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Factors Influencing the Bio-Tribo-Corrosion and Chemistry on Cobalt Alloys: A Brief Literature Review

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Abstract. Cobalt-chromium-molybdenum (CoCrMo) alloys are commonly employed for load-bearing implants, such as hip and knee prostheses, owing to their mechanical properties and excellent passivity characteristics to reduce corrosion. The interaction between biological environment and the metallic surface under an articulating condition is extremely complex. The metal is exposed not only to the harsh biological environment as well as mechanical loading, leading to the conjoined action of mechanical (wear) and electrochemical (corrosion) degradations of the material; termed as tribo-corrosion. Tribo-chemical film formation can affect the surface response towards tribo-corrosive processes. The fundamental mechanisms of the tribo-corrosive degradation and surface chemistry are recognized to contribute to the orthopaedic implants' longevity problem; an understanding of those mechanisms is therefore essential. This manuscript aimed to briefly review the current knowledge of the tribo-corrosion and tribo-chemistry phenomena from the pre-clinical studies' point of view on the CoCrMo alloys used for hip bearing implants. This overview highlighted that the tribo-corrosion and tribo-chemical reactions during sliding are sensitive to the biological species interaction, as well as several unexplored factors in the environment. These findings contribute to the further knowledge and pre-clinical understanding of protein-metal interactions occurring in films formation and the system variables effect on the metallic load-bearing surfaces.

Keywords: biomedical alloys, bio-tribo-corrosion, tribo-chemistry, hip implant, CoCrMo alloy

INTRODUCTION

Total joint replacement is a common type of orthopaedic surgery, where the natural joint is replaced by an artificial one (prosthesis or implant) to restore function and relieve pain when it becomes damaged or worn [1]. Joint replacement become one of the most successful surgical inventions for osteoarthritis since the seminal work by Sir John Charnley. This development has been through several stages of design and material improvements to date. The number of procedures for the total joint replacement, in this case, is hip replacement, has increased in the last 15 years and is expected to keep growing [2, 3]. Metallic-based alloys are one of the reliable materials used for the Total Hip Replacement (THR), other than polymers and ceramics. The second generation of Metal-on-Metal (MoM) was reintroduced in the 1990s as, promising a lower aseptic loosening rate for the younger patients and smaller wear particle size compared to polymer bearing [4]. However, the developing risk of adverse tissue reactions due to the released metal ions and debris become a matter of concern for both experts and patients. Thus, several products have been recalled from the market and withdrawn from use [5]. Nevertheless, MoM hip resurfacing and other metallic-coupled bearings (e.g., Metal-on-Polymer (MoP) and Ceramic-on-Metal (CoM)) are still being implanted today with reported success [6].

Since the issues of implant-derived debris, tribology (interacting surfaces in a relative motion [7]) and corrosion (electrochemical degradation) of metallic hip prosthesis have been brought to the fore in the last decade, burgeoning research activities designed to understand the degradation mechanisms associated with the implants have been

undertaken. Whilst studies [8-13] have recently suggested that the success of load-bearing implants is, in part, due to the chemical reactions between the biological species from synovial fluid and the metal surface during sliding, the material degradation mechanisms are still largely unknown due to the complex environment. Therefore, the knowledge gap motivated this manuscript to review the corrosion and surface chemistry roles on the cobalt alloys from several pre-clinical studies' point of view. The aim was to introduce the understanding of the tribo-corrosion and tribo-chemistry phenomena, as well as highlight various potential influencing factors as critical aspects to be considered in future studies. This overview is also relevant to other devices where at least a metallic component works at the tribological interface.

CORROSION IN TRIBOLOGICAL INTERFACES

Corrosion is thorny problem for metallic implant longevity and biocompatibility, entailing the destruction or loss of material due to an electrochemical reaction with its surrounding environment [14]. Biomedical devices commonly use passive-type metal alloys, such as cobalt-chromium alloys, owing to their ability to form a passive oxide film for corrosion resistance. The passivation is a secondary reaction, following metal oxidation in Equation (1) as the immediate reaction. Equation (2) demonstrates how the oxidized chromium from the metal surface reacts with the water or oxygen from the environment at the metal-solution interface. The reaction produces a passive oxide film containing chromium oxide (Cr_2O_3), which acts as a protective barrier to inhibit the charge transferred from the bulk metal surface to the electrolyte [15].

