates considered for the implant components became very limited. An alternative approach of protecting the surfaces from corrosion, wear, and degradation by introducing lubricating solutions to the contacting interfaces, in the forms of coatings or advanced lubricants helps to improve the performance of the components and increase the viability of the materials.

2.2.3. Friction and wear characteristics

While enhancing the resulting mechanical properties of the systems using 2D materials is impressive, specifically for biotribological applications, their capability to improve a material's tribological properties (i.e. to decrease wear and friction) is of uttermost importance. The lubrication mechanisms involving 2D materials are driven by low shear resistance of ultra-thin layered structures (interlayer sliding) [64]. In this regard, 2D materials can be directly adsorbed or deposited on the contacting surfaces thus enabling tribochemically formed tribo-films (Fig. 2b) [62]. Moreover, 2D materials can fill surface cavities to reduce wear and self-replenish surfaces (Fig. 2c). Besides shearing films, 2D materials may act as nano-rolling bearings, thus potentially changing the friction mode from sliding to rolling friction while providing a certain load-bearing capacity (Fig. 2d) [61]. Lastly, 2D materials are able to regulate the fluid flow in full film lubrication regime (Fig. 2e) [75,96].

Therefore, 2D films were employed in a wide range of tribological systems across various scales [64] and environment conditions [50,63] as friction and wear-reducing materials inducing even zero-friction and zero-wear states (superlubricity) [74,83,87]. Similarly, polymer composites reinforced with 2D materials demonstrated a notably improved wear resistance, decreased coefficient of friction (COF), and improved durability in tribological experiments. At the same time, another research direction relates to the tribological performance of liquid lubricants containing 2D materials [43,97], as it addresses the challenges of 2D materials exposure to natural fluids even if the wear of the bulk composites or coatings proceeds.

2.3. Interaction with biological matter

Irrespective of their use as (composite) coatings or reinforcement phases in composites, it is inevitable that 2D materials will come into

contact with the human body and be recognized by the human immune system, which may induce a reaction of the body's first defense system [98–101]. The major challenge of 2D materials is the interaction with the immune system cells, potentially causing immunostimulation, such as the production of antibodies (or macrophages) attacking the invading material, or immunosuppression, which reduces the resistance of organisms against infections and cancerous cells [98,99,102]. Therefore, it is of utmost importance to elucidate their biological interaction and response towards the human body/tissue (biocompatibility, antibacterial as well as antiviral activity, and toxicity towards biological matter) when using 2D nanomaterials for biotribological applications [101–105].

A range of advantageous properties makes the use of 2D materials promising for biotribological applications. Due to their layered structure combined with low interlayer distances, they possess high specific surface areas (surface-to-volume ratios), which can be used for adsorption, to increase adhesion, to chemically functionalize these materials, to deliver drugs (antibiotics) or other therapeutic molecules [56,99,102,106–108]. Certain 2D materials, including graphene and its derivatives as well as MXenes, offer the possibility to transform electromagnetic radiation into heat, which makes them attractive to be used in photothermal therapies (cancer treatment) [99,109,110]. Their inherent biocompatibility coupled with antibacterial and antiviral properties, which holds true for numerous 2D materials including graphene, GO, rGO, MXenes, MoS₂, among others, represents the most striking property regarding their usage in biotribological applications [98,100]. In particular, graphene's toxicity highly depends on the superficial or chemical properties of the nanomaterial [111]. The main influencing parameters are dimension, carbon-oxygen atomic ratio, and a number of layers [112,113]. Comprehensive reviews about graphene and derivatives with their primary cytotoxicity mechanism being the cell damage induced by the formation of reactive oxidation species can be found in [114–116]. Concerning the toxicity of TMDs, the release of chalcogen is related to their toxicity [117–119], which can be reduced in the presence of selenium and vanadium [120]. Similar to graphene, BP generally shows good biocompatibility with low cellular cytotoxicity, whereas their functionalization allows for control of their biocompatibility [121].

While there are still some ongoing debates about the fundamental mechanisms of the antibacterial/antiviral properties of 2D materials, experimental and numerical works have determined some of the main influencing aspects. In this context, the direct interaction of 2D materials with bacteria can irreversibly damage cellular components, including nucleic acids, lipids, and proteins, thus causing cell death shortly after the interaction [100,103,122,123]. Oxidative stresses generated through the formation of reactive oxide species subsequently affect the bacteria's enzymatic cycles [103,124,125]. Due to the existing surface terminations on their exterior surface, 2D materials tend to be affine to the cell membrane. The sharp edges of 2D materials can provoke a rupture of the membrane ("nano-knife-effect"), leading to the death of the respective organism [126,127]. The 2D nature of these materials can also induce a wrapping effect. In this regard, the considerably large lateral dimensions and high flexibility of 2D sheets are beneficial to wrap up bacteria and viruses, thus separating them from the remaining environments. Due to the impermeable nature of 2D materials, this separation cuts the access to essential supplements, ultimately causing cell death [128–130]. Certain 2D materials have been reported to induce anti-inflammatory responses in the human body, which can further help fight bacteria and viruses. For more detailed information about the specific biological response of different 2D materials regarding different bacteria and viruses, the interested reader is referred to [98,99,103,105,123–125,127,129,130].

3. Usage of 2D materials in biotribological applications

In this section, we summarize and critically discuss the existing

findings regarding the application of 2D materials in load-bearing artificial (Section 3.1) and dental implants (Section 3.2), bone fracture fixation (Section 3.3), invasive surgical (Section 3.4), and cardiovascular devices (Section 3.5), contact lenses (Section 3.6) as well as bioMEMS/NEMS (Section 3.7). This includes a brief overview of the existing challenges and how 2D materials can help to overcome them.

3.1. Load-bearing artificial implants

Natural synovial joints represent a unique biotribological system subjected to sustain variable mechanical stresses. While its damage often relates to trauma, overloading, or diseases, synovial cartilage, which is essential for proper joint function, has a minimal healing ability [131]. As a result, cartilage damage leads to painful motion or even motion disability, which holds especially true for hip or knee joints. Although viscosupplementation by hyaluronic acid (HA)-based solutions may result in short-term relief, the only existing effective treatment is a total hip and total knee arthroplasty (THA and TKA). The involved contact pairs for joint replacements can be classified as hard-on-hard (metal/ceramic femoral head articulating with metal/ceramic acetabular cup) or hard-on-soft (metal/ceramic femoral head against polymer acetabular cup or metal/ceramic femoral component in articulation with polymer tibial insert) [132]. The main reasons for implant failure are frequently connected to aseptic loosening due to wear-particle-induced osteolysis [133,134]. Therefore, an improved tribological/wear performance of artificial joints is essential to avoid implant failure and to extend service life.

Owing to its outstanding mechanical properties, biocompatibility, and low friction, ultra-high molecular weight polyethylene (UHMWPE) is widely used for hard-on-soft load-bearing implants [135,136]. However, the material is known to be prone to wear. To address the wear issue, several potential solutions were considered. Recently, attention has been paid to developing highly cross-linked polyethylene (HXPE) used for acetabular cups/tibial inserts with the overall goal of lowering wear rate, thus minimizing the number of particles possibly interacting with surrounding tissues [137,138]. Furthermore, surface texturing of rubbing surfaces is considered to be one of the methods considerably improving the tribological performance of implant materials [139–141]. Another way is in coating the implants with diamond-like carbon (DLC) [142–146], titanium nitride [147], or silver [148]. Alternatively, the introduction of a nanodiamond-containing lubrication solution was proposed for improving the tribological properties of the implant materials [149,150]. While these approaches show their potential for improving the polymer surface resistance to wear, an alternative solution involves reinforcement of the bulk characteristics of the polymers. Moreover, the use of 2D materials opens new perspectives to tailor the tribological performance of load-bearing implants through their applications in bio-scaffold for bone tissue engineering along with bone therapy such as bone regeneration [151–153], cell proliferation and osteogenesis [154], wound repair [155], antibacterial ability, and osseointegration [156].

