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TRIBOCORROSION – INSIGHT INTO MATERIAL DEGRADATION IN SPECIFIC ENVIRONMENTS

Sorin-Bogdan BAŞA[1,2], Lidia BENEA[1,*]
[1]Research (Competences) Centre: Interfaces-Tribocorrosion and Electrochemical Systems (CC-ITES), Faculty of Materials and Environmental Engineering, ”Dunărea de Jos” University of Galati, 47 Domnească Street, RO-800008, Galati, Romania
[2]Department of Metallurgy and Materials Engineering, Katholieke Universiteit Leuven, Kasteelpark Arenberg 44, B-3001 Leuven, Belgium
[*]Corresponding author
e-mail: Lidia.Benea@ugal.ro

ABSTRACT

Tribocorrosion is an irreversible surface degradation mechanism of a metallic and/or non-metallic material as a result of its physicochemical and mechanical interactions during relative motion (friction, sliding, impact, abrasion, erosion) in corrosive media. It encompasses synergistic effects between mechanical wear and chemical / electrochemical processes which interact with each other. Tribocorrosion processes lead to uneconomical material loss as well as to the decreasing of the following characteristics: durability, reliability, safety, performance, energy efficiency, pollution and health. Recent activity in tribocorrosion research aims at addressing the need to select or design new surfaces for future equipment as well as minimizing the operating costs and extending the life of existing machinery and medical devices. The work presents an overview and some experimental results from tribocorrosion tests of biomaterials and nanocomposite coatings in specific environments from physiological solutions to industrial environments.

KEYWORDS: corrosion, tribocorrosion, nanocomposite coatings, biomaterials, material degradation, specific environments

1. Introduction

Tribocorrosion is regarded as the science of surface transformations as a result of the chemical reactions and mechanical disturbances that occur between the elements of a tribo-system exposed to a corrosive environment.

In the recent years, ***tribocorrosion*** a, research activity that combines the science under focus corrosion and the science of tribology, has been focused by both engineers and scientists from different research areas, contributing to the development of a new topic of research beyond conventional ones.

Tribocorrosion involves mechanical and chemical/electrochemical interactions between surfaces in relative motion between one another, and in the presence of a corrosive environment. The tribocorrosion process is common in many areas where it can cause premature destruction of devices, equipments and even vehicles. It is also found in living systems in case of metal implants in the human body such as artificial joints, orthopedic plates and screws and dental implants. Therefore all of this complex tribocorrosion process is studied and researched in terms of two broad categories of applications [1-10]:

- tribocorrosion in industrial systems;
- tribocorrosion in living systems.

Tribocorrosion applications

The study on tribocorrosion evolves to be an active research area, due to its wide existence in a variety of industries, such as mining, oil, automotive, food, nuclear, offshore marine and biomedical, Figure 1 (a-f). Tribocorrosion is a surface degradation process resulting from simultaneous tribological and electrochemical actions in a corrosive environment [11-14].

The tribocorrosion process cannot be simply predicted from the knowledge of isolated wear and corrosion behaviors of the material, since synergistic effects of these two processes can accelerate the mass loss in the tribocorrosion test. Tribocorrosion can cause material degradation, and affects the friction, wear and lubrication behavior of the tested materials.

In the biomedical applications the tribocorrosion process is increasingly interesting to researchers and is studied under two major aspects: orthopaedic science and surgery and dental implants.