$$\frac{1}{2}M^{n+} + \frac{n}{2}H_2O \rightarrow \frac{n}{2}MO + nH + ne^-$$
(1)

$$2Cr^{3+} + 3H_2O \rightarrow Cr_2O_3 + 6H + 6e^-$$
(2)
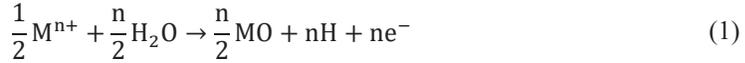
However, the passive film can be removed mechanically under tribological conditions, leaving the bare surface exposed to the environment. Thus, the metal ion can be pulled out from the exposed surface due to the rapid oxidation process. The combined processes are termed as tribo-corrosion; a complex synergism of surface degradations due to mechanical wear (i.e., friction, abrasion and erosion) and chemical/electrochemical interactions with an aqueous/corrosive environment [16]. Therefore, the rate of removed material when both mechanisms take place will be higher than when each is present individually [16, 17]. Currently, there are several approaches to quantitatively evaluate the tribo-corrosion mechanism, such as synergistic, mechanistic, third-body and nano-chemical wear [16, 18].

In 1995, Watson et al. [19] established the synergistic approach to calculate wear-corrosion synergism, that has been standardized in ASTM G119 [20]. This approach demonstrates that the overall material loss is affected by a synergism factor (S), as in Equation (3). The mechanical wear mechanism not only produces a pure mechanical loss (W_0) but also change the corrosion rate due to wear (ΔC_w) that coincides. The electrochemical process occurs as pure passive corrosion (C_0) outside the contact, as well as changes the wear due to corrosion (ΔW_c). As a result, all products can be accumulated as a total of degraded materials (V_{total}, while using volumetric unit).

Equation (4) shows the synergistic components. The interactions can be classified into 'antagonistic or additive' and 'synergistic' [21]. An antagonistic interaction is identified when the corrosion and wear interplay decreases the total material loss, producing a beneficial effect, e.g. tribofilm formation in an MoM's hip joint due to sliding with protein presence [22]. On the contrary, a synergistic interaction is found when corrosion increases the wear and total material loss. Passive oxide film removal due to sliding or fretting can be an example of a synergistic behavior [21].

$$V_{total} = W_0 + C_0 + S$$
(3)

$$S = \Delta C_w + \Delta W_c$$
(4)

This section describes the aforementioned equation into a chronological stage of the synergistic approach in a sliding cycle, as summarized by Fuentes et al. [23]. In metal passivity, it has been explained that passive-type metals will form a passive oxide layer spontaneously to resist corrosion. After the two articulating surfaces are rubbed with various normal load, velocity, and direction, the passive oxide layer and other wear debris are destroyed and removed from the surface (de-passivation). The fresh area at the de-passivated surface is exposed to the electrolyte and available

for corrosion (wear-enhanced corrosion, or C_w henceforth). The corrosion roughens the surface and simultaneously causes corrosion-enhanced wear (or W_c henceforth). Meanwhile, the surface reforms the passive oxide film at some time after the removal (re-passivation). On the other hand, the removed wear debris (containing the oxide layer, metal ions, and the removed bulk surface) can oxidize, bind and react with the electrolyte or biological species. The metal debris-electrolyte products, such as hard oxides or other complex compounds, may accumulate and contribute as third-bodies of abrasive wear in the tribological contact, which increases the wear loss (corrosion-enhanced wear, or W_c henceforth). The stages are repeated in a cyclical process as long as there is a tribological condition; either sliding or fretting contacts.

BIO-TRIBO-CHEMICAL INTERACTIONS ON METALLIC SURFACES

The synovial joint fluid, as the natural lubricant for the cartilage inside the human body, contains protein, glucose, uric acids and many other nutritional bio-substances. As the predominant species in the joint fluid, proteins were observed to adsorb on the exposed surface; affecting the behavior of passive oxide film at a metallic implant surface, causing less stable metal-oxide bonding and changing the corrosion reaction [24]. Moreover, the protein species, such as albumin, was known to have a high affinity with metal ions; thus, it can interact with the dissolved metal ions in the bulk solution [25, 26]. Under a tribological contact, proteins were reported to form a biofilm on the articulating surface, which deposited a complex structure of inorganic and organic compounds and enhanced the lubricity [13]. The surface potential, along with the contact pressure and normal load in the articulating surfaces, can affect the lubricating ability of the protein films [13, 22, 27]. Hence, it is useful to discuss the chemical reactions between biological species and the metallic surface under tribo-corrosion condition; termed as tribo-chemistry.