Liu et al. reported an increased hardness, hydrophilicity, crystallinity, and mechanical characteristics thus improving the overall wear resistance for graphene/UHMWPE composites (1.5 wt.-% of graphene) fabricated by octa-screw extrusion [157]. Martinez et al. investigated the effect of functionalized graphene and UHMWPE composite coatings on a UHMWPE substrate, verifying that they improve adhesion and ductility [158]. The mechanical and tribological properties depended on the graphene content and fabrication technique. The addition of 0.5 wt.-% graphene resulted in a wear reduction of 20%. Lahiri et al. manufactured graphene-reinforced UHMWPE by hot pressing and used electrostatic spraying to fabricate graphene-reinforced UHMWPE coatings [159,160]. At 1 wt.-% graphene-reinforced UHMWPE, the wear resistance increased a factor of 4.5 and the COF was reduced by 68% compared to pure UHMWPE. Additionally, low graphene concentrations showed better biocompatibility as a result of a reduced graphene

agglomeration tendency. Taromsari et al. sought to take advantage of hydroxyapatite's (HAp) outstanding biocompatibility by integrating it with graphene in UHMWPE, using solvent mixing and ultrasonication [161]. They verified that using 1 wt.-% graphene and 10 wt.-% HAp enhanced the elastic modulus and yield strength by 114% and 24%, respectively. Thereby, the tribological tests showed an 84% reduction in wear rate and a 55% friction reduction. The increased oxidative stress, and thus increased cytotoxicity, caused by graphene, were balanced by HAp, which boosts cell adhesion. The biocompatibility of graphene with HAp-reinforced UHMWPE was increased by establishing nucleation sites for apatite mineralization and preferential cell adhesion, as well as improved mechanical properties [162]. Graphene used as composite coatings or reinforcement phase in composites has been used for/titanium (Ti) alloy [163–165], magnesium (Mg) [166], Mg alloy [167], alumina [168], and polyetheretherketone (PEEK) [164,169]. The majority of these graphene-based investigations enhanced mechanical or tribological properties while taking biocompatibility into account.

In addition to pure graphene, attention was given to GO or rGO used for load-bearing implants due to their simple fabrication. Several studies combining GO with UHMWPE have been presented [170–175]. Tai et al. investigated the tribological behavior of GO/UHMWPE nanocomposites dependent on the GO content using a reciprocating ball-on-disc tribometer [174]. The addition of GO to UHMWPE increased the wear resistance by forming a transfer film with slightly increased COF. Chen et al. comprehensively studied the effect of GO and sodium chloride (NaCl) on the surface porosity of GO/NaCl/UHMWPE composites [171]. The dissolution of NaCl introduced new pores, which enabled the storage of fresh lubricious material, thus decreasing friction and wear. Lu et al. examined the effect of GO addition and chemical crosslinking on friction and wear of GO/UHMWPE nano-composites [175]. By pin-on-disc testing in simulated body fluid (SBF), they demonstrated that doping polyethylene with GO accompanied by crosslinking decreased both friction and wear due to enhanced bonding and linkage between GO and UHMWPE. In contrast, increased friction and wear were reported for samples immersed for six months in SBF due to swelling. Recently, Sharma et al. evaluated the tribological performance of GO/HDPE/UHMWPE nano-composites using pin-on-disc testing in phosphate buffered saline (PBS) [170]. The addition of GO reduced friction and induced a lower wear rate, which was mainly attributed to the self-lubricating ability of GO. Melk and Emami investigated the mechanical and thermal properties of vitamin-E doped UHMWPE reinforced by multiwalled carbon nanotubes (MWCNTs), GO, and nanodiamonds (ND) [176]. The results showed that all carbon nanoparticles positively impacted the thermal stability of UHMWPE, thus reducing the oxidation degradation temperature. Suner et al. investigated the wear rate and biocompatibility of GO/UHMWPE nanocomposites under lubrication with diluted bovine serum (BS) [177]. Lower contents of GO (0.5 wt.-%) did not affect the resulting wear rate while 2 wt.-% were needed to observe a 30% wear rate reduction. Similar results were observed by Pang et al. [178]. Marimuthu and Rajan demonstrated improved fatigue and wear properties for GO-reinforced high-density polyethylene (HDPE) nanocomposites [179]. Recently, Çolak et al. studied the effect of rGO on the performance of UHMWPE in reciprocating ball-on-disk tests using an alumina counter-body under water lubrication [180]. Friction was reduced by up to 37% for composites containing 0.7 wt.-% of RGo. An important drawback of most of the aforementioned studies is that the biotribological performance of the 2D materials was studied in absence of a biological lubricant. To overcome this aspect, Hussain et al. reinforced UHMWPE by graphene (using vitamin C as an antioxidant) to study their tribological response in reciprocating pin-on-disk tests under dry and lubricated (human serum) conditions [181]. They demonstrated a wear rate reduction by 99% for the novel UHMWPE/Vitamin C/graphene nanoplatelets (GNP) nanocomposite with 2 wt.-% of graphene.

In addition to UHMWPE, GO has been used to reinforce hydrogels for articular cartilage (avascular structure), which is mainly composed of collagen fibers. Hydrogels are well-known for retaining enormous