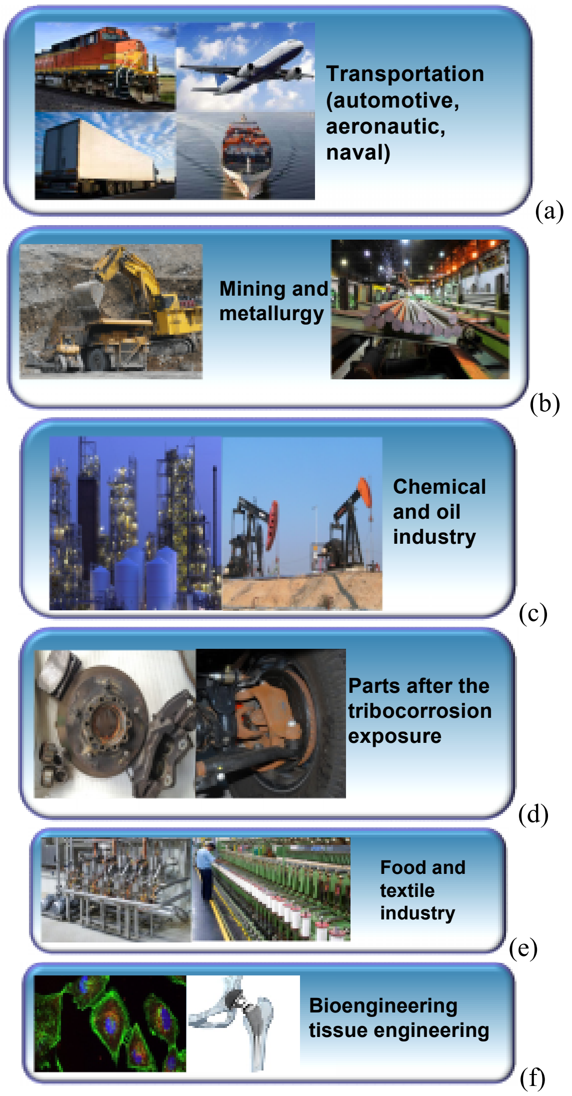


***Fig. 1**. The variety of tribocorrosion process applications and needs of knowledge: (a) - transport; (b) - mining and metallurgy; (c) - chemical and oil industry; (d) - different parts of mechanisms in moving conditions under corrosive environments; (e) - food and textile industry; (f) - bioengineering and biomaterials*

***Orthopaedic science and surgery**.* The tribological aspects of bone implants, particularly hip and knee joints are addressed by many researchers to increase their lifetime and to avoid that patients undergo too often a repair surgery (recovery). However, as the tribological aspects in such implants are influenced by human body fluids at the interface (Periprosthetic fluids), the complex tribocorrosion process must be taken into account. The tribocorrosion process can occur in hip joint implants and many authors already stress the importance of better understanding the electrochemical behavior of metallic implants after mechanical destruction and surface destabilization and the protective passive film stability in the presence of simulated body physiological solutions [1].

Dental implants are another area where tribocorrosion processes have direct applications. In fact, each mastication or biting process is a tribocorrosive cycle, friction taking place between teeth and food particles in the presence of liquid called saliva. The behavior of materials used is strongly influenced by the solutions pH and acidification improves the electrochemical response of the material. Evolution of the material repassivation is strongly influenced by the nature of the electrolyte [1].

The tribocorrosion processes are still poorly understood and explained, the literature being still quite poorly represented. Investigation of the tribocorrosion processes with industrial and biomedical applications involve a multidisciplinary approach to know and understand a system composed of a surface of a material, an environment (electrolyte, solution, human fluid) and a mechanical contact [1].

Integration of researchers from several disciplines such as materials science, mechanical engineering, electrochemistry/chemistry, tribology, biology and medicine may lead in the future to better knowledge and interpretation results.

The ***tribocorrosion*** process can be characterized by its synergy effects resulting from the combination of mechanical and environmental effects, Figure 2.

This synergism leads to degradation, and thus to a loss of material, which is often much greater than the one which we would expect by simply summing the degradation of the two processes separately.

$$W_T \neq W_E + W_M$$

Where: W_t – total volume of the lost material; W_E – volume of material lost by electrochemical corrosion and W_{M-} volume of material lost because of mechanical wear.

There is a synergetic interaction between friction and corrosion. Friction induces tension in materials and plastic deformation, residual stress and in a number of materials can even cause structural modifications.

Also, friction induces local destruction of the layer formed by material surface interaction with the environment. In general, the adsorbed layers are present on surfaces. Passive layers or corrosion products may also be present on surfaces. On the passivable materials, a thin (passive) layer protects material against corrosion. As a result, the reactivity of the surface is deeply modified by the friction on the contact surface and thus it modifies completely the kinetics of corrosion and passivation. On the other hand, corrosion, or electrochemical reactions that occur on the material surface, can strongly influence the tribological conditions and/or mechanical wear.