The tribo-chemistry can be referred to as the reactions that chemically occur at a tribo-contact activated by friction, shear stress and wear mechanism [28, 29]. Generally, the reaction generates a tribo-chemical film (tribofilm or tribomaterial) that is influenced by affecting factors, such as contact pressure [30], sliding speed [31], sliding distance [32], lubricant constituents [33], rubbing cycle [34] and surface materials [35]. The tribo-chemical reactions in the hip bearing occur when the biological species in the synovial fluid interact either with the articulating implant surfaces or the other species in the body fluid, which eventually produce such deposited or precipitated organometallic compounds and passive oxide film that help to reduce the friction and degradation at the contacting bodies [36].

OVERVIEW OF THE INFLUENCING FACTORS IN PRE-CLINICAL STUDIES

Bio-tribo-corrosion studies have been conducted using several methods for controlling the testing conditions to make them as close as possible to a real implant environment. In order to mimic these conditions, tribometer and hip simulators are usually employed with suitable sample materials, biological fluids and environmental settings. In our research group, the bio-tribo-corrosion studies by Yan [37], Hesketh [38], and Beadling [39] used either a reciprocating pin-on-plate tribometer or hip simulator to re-create the sliding motion of the hip bearings. Bryant [40] and Oladokun [41] also investigated the bio-tribo-corrosive behavior with the simulator and fretting tribometer for a fretting motion of cemented stem and modular taper, respectively. The three-electrode cells were commonly employed to assess *in situ* or control the corrosion in the tribo-couples, instrumented to a reciprocating pin-on-plate tribometer. The protein-containing serum was used as the lubricant to simulate the synovial joint fluid in the human body [9, 31]. Several surface analyses, including the wear volume, roughness and chemistry, are typically conducted following the bio-tribo-corrosion (sliding) tests.

Espallargas et al. [11] categorized the possibility of tribofilm specifications in a metallic implant based on published results, i.e. (i) a pure passive Cr-oxide film with thickness up to 100 nm, (ii) layer of organic species covering the metal surface without a transitional oxide film and (iii) a high organic species concentration with thinner Cr-oxide film covering the metal surface. These categories are for metallic implant since an oxide film automatically reacts when the passive-type metal surface is in contact with the environment. However, the combination of the films might be the closest formation to the real condition. It seems that the formation of organic species must compete with the bonding of metal-oxide in the passive film during the deposition process. Fan et al. [9] found that this proteinaceous layer build-up occurs in the boundary lubrication regime. As a result, the protein film can affect the tribological responses; e.g., by reducing friction and wear loss on the metallic surfaces. Mathew et al. [42] observed that protein resulted in a 23% decrease of the total volume loss from the CoCrMo surfaces when tested under boundary lubrication conditions. Liao et al. [12] observed the tribological layer in the hip resurfacing MoM with self-mated CoCrMo alloys. Using EELS and Raman spectroscopy, the signal indicated clear evidence of the presence of graphitic carbon as the

main structure of tribofilm. The graphic carbon deposition was also claimed to reduce friction and protect the surfaces from wear as well as corrosion. Furthermore, the authors have identified Mo-S speciation via XPS in a previous study and hypothesized to influence the wear and friction during tribo-corrosion [43]. However, it still needs to be clarified how the protein can degrade into those complexes under the sliding condition, which produces the solid lubricant and protective effect associated with the denaturing state of the protein structure.