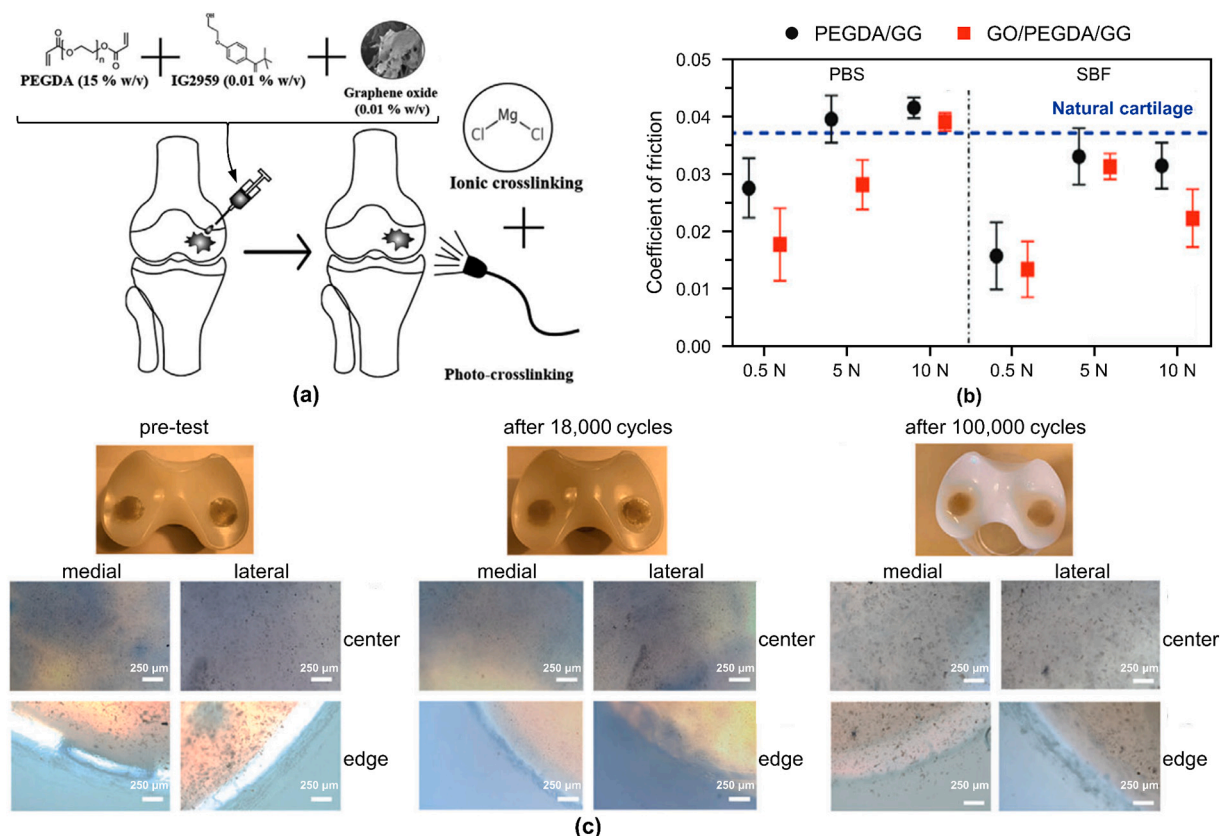


Fig. 3. (a) Schematic illustration of the targeted concept. (b) Average COF at varying normal forces under PBS and SBF lubrication for PEGDA/GG and GO/PEGDA/GG composites. (c) Macroscopic and brightfield images of medial and lateral condyles prior to testing as well as after 18,000, and 100,000 cycles, respectively. Redrawn from [183] with permission (Wiley, 2021).

amounts of water and preserving a specific shape, but they cannot provide sufficient structural or mechanical support on their own. Therefore, GO with polyethylene glycol (PEG) wrapped in Chitosan (CTS)/sodium glycerophosphate (GP) hydrogels helped to improve lubrication efficiently while reducing wear with no cytotoxicity for low GO contents [182]. The hydrogels for cartilage application were characterized using the μ -CT technique to study the influence of GO on wear of the blends of gellan gum (GG) and polyethylene glycol diacrylate (PEGDA) doped with/without GO reinforcement [183,184]. GO was found to reduce the roughness modification, resulting in reduced friction and improved wear resistance (Fig. 3).

Wang et al. investigated the electrochemical and biotribological characteristics of laser-textured, GO-coated Ti-Al-V plates with variable groove widths demonstrating the superior corrosion and wear resistance of the coated alloy samples [185]. GO has also been studied in conjunction with HAp for load-bearing implants as a composite coating on various metal substrates. The presence of GO enhanced the mechanical properties, corrosion resistance, and coating adhesion [186,187].

Though the major focus was directed to graphene and graphene-based systems, other 2D materials, such as TMDs, h-BN, or BP have also been investigated for load-bearing implant applications. Salem et al. discovered that the inclusion of MoS₂ increased the wear resistance of HDPE composites [188]. BP affected the mechanical properties, corrosion behavior, and biocompatibility (cell proliferation, osteogenic differentiation, antibacterial properties) of load-bearing implants [154,189,190]. Li et al. discovered that BP/carbon fiber (CFR)/PEEK coatings showed a better wear resistance and lower friction, possibly due to the lower propensity of BP oxidation compared to the BP/CFR/PEEK coatings [191]. Sun et al. showed that PEEK/PTFE composites with BP reinforcement featured superior wettability and antibacterial

properties as well as lower friction (reduction by 73%) and wear (reduction by 95%) compared to the pure matrix materials due to the good lubricous properties of the BP transfer film [192]. Thereby, 0.5 wt. % of BP showed the best performance.

To summarize, 2D materials are promising for improving the mechanical and biotribological properties that affect the longevity and functionality of load-bearing implants. However, there are still some challenges in characterizing the *in-vivo* biotribological performance as well as studying the coupled effect on the articulating and the fixation interfaces. The experimental studies presented, therefore, demand further clinical validation.

3.2. Dental implants

Human teeth are usually exposed to substantial wear during biting and chewing [193,194]. An increasing number of biotribological studies focus on dentistry with two main research directions, namely tooth restoration devices and dental implants [5]. The materials employed must be wear-resistant without deteriorating the counterparts (natural or restored teeth) during chewing. Moreover, they must resist fretting corrosion and provide adequate fixation stability to ensure prolonged lifetimes [5,195–198]. Besides that, aesthetic aspects have continuously become more important. Metals and their alloys are mainly used for orthodontic appliances and dental implants but are less frequently utilized for restoration purposes due to their undesirable color [5]. In contrast, polymeric alternatives tend to have high wear rates and poor gloss retention [199]. Dental ceramics based on yttria-zirconia (3Y-ZrO₂), Leucite (KAlSi₂O₆), or Fluorapatite (Ca₅(PO₄)₃F) are generally used as restoration materials due to their excellent wear resistance and overall appearance mimicking natural teeth.