The surface composition is modified by electrochemical reactions and particularly by corrosion, as a result, the mechanical properties of the surface change too, along with the resistance to mechanical wear.

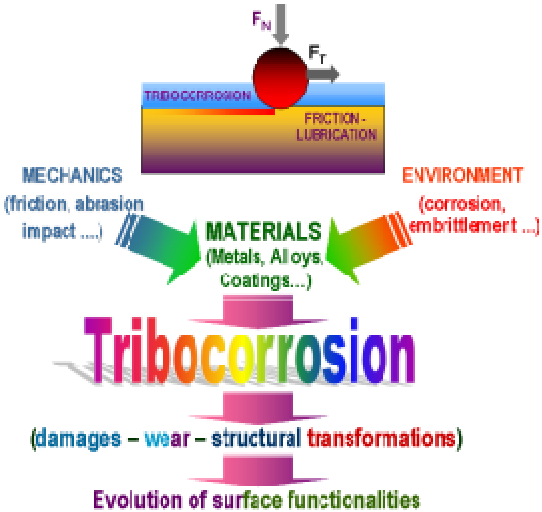


Fig. 2. *Synergy of friction and corrosion processes occurring simultaneously on the surface of a material*

Corrosion by-products may contribute to the formation of a third body in the process of degradation. They may also lead to changes in the composition and properties of lubricants. Finally corrosion affects geometric characteristics and surface roughness. Consequently the action of corrosion on a tribo-surface can substantially alter the friction coefficient, and the kinetics and mechanism of wear.

First tribologist 2400 B.C.

Tracing back the history, we may find that the understanding of the role of friction in man-made machine certainly goes back to the ancient Egyptians (ca. 2500 BC). Overcoming friction in moving large pieces of stones and statues must have been a great challenge for them.

Some carvings and images of that time tell that the use of water might have given some reduction in the friction and thus less work for the slaves who had to carry out this monumental task of building the Pyramids, Figure 3 (a, b). Also, there is a clear indication of the use of lubricated wooden planks to reduce contact area in moving very large pieces of stones or statues. A situation much closer to that of today's time was faced in the case of the lubrication of the chariot wheel hubs, for war or other usages. Around 1500 BC, the art of chariot building was quite advanced in Egypt and the use of leather and animal fats to lubricate the wheel hub was generally practiced [15].

A more famous Tribologist - 500 years ago or another Leonardo da Vinci revelation?

Modern tribology began some 500 years ago, when Leonardo da Vinci deduced the laws governing the motion of a rectangular block sliding over a planar surface [16, 17]. Leonardo da Vinci (1452-1519) can be named as the father of modern tribology.

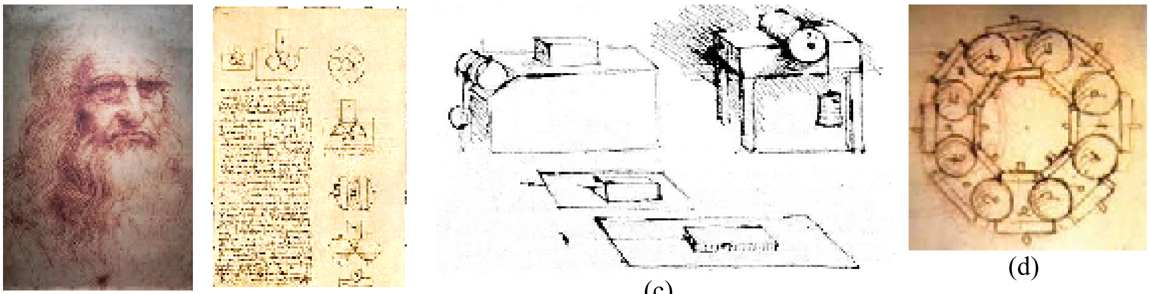
(a)

(b)

Fig. 3. *Indication of the use of tribology in ancient Egypt: (a) - Transporting the statue of Ti – from a tomb at Saqqara, Egypt; (b) - The first recorded tribologist – pouring lubricant (water or oil) in front of the sledge in the transport of the statue of Ti*

He studied an incredible manifold of tribological subtopics such as: friction, wear, bearing materials, plain bearings, lubrication systems, gears, screw-jacks, and rolling-element bearings. 150 years before *Amontons' Laws of Friction* were introduced, he had already recorded them in his manuscripts. Hidden or lost for centuries, Leonardo da Vinci's manuscripts were read in Spain a quarter of a millennium later [16, 17]. In Figure 4 there are presented some aspects of tribological studies from his manuscripts.