Figure 1 illustrates the key factors that should be considered in order to understand tribo-corrosive degradation and tribo-chemical film phenomena for articulating hip implants, based on the pre-clinical studies reviewed in this section. Previous study have shown that the tribo-corrosion and tribo-chemistry behaviors are sensitive to the change in electrolyte species [43]. Several other factors that might potentially affect the system are discussed in the following sub-sections, based on several pre-clinical studies.
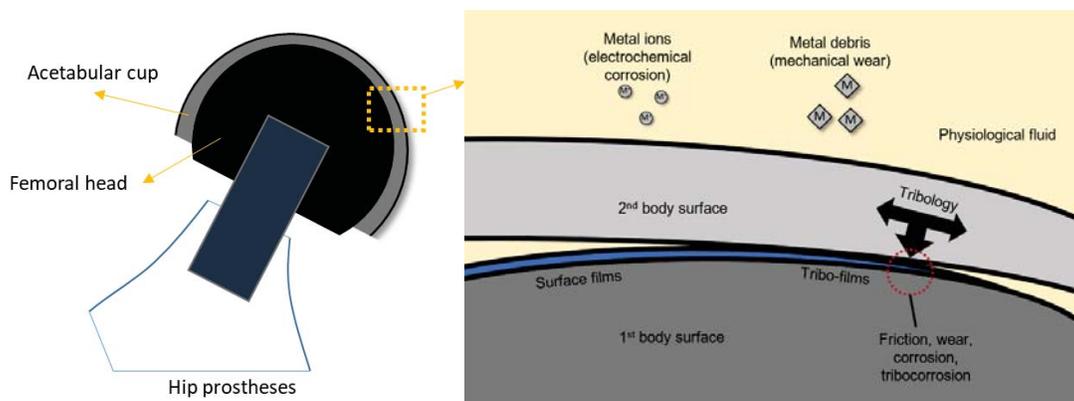

FIGURE 1. (a) A schematic for the contacting bearings in total hip replacement with a modular interfaces design and (b) key parameters related to the tribo-corrosion and tribo-chemical film phenomena for articulating the implant surfaces

Electrochemical Potential Condition

The tribo-corrosion process is driven by the difference in the electrochemical potential established across the surface affecting the material passivity and the formation of the surface film. Metal implants inside the human body establish electrode potentials which are inconsistent over the whole surface [44]. These phenomena can be shown on removing passive oxide film while rubbing, resulting in a potential drop compared to the remaining area resulting in localization of anodic/cathodic areas and increased charge transfer [45]. The change in the electrode potential can be influenced by the surface pH [46], metal ion release [47], material and biological fluid [33, 48]. These indicate that the actual value of the surface potential in the body can vary greatly and even differ at different implant area sites.

Wang et al. [44] reported that applied potentials (comparing at +0.2 V vs silver-chloride (Ag/AgCl) electrode, Open Circuit Polarization (OCP) and -0.8 V vs silver electrode) affect the subsurface deformation. The result indicates that the surface potential modified the passivity and protein adsorption rate on the CoCrMo surface. A study by Kerwell et al. [49] evaluated tribofilms formed on CoCrMo alloy at various anodic potentials (-0.4, +0.6, +0.7 and +0.8 V). The results showed that electrochemically-induced surface films at the specific potential condition could affect corrosion resistance. Thus, these results indicate that electrochemical conditions can be sensitive, even with a small potential difference, in changing the protein adsorption on a metallic implant. Munoz et al. [50] highlighted that the protein adsorption rate is higher at cathodic -1 V than observed at the more passive potentials. Yan et al. [37] attempted to decrease the total volume loss of cobalt-based alloy under sliding at -0.8 V. Therefore, it is essential to investigate the effect on film formation in the organic-containing lubricant under tribological and applied potential conditions.

Flash Temperature and Protein Concentration

Proteins are known to be sensitive to elevated flash temperatures and shear [51], all of which are present within a tribological contact, leading to the re-arrangement of and damage to the protein structure [11]. A higher temperature than the normal human body condition can weaken and even break the disulphide chain, which results in an unstable

secondary structure and reduced protein biological activity [52-54]. A study by Mishina and Kojima [55] found that protein denaturation begins to occur in temperatures ranging from 323 to 328 K. Lin et al. [56] reported that the conformational protein structure is reversible at temperatures below 323 K. However, it becomes irreversibly unfolded at temperatures 333 K. In another study, Wimmer et al. [57] estimated that the decomposition of protein occurs at above 330 K. As reported by Bergmann et al. [58], the implant temperature could reach a range of 293-331 K, measured *in vivo* using an instrumented hip implant (at a sampling rate of 2-10 Hz and accuracy of 0.1°C) with telemetric data transfer. Their study has proven that the artificial joint temperature can increase, depending on the walking cycles and speed, material combinations and surrounding environment temperature. It is hypothesised that the protein will more easily form a tribofilm when in a denaturing state.