To enhance the wear- and corrosion-resistance of tooth restoration or

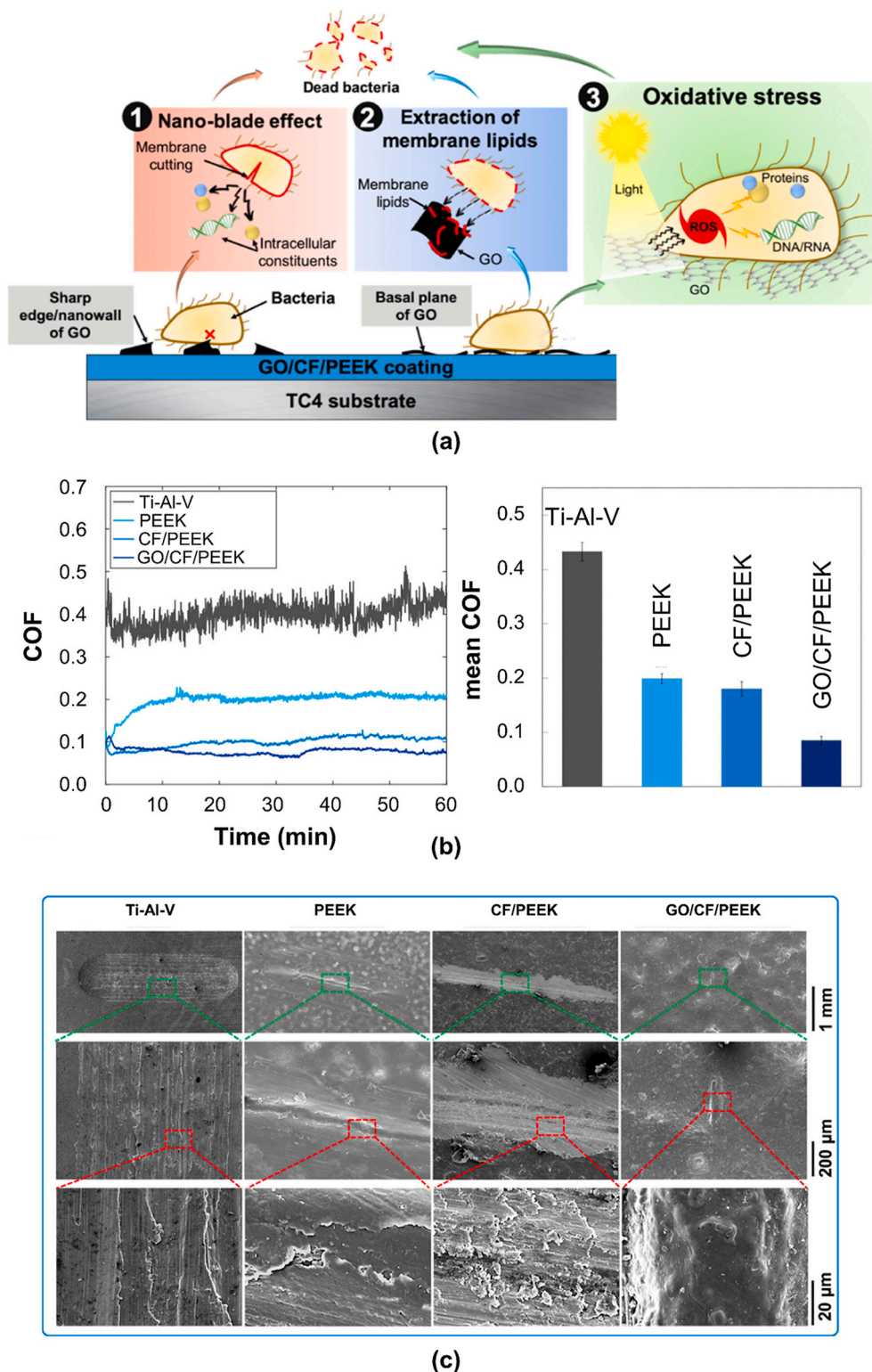


Fig. 4. (a) Schematic illustration of antibacterial mechanisms of the GO/CF/PEEK composite coatings. (b) COF and (c) worn surface morphologies of the Ti-Al-V, PEEK, CF/PEEK, and GO/CF/PEEK surfaces, respectively. Redrawn from [164] with permission (Elsevier, 2021).

dental implant materials, composites appear to be an interesting alternative due to the possibility to mimic the characteristics of natural teeth and restore cavities as well as replace carious tooth structures, thus improving clinical efficiency [200,201]. In this context, resin-based organic matrices with inorganic filler particles, e.g. zirconia, borosilicate glass, quartz, or alumina [202–206], as well as hybrid ceramic-polymer composites [207–210] have been recently studied for dental

restoration [5].

Regarding dental implants, Ti, zirconium (Zr), tantalum (Ta), and their alloys are used for bone fixation due to their good cytocompatibility [211–215]. Although their elastic moduli are suitable for such applications, their biotribological performance is rather poor. Additionally, toxic elements such as aluminum (Al) or vanadium (V) can be released due to micro-movements and tribo-corrosion, leading to

inflammatory reactions in biological environments [5]. Therefore, efforts have been recently dedicated to modifying and coating implant surfaces [216–220] or integrating porous structures [221–224] to enhance their mechanical properties, wear-resistance, bone ingrowth, and osseointegration. In this regard, 2D materials can potentially help to address these challenges intensifying the efforts for exploring graphene- and GO-reinforced composite ceramics or composite coatings on metal alloys [225] for dental restoration [226,227] or implant materials [164,228–230].

Zhang et al. fabricated GO-reinforced 3Y-ZrO₂ ceramics with different concentrations using hot-press sintering and good mechanical properties due to a homogeneous GO distribution [226]. Moderate GO contents (0.1 or 0.15 wt.-%) improved the flexural strength and fracture toughness by 200% and 41%, respectively, which was related to crack deflection and bridging as well as GO put-out. Lower (0.05 wt.-%) or higher (0.20 wt.-%) contents of GO downgraded the overall structural and mechanical properties. Linear-reciprocating ball-on-disk biotribological studies demonstrated that moderate GO contents induced the best performance with a wear rate reduction by 33% (0.10 wt.-%) and 63% (0.15 wt.-%), respectively. Lower and higher GO concentrations did not show any significant effect or even induced a downgraded performance with an increased gravimetric wear rate. The beneficial effects were connected with changes in the predominant wear modes, which changed from severe micro-ploughing and plastic deformation for the pure ceramic to very mild wear for the GO-reinforced composites (wear-resistant tribo-film). Subsequently, Zhang et al. verified lower contact angles on GO-coated surfaces compared to pure 3Y-ZrO₂ indicating enhanced wetting properties, which was attributed to the existing hydroxyl groups [227]. No signs of cytotoxicity were observed, and the GO-reinforced ceramics promoted proliferation, growth, and adhesion of fully elongated cells. The authors demonstrated that the composite ceramic can be fabricated by stereolithography from GO/3Y-ZrO₂ suspensions, which is an important step towards the formation of complex and custom-designed dental implants [231].

Sun et al. prepared bright white color fluorinated graphene (FG) for fabricating glass ionomer cement (GIC) composites reinforced with various FG concentrations [232]. The analysis demonstrated that the Vickers microhardness and compressive stress of such composites reduced, and the COF in zirconia ball-on-disk tribometer tests under artificial saliva lubrication decreased. The composites also showed the antibacterial effect against *S. aureus* and *S. mutants* correlated with the FG content.

Regarding metallic implants, Kalisz et al. compared the mechanical and corrosion properties of graphene deposited on a Ti alloy with a 210 nm thick, magnetron-sputtered niobium pentoxide (Nb₂O₅) layer as a reference [228]. While Nb₂O₅ could withstand a steel wool scratch-test without substantial scratches, the graphene coating was easily removed from the Ti-Al-V surface. However, graphene featured much better inferior corrosion resistance than Nb₂O₅. In a 0.5 M/l NaCl, 2 g/l KF, pH 2 electrolyte, which is a more aggressive environment than SBF or artificial saliva, the corrosion current density was 24 times lower (0.01 $\mu\text{A}/\text{cm}^2$) for the graphene-coated Ti-Al-V surface compared to Nb₂O₅ (0.24 $\mu\text{A}/\text{cm}^2$). The authors concluded that hybrid coatings containing graphene or its derivatives can improve the mechanical and corrosion resistance properties of metallic implants [228].

Mahmoodi et al. investigated the *in-vitro* corrosion, and biotribological behavior of 2 μm thick GO-reinforced HAp composite coatings electrophoretically deposited on Ta substrate [229]. The surface roughness increased from about 35 nm (uncoated) to 120 nm (composite coating). The stiffness was 8-fold higher, the nano-indentation hardness (H_{IT}) increased by a factor of 18, while the elastic modulus (E_{IT}) was 14 times higher. Consequently, the fracture toughness (H_{IT}/E_{IT}) and the resistance to plastic deformation (H_{IT}^3/E_{IT}^2) were enhanced. The COF of the coated samples was 30% lower (0.30) than that of uncoated reference (0.43). Moreover, the hybrid coatings featured a substantially lower corrosion current density (5.0 versus 18.6

$\mu\text{A}/\text{cm}^2$) in Hanks' solution and 3.5 times higher corrosion resistance. This indicated that GO-reinforced HAp composite coatings bear tremendous potential for dental implants' surface protection. Even more beneficial, they verified a high antibacterial activity against *S. aureus* and *E. coli* and hindered the adhesion of bacteria for the composite coatings. The increased hydrophilicity and roughness of the coated surfaces further improved MG-63 cell proliferation, indicating good biocompatibility.

Salehi et al. employed plasma-spraying to fabricate GO/fluorapatite (FA)/zinc oxide (ZnO) nanocomposite coatings on Ti-Al-V alloys [230]. After optimizing the deposition parameters to obtain homogeneous, uniform coatings with decreased crystallites, it was demonstrated that the addition of GO increased the micro-hardness from 450 to 760 HV while roughness and contact angle decreased from 6.4 μm to 4.1 μm and from 81° to 58°, respectively. Moreover, the GO/FA/ZnO coatings induced a bone-like apatite formation after 14 days of immersion in SBF solution and promoted MG63 cell viability and spreading. Zirconia ball-on-disk tribo-testing under SBF lubrication showed a reduced COF from 0.6 to 0.4 and a decreased wear rate (by 60%,) while the wear mechanisms remained unchanged.