Leonardo da Vinci introduced the first concept of friction. He found the dependence of friction on load and the independence of geometrical contact area.



(a) (b) (c) (d)

Fig. 4. *Leonardo da Vinci's Studies of Friction from writings and sketching in the Codex Atlanticus, the Arundel MSS. 263 and the Codex Madrid discovered as recently as 1967 confirm his acute recognition and understanding of many basic features of tribology: (a) Leonardo da Vinci; (b) Ball Test Geometry; (c) - Sled Friction Test Geometry; (d) Leonardo's Ball Bearing with Cage*

Nanotribology

Since the 1990s, new areas of tribology have emerged, including the nanotribology, biotribology, and green tribology. These interdisciplinary areas study friction, wear and lubrication at the nanoscale (including the Atomic force microscopy and micro/nano electromechanical systems, MEMS/NEMS), in biomedical applications (e.g., human joint prosthetics, dental materials), and ecological aspects of friction, lubrication and wear (tribology of clean energy sources, green lubricants, biomimetic tribology).

Micro/nanotribology as a field is concerned with experimental and theoretical investigations of processes ranging from atomic and molecular scales to the microscale, occurring during adhesion, friction, wear, and thin-film lubrication at sliding surfaces [18-20].

This involves determination of the chemical, physical and mechanical properties of the surfaces undergoing relative motion at length scales of the order of nanometers. Interaction between rubbing surfaces occurs at asperities [roughness of surfaces] at which the local pressure and temperatures can be very high.

These conditions can lead to formation of tribochemical films with the unusual properties necessary for efficient wear protection. The nanomechanical properties of these films are being investigated by interfacial force microscopy (IFM) which is capable of determining the elastic constants and unelastic behavior of the films in boundary layer lubrication [18-20].

The potential for nanotechnology to transform civilization as we know it is breathtaking and the nanomechanical systems of the future will all require new atomic lubrication schemes to overcome the debilitating effects of friction. In order for this impending revolution to be fully realized, we need a fundamental understanding of friction and corrosion at the atomic to meso-scale.

Electrodeposition, nanocomposite coatings (layers) and tribocorrosion systems

Electrodeposition of new and advanced nanocomposite coatings is a process of low energy consumption, and therefore very convenient for the surface modification of various types. Published research works in tribocorrosion (wear - corrosion) behaviour of electrodepositd nanocomposite coatings are also in the attention of our researchers starting from 2001. One example is the starting work dealing with electrodeposition and characterisation of Ni-SiC nanocomposite coatings, which since its publication (2001) has generated over 186 citations in ISI international journals [21, 22].

Tribocorrosion process and damage effects

As it is reflected in Figure 5, corrosion ***leads to***: material loss, reduced reliability, reduced durability, reduced energy efficiency, reduced safety performance, increased health hazards, increased pollution, etc.

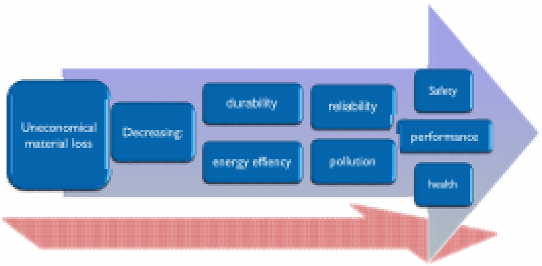


Fig. 5. Damage effects of tribocorrosion processes

2. Experimental set-up for tribocorrosion study

Electrochemical Cell

Uni-directional or bidirectional pin-on-disk contact geometry can be used and an electrochemical cell connected to a tribometer. Schematic set-ups for tribocorrosion studies are given in Figure 6.