Previous studies have successfully investigated the effect of protein concentration and temperature on the corrosion and tribo-corrosion properties of CoCrMo alloys. Namus et al. [59] successfully employed Electrochemical Impedance Spectroscopy (EIS) to investigate the effect of protein (calf bovine serum) concentration and temperature on the CoCrMo surface responses under cathodic polarisation to simulate the unworn area, that will be more cathodic. A significant reduction in oxide film resistivity was observed with the increasing concentration of protein and temperature up to 323 K. Atomic Force Microscope (AFM) confirmed that the protein concentration in the electrolyte influences the density of the surface adsorbed protein [60]. These findings indicate that the temperature could affect the protein adsorption and electrochemical processes on the metal surface. Vidal et al. [61] also used electrochemical tests to observe the effect of temperatures ranging from 298 to 333 K and the protein concentration of bovine serum albumin on the adsorption process. The results clearly show that the higher temperatures will increase the passive film dissolution rate of CoCrMo alloys and reduce the transpassive (pitting) potential. During static immersion, the bulk protein concentration has been proven to accelerate the anodic reaction since the adsorbed protein can bind and transport the metal ions away from the solution-surface interfaces.

During sliding, the protein concentration's effect on the tribo-corrosion of CoCrMo alloys has been investigated by Sun et al. [62]. The results show that the protein's increasing concentration can enhance the adsorbed protein density and thickness, reducing the polarisation resistance and increasing lubricity of the surfaces. However, the research on the effect of protein-metal interactions during tribo-corrosion as a function of temperature remains limited. It is important to investigate the effect of elevated temperature and protein concentration on the tribo-corrosive behaviour at the metallic surface in order to develop a clearer understanding about the failure mechanism of the metal bearing.

Dissolved Metal Ions

It is well-known that the released metal ions in the physiological fluid can cause the adverse local tissue reactions. There were only few studies that have indicated the effects of the dissolved metal ions on the implant system. The protein interaction with molybdenum ions was assumed significantly to affect the passivity behaviour on a CoCrMo surface [63, 64]. The molybdenum-protein interaction then increases the tribofilm performance to reduce wear and corrosion on the metal implant. Indeed, there are specific conditions for sliding and the electrochemical potential applied. The interaction then leads to the growth of surface film formation. A recent study by Martin et al. [65] found that Mo ions can deposit the proteins to the surface compared with the Co and Cr presence. This result shows an independent role for each metal ion type (Co, Cr, and Mo in this case) on the protein films' deposition. However, the interaction between the metal ions and the proteins, as well as their effect on material degradation during tribo-corrosion, are not well established. The effect of dissolved Co and Cr ions on the surface chemistry and degradation is still not explored yet. Even though there have been only a few studies about the role of Mo ion in the adverse reaction, it was hypothesised that dissolved metal ions in the bulk solution could interact with the proteins, influencing the tribo-corrosion and surface chemistry of the metal implant.

CONCLUDING REMARKS

A combined and holistic understanding of the fundamental mechanisms of wear and corrosion, or tribo-corrosion, has been acknowledged as a key enabling technology to enhance the longevity of orthopaedic implants. This overview has highlighted the current understanding of the tribo-corrosion and tribo-chemistry phenomena of the CoCrMo alloys from the related pre-clinical studies. Also, this review contributes to the pre-clinical understanding of protein-metal interactions occurring in tribofilms' formation and the potential system variables effect on the metallic bearing surfaces. Tribo-chemistry, an interaction between electrolyte species and a metal surface during the articulation of

surfaces resulting from shear stress and/or chemical interactions, is also shown to affect surface degradation. However, due to the complex nature of the environment, researchers are still striving to create the optimal system.

Based on this literature review, the following factors are indicated to have important role upon the behaviors of tribo-corrosion and tribo-chemistry on the CoCrMo alloy:

- The understanding of the tribo-corrosive behaviour and tribo-chemical film processes of the CoCrMo surface in several organic-containing electrolytes and electrochemical potential conditions remains limited.
- The mechanisms pertaining to the formation of the tribofilms, when subjected to various protein conformations and concentrations, still requires further investigation.
- Besides the biological tissue reaction, the role and effect of released metal ions, e.g., Co, Cr and Mo ions, from the CoCrMo alloy surface in the physiological fluid, on the surface tribo-chemical film and tribo-corrosive degradation, remain largely unknown.

Therefore, the understanding of the tribo-corrosive behavior and tribo-chemical film processes of the CoCrMo surface in several organic-containing electrolytes and the aforementioned influencing factors needs to be further investigated in the future experimental studies.

ACKNOWLEDGEMENT

CONFLICT OF INTEREST

No conflict of interest was reported by all authors.

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