Qin et al. studied GO/CF/PEEK composite coatings with 0.02 wt.-% GO and 25 wt.-% CF powder sprayed on Ti-Al-V alloys [164]. In antibacterial tests, the GO/CF/PEEK composites exhibited good suppression towards *E. coli* and *S. mutans* as well as an excellent activity against *S. aureus* due to a nano-blade effect, the extraction of membrane lipids and oxidative stresses induced by the randomly distributed and oriented GO nanosheets (Fig. 4a). Moreover, a reduced COF (Fig. 4b), as well as an improved wear-resistance of GO/CF/PEEK, compared to Ti-Al-V, PEEK, and CF/PEEK without GO (Fig. 4c) were demonstrated in reciprocating sliding against Si₃N₄ under SBF lubrication.

To summarize, the clinical outcomes of dental restoration and implants can be considerably improved due to the enhanced biotribological performance boosted by graphene or GO used as material reinforcement or composite coatings. Although being highly promising, there are still many challenges and drawbacks to be overcome. Most studies focused on studying the cytocompatibility, anti-bacterial properties, and wear behavior *in-vitro*. The long-term wear behavior and mechanisms are yet to be elucidated. The lack of research on *in-vivo* cell attachment, bone ingrowth, and osseointegration and the missing knowledge about the involved wear mechanisms when exposed to the oral microbial environments represent the main obstacles in the employment of 2D materials for dental materials, which urgently ask to be addressed.

3.3. Bone fracture fixation

Many orthopedic devices, including plates, nails, wires, and screws, are used to re-set and fix the fracture in place when a bone is broken. Stainless steel, Ti, cobalt-chromium (CoCr), nickel-titanium (Ni-Ti), or Mg alloys are employed for permanent or temporary/degradable prostheses [233–235]. Thereby, fatigue processes of bone plates at screw holes due to cyclic-loading [236], as well as crack acceleration/propagation, abrasion, and fretting corrosion induced by micro-movements are major causes for destabilization and premature failure [5,237]. Since these elements are in direct contact with cortical bones, their biotribological performance in terms of wear particle and ion release, which can result in inflammation and carcinogenic effects [238,239], is essential. Therefore, efforts have been recently dedicated towards optimizing the fixation mechanisms designs [238,240] via tailoring microstructures by processing routes [241], ion nitriding [242], or the application of coatings [243–247]. With respect to 2D materials, h-BN-reinforced composite coatings on Ti alloy [248,249] as well as GO coatings [250] and graphene as reinforcement phases [167,251] for Mg alloys have been studied.

Tozar and Karahan electrophoretically deposited h-BN- and collagen- (COL) reinforced HA/CTS composite coatings on Ti-Al-V

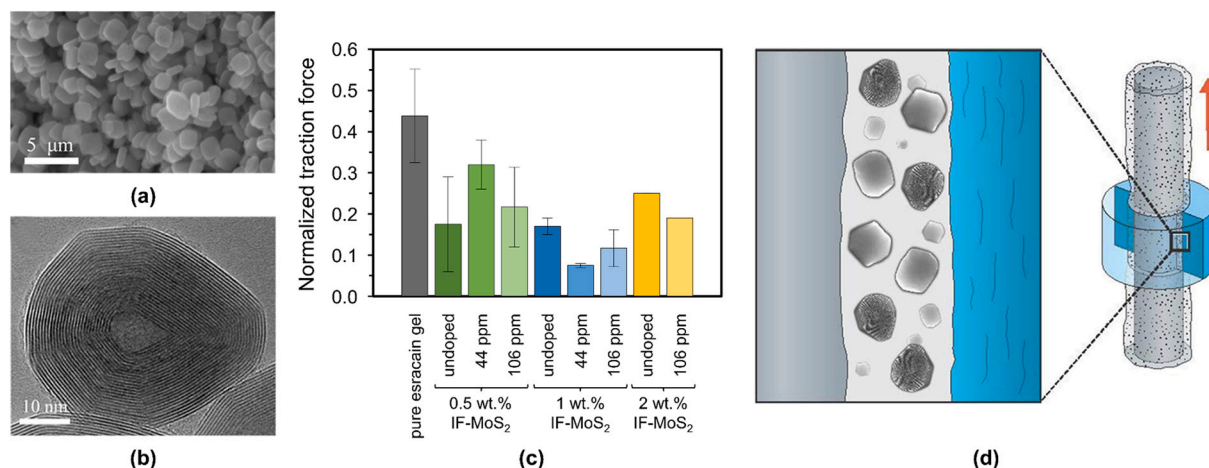


Fig. 5. (a) Scanning electron and (b) transmission electron microscopy images of IF-MoS₂ nano-particles, reprinted from [258] with permission (2014, Springer). (c) Traction forces to move the lead through the urethra model ring using differently formulated gels normalized to dry conditions [259]. (d) Schematic illustration of the lubrication gel mixture between the lead and urethra model ring, reprinted from [258] with permission (2014, Springer).

[248]. The combination of both reinforcement phases (h-BN/COL/HA/CTS) provided superior corrosion protection in SBF compared to single COL- or h-BN-reinforced and pure HA/CTS composite coatings. While the addition of just COL decreased the elastic modulus and increased friction compared to HA/CTS, the reinforcement with h-BN enhanced the mechanical properties (higher elastic modulus and hardness) and decreased the surface roughness as well as friction. Subsequently, Tozar and Karahan demonstrated that the corrosion protection performance, the mechanical properties, and the frictional behavior improved with increasing the h-BN concentration up to 5 g/L [249]. For higher concentrations, downgraded effects were verified. Moreover, no cytotoxic effects were observed after 12 weeks of sample immersion into the SBF. Shahin et al. reported that the addition of graphene to zirconium alloyed Mg led to a microstructure refinement, thus increasing the hardness and stiffness of materials [167]. The addition of graphene was superior to solely alloying with Zr, thus reducing friction by as much as 71% and improving the wear resistance by up to 94% compared to pure Mg.

To summarize, the usage of 2D materials as reinforcement phase in composite coating is promising to address the biomechanical and biotribological challenges that affect the lifetime and functionality of fracture fixation devices. However, research in this area is still in its infancy. Therefore, the mechanisms leading to improvement, the effect of different 2D materials, as well as the influence of wear on the performance and osseointegration of fracture fixation components, are yet to be investigated.

3.4. Invasive surgical devices

Minimally invasive surgical devices such as needles, graspers, endoscopes, or catheters are used in various medical operations. Thereby, friction greatly influences device-tissue interactions, device positioning, the accuracy of force control, and tissue damage, which may lead to long-lasting trauma. Consequently, these aspects drive the development of innovative minimally invasive needles or grasping forceps [5], for which current trends are mostly directed towards the optimization of motion [252,253] and geometry [254–257] rather than materials utilized. The insertion and operation of different medical devices, such as endoscopes, catheters, laparoscopes, and colonoscopies, represent another relevant biotribological application. Thereby, the cylindrical polymeric or metallic instruments need to pass through the mouth, esophagus, or anus, among others, to reach the target position. During this procedure, the much softer human body tissue coming in contact with these instruments can be easily damaged due to compression, stretching, and friction [5]. Therefore, water-soluble and sterile gels are

typically employed for lubrication and local anesthesia [258].

Goldbart et al. added fullerene-like MoS₂ (IF-MoS₂) and rhenium-doped MoS₂ (Re:IF-MoS₂) nano-particles (Fig. 5a, b) in different concentrations to a commercial 2% lidocaine-hydrochloride (Esracain) gel [259]. The resulting gel mixture was applied on a metallic lead and tested against a soft polydimethylsiloxane (PDMS) ring to simulate metal-urethra interaction under linear motion. It was shown that friction could be substantially reduced for Re:IF-MoS₂ (Fig. 5c). Since the nanoparticles did not show any signs of peeling-off or transferred layers, the friction reduction was attributed to fullerenes' ability to roll, thus separating contacting surfaces, which changed the mechanism from sliding to rolling friction (Fig. 5d). While the traction force decreased with increasing concentration from 0.5 to 1 wt.%, a higher concentration (2 wt.%) led to a degradation of the gel and particle agglomeration, resulting in higher viscosity and, thus, in increased friction (Fig. 5c).

Adini et al. deposited a cobalt coating with impregnated fullerene-like WS₂ nano-particles on Ni-Ti endodontic files used for root canal treatment [260]. Torque measurements of endodontic files clamped between two stainless steel plates at defined loads indicated a substantial improvement in the fatigue resistance and time to breakage of the coated files due to reduced friction between the file and the surrounding tissue.

To summarize, the application of 2D materials as additives in lubricating gels or as coatings appears to be promising to reduce friction and/or wear thus enhancing the overall performance and reliability of invasive surgical devices such as endoscopy or root canal treatment. However, the entire topic is greatly underexplored, with many directions for future systematic investigations. So far, only the application of TMDs (MoS₂ and WS₂) has been investigated, which implies that the usage of other 2D materials is highly prospective. Moreover, more research has to be dedicated towards *in-vitro* model setups and materials that adequately reflect the complex *in-vivo* conditions. Particular emphasis should also be paid to the biocompatibility of the used materials and the dynamic interaction of the device surfaces with human skin, tissue, and blood cells.

3.5. Cardiovascular devices

Various devices are employed for the treatment of cardiovascular diseases. In these procedures, biotribological issues can induce serious complications leading to hemolysis, thrombus formation, or device failure [5]. For instance, stents and catheters used inevitably interact with vessel walls and potentially damage the endothelium and tissue by frictional motion [261,262]. Moreover, fretting wear can occur between

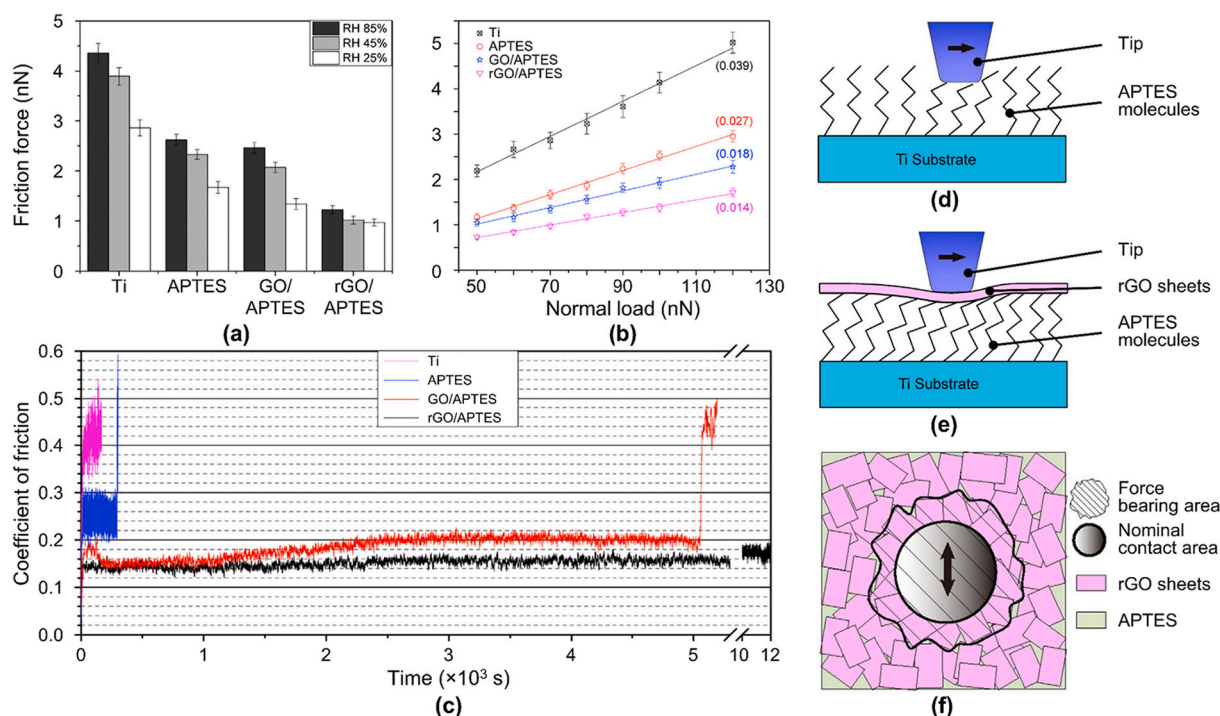


Fig. 6. Friction force of Ti, APTES, GO/APTES, and rGO/APTES at (a) different relative humidities and (b) normal loads. (c) COF versus time for Ti, APTES, GO/APTES, and rGO/APTES. Schematic illustration of the interaction between the AFM tip and the (d) APTES or (e) rGO/APTES films as well as (f) force bearing area enlargement due to the rGO/APTES coating. Reprinted from [291,292] with permission (Elsevier, 2018).

overlapping stents [263] and blood friction can be related to the migration of the stent from the intended position [264] and corrosion of the stent materials [265]. In this context, 2D materials such as GO coatings on biodegradable vascular stents made of Mg alloys may enhance corrosion resistance, blood compatibility, and antibacterial activity [266,267]. Furthermore, friction and wear of the moving components in mechanical heart valves or ventricular assist devices such as pulsatile or rotary pumps represent a major concern as they lead to blood clots and tissue damage due to excessive shear stresses and wear particle wash-out and contamination [268–270].

The cardiovascular devices include a number of components such as rolling, journal, or thrust bearings [271–273], which either are in direct contact with blood, lubricated by a saline-blood mixture, or have an indirect influence through friction-induced heating even when they are sealed [5]. 2D materials can be applied as coatings or reinforcement phases for mechanical components, thus effectively reducing friction, wear, and preventing local heating [42]. Regarding biomedical applications, Arokia Raj et al. transferred monolayer graphene from a CVD-coated copper (Cu) foil to bi-leaflet mechanical mitral valve surfaces using polymethylmethacrylate (PMMA) as a sacrificial layer [274]. Accelerated wear tests under saline lubrication were performed, and the presence of a protecting graphene layer on the valves was verified after 40 million cycles (corresponds to an operation of 1 year). The authors rated the risk of toxicity as negligible due to the low amount of graphene and the reduced probability of accumulation in non-desired body parts. Moreover, they hypothesized that conjugating the graphene coating with glucose oxidase may be useful as an auto-thrombolytic method in mechanical heart valve therapy and reduce the need for anticoagulation.

Even with the existing success of using 2D materials in highly-stressed machine elements such as bearings, pumps, or sealings, which can be also found in cardiovascular devices [42], the application of 2D materials in these devices is greatly under-explored. Reasons for this can certainly be traced back to the uncertain operating conditions under *in-vivo* conditions as well as the complex and individually varying mechanisms of blood lubrication or, for instance, reactions of the component

materials with anticoagulants.

3.6. Contact lenses

Contact lenses are ocular prosthetic devices employed for vision correction, therapeutics, and cosmetics [275]. Recent research has been directed towards the optimization of chemical and physical material properties, the development of tailored manufacturing processes, and the avoidance of microbial contamination and other ocular complications to minimize adverse effects associated with contact lens wearing, maintain a regular corneal metabolism, and preserve tear film stability [275]. Contact lenses can be grouped into soft, rigid, and hybrid compositions, whereby polymethylmethacrylate (PMMA), (fluoro-) silicone acrylate, and water-containing polymer hydrogels are commonly used [275]. Comfort, visual performance, and durability are determined by their mechanical properties, wettability, and soaking characteristics [275]. Friction between the eyelid and contact lens is another important aspect. Although too low or too high friction can be irritating, no standardized reference values are available yet [276]. The utilization of 2D materials such as graphene is currently more concentrated on therapeutic applications, thus extending the therapeutic function of the contact lens by sensing platforms [277,278], e.g. for full-corneal electroretinogram recording [279] or electromagnetic interference shielding [280] and dehydration protection [281].

Regarding lubrication purposes, Huang et al. [282] directly loaded silicon contact lenses with HA and rGO. Moderate amounts of rGO featured permissible swelling and transmittance properties as well as a low burst with release times up to 96 h compared to 24 h for conventionally soaked lenses. HA/rGO-loaded lenses showed a high HA-tear fluid concentration and an improved tear fluid volume. Therefore, the usage of HA and rGO bears the potential to lubricate the contact lens without protein adherence and the necessity for eye drop solutions when treating corneal diseases like keratoconus or dry eye syndrome. Also, considering previous efforts in various lubrication systems [65,75,76], we hypothesize that 2D materials, specifically graphene with its

Table 1

Summary of 2D materials as well as their main effects when employed for load-bearing implants.

2D material and form of application	Substrate or matrix	Biotribological evaluation	Main findings	Ref.
GO reinforced composite polymer	UHMWPE	Dry rotational pin-on-disk testing	Enhanced wear resistance, increased hardness, hydrophilicity, crystallinity, and yield strength	[157]
GO/UHMWPE composite coating	UHMWPE	DI water-lubricated rotational alumina ball-on-disk testing	Wear and friction reduction, improved adhesion and ductility	[158]
GNP/UHMWPE composite coating	UHMWPE	Dry nano-scratch testing	Wear and friction reduction, improved biocompatibility, fracture toughness, and tensile strength	[159,160]
GNP/HAP composite polymer	UHMWPE	Dry rotational steel pin-on-disk testing	Wear and friction reduction, enhanced elastic modulus and yield strength, increased oxidative stress	[161]
GO/CS composite coating	Ti-6Al-4V	Dry rotational steel ball-on-disk testing	Enhanced wear resistance and cytocompatibility	[163]
Al ₂ O ₃ /GO/HAP	Ti-6Al-4V	Dry rotational pin-on-stainless steel disk testing	Wear and friction reduction, decreased microhardness	[165]
GO reinforced composite polymer	UHMWPE	Dry reciprocating zirconia ball-on-disk testing	Wear reduction, slight friction increase, increased hardness	[174]
GO reinforced composite polymer	UHMWPE	BCS lubricated reciprocating Si ₃ N ₄ ball-on-disk testing	Wear and friction reduction	[172]
GO reinforced composite polymer	UHMWPE	SBF lubricated rotational pin-on-CoCrMo disk testing	Wear and friction reduction	[175]
GO reinforced composite polymer	HDPE/ UHMWPE	PBS lubricated rotational stainless steel pin-on-disk testing	Wear and friction reduction	[170]
GO/NaCl/UHMWPE composite polymer	UHMWPE	Water lubricated reciprocating steel ball-on-plate testing	Wear and friction reduction	[171]
GO reinforced composite polymer	UHMWPE	BS lubricated reciprocating pin-on-CoCr plate testing	Enhanced wear resistance	[177]
GO reinforced composite polymer	UHMWPE	Dry, (sea) water lubricated rotational block-on steel ring testing	Enhanced wear resistance	[178]
GO reinforced composite polymer	HDPE	Dry rotational pin-on-abrasive wheel testing	Enhanced wear resistance and fatigue properties	[179]
rGO reinforced composite polymer	UHMWPE	Water lubricated reciprocating alumina ball-on-disk testing	Friction reduction, slight wear reduction, increased hardness	[180]
GNP reinforced composite polymer	UHMWPE	Dry, human serum lubricated reciprocating pin-on-Ti6Al4V disk testing	Wear reduction	[181]
GO/PEG reinforced composite polymer	CS/GP hydrogel	Water lubricated reciprocating ball-on-CoCrMo disk testing	Enhanced lubrication properties	[182]
GO reinforced composite polymer	PEGDA/GG hydrogel	PBS, FBS lubricated rotational pin-on-disk and CoCr swinging femoral-on-tibia insert testing	Wear and friction reduction	[183,184]
GO coating	Ti-Al-V	Dry and BSF lubricated reciprocating Si ₃ N ₄ ball-on-plate testing	Superior wear and corrosion resistance, friction reduction	[185]
MoS ₂ reinforced composite polymer	HDPE	Dry reciprocating stainless steel ball-on-plate testing	Wear reduction, slight friction reduction	[188]
BP reinforced composite polymer	PEEK/PTFE	Dry reciprocating GCr15 steel ball-on-plate testing	Wear and friction reduction	[192]

derivatives, can be considered as an additive to eye drops solutions for ocular applications [283].

3.7. Bio-MEMS/NEMS

Electrokinetic- or microfluidic-based biomedical micro- or nano-electromechanical systems (bio-MEMS/NEMS) [284] are increasingly employed as miniaturized biosensors [285,286], microarrays for genomic, proteomic, and point-of-care diagnostics [287,288], medical implants for drug delivery, microtools for surgery or cell and tissue engineering [287]. Besides silicon, glass, or polymers, Ti, Al, Cu, or gold (Au) are used due to their electrical and mechanical properties as well as good biocompatibility [289]. Due to their unique electrical, physical, and optical properties, 2D materials are used to modify the involved surfaces for biosensing purposes. The high specific surface area to volume ratio of 2D materials enables a strong response to the surface activities suitable for biosensing.

The stable operation of bio-MEMS/NEMS devices also depends on the surface properties and the control of adhesion, friction, and wear level of surfaces coming into contact during their operation [290]. For instance, Ti alloys possess disadvantages regarding their biotribological behavior, thus focusing on their surface modifications [289].

However, only a few biotribological studies including 2D materials for biosensors have been presented to date. Li et al. chemisorbed rGO (thickness of 1.3 nm) on β -type Ti-Nb-Ta-Zr (TNTZ) alloys using 3-aminopropyltriethoxysilane (APTES) [291,292]. Thereby, the silanol moiety of APTES chemically bonded to TNTZ, while the amino group was

covalently anchored to the oxygen groups of GO. A thermal reduction process generated self-assembled rGO/APTES layers with a thickness of roughly 3.3 nm. In AFM experiments using a square pyramid Si₃N₄ tip, the rGO/APTES film reduced adhesion and friction and showed less sensitivity to humidity and load (Fig. 6a, b). A superior wear-protection with an increased time until failure was observed in linear-reciprocating ball-on-disk experiments using a sodium chloride aqueous solution, (Fig. 6c). This observation was traced back to the self-lubricating properties of rGO and a “hard cover on molecular springs” mechanisms (Fig. 6d, e). Thereby, the chemically bonded rGO sheets distributed the load over a larger area, thus increasing the force bearing area compared to the nominal contact size of pure APTES or Ti (Fig. 6f). Wang et. al proposed a GO coating (coupling agent APTES) and a protein dopamine (DA) adhesion layer on Ti alloys [293]. The anticorrosion behavior under dry conditions and SBF lubrication was enhanced compared to the uncoated Ti-Al-V. Improved wear properties and a friction reduction were demonstrated for the GO-coated samples. Thereby, the tribological results of the GO coated samples were superior under dry compared to SBF lubrication conditions. Moreover, the DA adhesion layer enhanced the properties compared to the pure GO/APTES coating.

Laun et al. experimentally studied the wettability of MoS₂ surface with water and used molecular dynamic simulations to predict the slip-length of water on MoS₂ nano-sheets [294]. It was shown that low friction is related to the hydrophobic nature of the surface (large slip length ~ 1.8 nm) suggesting that MoS₂ suppresses permanent biomolecule adsorption on the surface. They anticipated that the developed

Table 2

Summary of biomedical applications beyond load-bearing implants and the applied 2D materials as well as their main effects.

Biomedical application	2D material and form of application	Substrate or matrix	Tribological evaluation	Main findings	Ref.
Dental implants	GO reinforced composite ceramic	3Y-ZrO ₂	Artificial saliva lubricated reciprocating steel ball-on-disk testing	Improved flexural strength and fracture toughness, wear reduction and shift into the mild-wear regime, transfer-film formation, good cytocompatibility	[226,227]
	Fluorinated GO reinforced composite ceramic	Glass ionomer cement	Artificial saliva lubricated reciprocating zirconia ball-on-disk testing	Enhanced hardness and compressive stresses, friction reduction	[232]
	Graphene monolayer coating	Ti-Al-V alloy	Dry steel wool scratch testing	Low adhesion, but enhanced corrosion resistance	[228]
	GO/carbon fiber/PEEK composite coating	Ti-Al-V alloy	SBF lubricated rotational zirconia ball-on-disk testing	Enhanced friction and wear behavior, good cytocompatibility	[164]
	GO/FA-ZnO composite coating	Ti-Al-V alloy	SBF lubricated reciprocating Si ₃ N ₄ ball-on-disk testing	Friction and wear reduction, good antibacterial activity, and cytocompatibility	[230]
Bone fracture fixation	GO/hydroxyapatite composite coating	Ta alloy	Dry nano-scratch testing	Enhanced wear and corrosion resistance, good antibacterial activity, and cytocompatibility	[229]
	h-BN reinforced COL/HA/CTS composite coating	Ti-Al-V	Dry nano-scratch testing	Enhanced corrosion resistance and friction	[248,249]
	GNP reinforcement	Mg-Zr, Mg-Zr-Zn	Dry nano-scratch testing	Enhanced friction, wear and corrosion resistance	[167,251]
Endoscopy	Fullerene-like structured undoped and rhenium-doped MoS ₂ nanoparticles	Esracain gel	Linear urethra model medial lead-in-PDMS ring testing	Friction reduction due to change from sliding to rolling friction	[258,259]
Root canal treatment	Fullerene-like WS ₂ nanoparticle impregnated cobalt coating	Ni-Ti-alloy	Endodontic file-against-clamped stainless steel plates testing	Friction reduction, increase of fatigue resistance and time to breakage	[260]
Heart valves	Graphene monolayer	bi-leaflet mechanical mitral valve surface	Accelerated valve wear testing	Preservation of the coating	[274]
Bio-MEMS/NEMS	Self-assembled rGO/APTES nano-coating	TNTZ	AFM and reciprocating Si ₃ N ₄ ball-on-disk testing	Adhesion, friction, and wear reduction due to a hard cover on molecular springs mechanism	[289,291,292]
	GO/APTES/DA coating	Ti-Al-V	Dry and SBF lubricated reciprocating Si ₃ N ₄ ball-on-disk testing	Enhanced corrosion resistance, friction and wear	[293]

force fields for MoS₂ will help to understand the bio-molecule and MoS₂ interaction in MoS₂-based biosensors.

To conclude, the addition of 2D materials generally improves the anti-wear properties and reduces friction along with the excellent anti-corrosion behavior thus enhancing the performance of bio-MEMS/biosensors. Apart from GO, rGO, and MoS₂, there are more 2D materials including BP [295] or graphitic carbon nitride (g-C₃N₄) [296] used for bio-MEMS/biosensors but their biotribological studies are yet to be explored. Additionally, the studies published to date address the biotribological behavior from a rather fundamental/general perspective and there is a lack of investigations on the influence of 2D materials on the component lifetime of bio-MEMS/NEMS.

4. Concluding remarks

Although notable progress has been made in the design of biomedical devices, issues related to the interaction of the contacting surfaces and the human body still exist thus asking for further development. As outlined in Sections 1 and 2, 2D materials have demonstrated great potential in biotribological/biomedical applications due to their unique physical, chemical, and structural properties combined with large surface-to-volume ratio, tunable surface chemistry, inherent biocompatibility, antibacterial/antiviral activity, as well as non-toxicity. In Section 3, we demonstrated that 2D materials can be employed as protective coatings, fillers in composites, or as additives in fluid mixtures to effectively tailor and manipulate the biotribological behavior. Regardless the application approach, the major role of 2D materials is to enhance the mechanical strength and wear resistance of the systems subjected to stresses, though there are certain advantages and disadvantages of each approach. The incorporation of 2D materials as composite fillers enables control of material release during operation but creates challenges of material uniformity, interface connections and

design modifications. The usage of coatings minimizes the requirements for the changes in the existing implant designs. However, the library of the applicable coatings is limited by the adhesion challenges and compatibility of the deposition techniques with complex designs. Moreover, eventual wear and degradation with no means of replenishment of the coatings significantly reduce their lifetime. Additives in the fluid mixtures enable easier replenishment of the lubrication solution. However, the localized delivery of such solutions is rarely feasible. At the same time, the diffusion of toxic elements to the human body and human organs raises concerns about inflammation and the chronic response of the body.

The applications of 2D materials in combination with the substrate or matrix materials, the biotribological test setup, as well as the main beneficial effects are summarized in Tables 1 and 2. It becomes evident that most investigations (~75%) have been focused on load-bearing and dental implants, while mainly using graphene, GO, or rGO. Considerably fewer studies (~25%) have been dedicated to elucidating the potential of other 2D materials, including MoS₂ or WS₂, in biotribological applications. Similarly, only a limited effort has been focused on other applications, such as bone fracture fixation, endoscopy, root canal treatment, heart valves, contact lenses, or bio-MEMS/NEMS.

Although 2D materials help to enhance the overall biotribological behavior in a wide range of different applications, there are still many challenges. While most studies focused on cytocompatibility, antibacterial properties, and *in-vitro* friction, wear, or corrosion, the long-term *in-vivo* mechanisms have not yet been satisfactorily elucidated in clinical studies. The lack of research on the *in-vivo* cell attachment, bone ingrowth, osseointegration, or interaction with human tissue and blood cells as well as the lack of understanding of the friction, wear, and corrosion mechanisms when exposed to the dynamic human body environment and *in-vivo* operation conditions are currently the main obstacles for the advanced employment of 2D materials. In this regard,

the usage of appropriately designed biotribological test rigs mimicking the real body conditions together with the proper selection of testing conditions and lubricants is of utmost importance. Moreover, more emphasis needs to put the comparability of the findings to allow for a fair comparison of results obtained from different research groups and the transferability to *in-vivo* conditions.

A striking feature of many 2D materials, which has been barely explored in the field of biotribological applications, is their rich surface chemistry, which enables a tailor-made chemical functionalization. We anticipate that this approach is highly suitable to induce specific matter-tissue interactions thus improving adhesion and bone ingrowth, among others. Furthermore, to date, there are hardly any theoretical/numerical approaches to facilitate investigations and simulations of the *in-vitro* and *in-vivo* behavior of 2D materials in biotribological/biomedical applications. Finally, more attention should also be paid to other 2D materials, such as BP, MXenes or MBenes, that have already been partially studied for tribological purposes but not yet in the context of biotribology.

Author contributions

Idea and conceptualization: M.M. and An.R.; Formal analysis: all authors; Writing original draft: M.M., D.B., D.N., N.E., An.R.; review and editing: all authors; Funding acquisition: all authors; All authors contributed to the discussion and have reviewed, edited, and approved the final version of the manuscript.

Declaration of Competing Interest

The authors declare no conflict of interest.

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