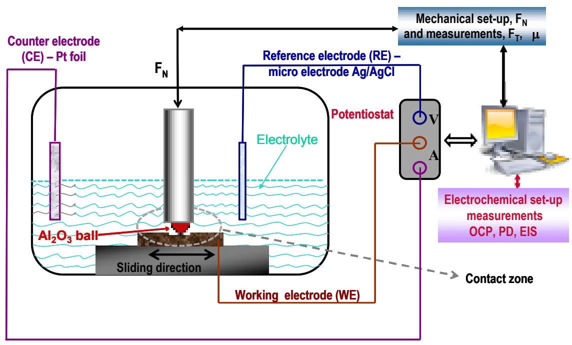


Fig. 6. *Description of the pin-on-disc set-up used to study the tribocorrosion behavior of materials and coating in aqueous solution: the electrochemical cell connected to potentiostat reference electrode (RE), counter electrode (CE), working electrode (WE) is the sample with the material or coating on the top of a cylinder or a plate, the mechanical set-up with counterbody (pin), made by corundum (Al_2O_3), wear track on the surface of the sample. The experiment is controlled by PC for imposed mechanical - electrochemical parameters and mechanical - electrochemical data*

The counter body (pin) is usually a hard ceramic like alumina (Al_2O_3) or zirconia (ZrO_2) cylinder (7 mm in diameter), mounted vertically on a rotating head, above the sample. The lower spherical end (radius = 100 mm) of the pin is then applied against the material surface (disc) with an adjustable normal force, correlated with the real application. When rotation is applied, the end of the pin draws a circular wear track (16 mm in diameter) on the working surface (for the uni-directional contact geometry) or a wear track with an amplitude of about 200 µm (for bi-directional contact geometry).

In – situ electrochemical measurements

In-situ applied electrochemical methods are: the open circuit potential measurements (OCP), the potentiodynamic polarization measurements (PD), the potential step from active to passive state and the electrochemical impedance spectroscopy (EIS).

Open circuit potential measurements

The measurement of open circuit potential gives information on the electrochemical state of a material, for example active or passive state in the case of passivable materials. However, open circuit potential measurements provide limited information on the kinetics of surface reactions. The open circuit potential recorded during uni-directional pin-on-disk sliding tests, in which the disk is the material under investigation, is a mixed potential reflecting the combined state of the unworn disk material and the material in the wear track. One must be aware that a galvanic coupling between worn and unworn parts on the disc surface may take place. The measured open circuit potential is an average value depending on current density distribution over the disk surface.

In - situ mechanical measurements

During the sliding tests, the normal force, tangential force, coefficient of friction, number of cycles, displacement amplitude as well as the open circuit potential are recorded.

Ex-situ wear track investigations

All samples with wear track can be examined ex-situ by SEM and high surface microtopography to evaluate the damages provoked and to estimate the mass, track depth or volume loss.

3. Experimental results for Ni/nano-WC layers used in industrial applications

Working on developing environmentally friendly materials with high corrosion and wear resistance by electrodeposited coatings were obtained ***Ni/nano-WC layers*** on 304L steel support. In this way it could be offered to the industry improved materials for longer and more efficient use in the cooling system of the nuclear power plants.

The Ni/WC nanocomposite coatings obtained by electro-codeposition of WC nanoparticles (60 nm mean diameter) with nickel from a dispersing nickel plating bath were characterized comparatively with pure nickel coatings for tribocorrosion behaviour.

Reciprocating sliding wear tests (as in the set-up presented in Figure 6) were performed in solution containing LiOH and H_3BO_3 on the sample sliding against corundum balls (10 mm diameter) for 10000 cycles. Al_2O_3 ball was selected as a counter body due to its high wear resistance, high chemical inertness and high electrical resistance. Tests were done at normal load of 5 N displacement amplitude of 200μm and the sliding frequencies of 1 Hz.

The coefficients of friction performed during wear tests in wet conditions is shown in Figure 7. During friction in the wet environment the coefficient of friction oscillates between values. As an average it was appreciated 0.20 for Ni/nano-WC and 0.37 in the case of pure Ni at the normal applied force of 5N (Figure 7).

The open circuit potential (OCP) data recorded on the Ni/nano-WC and pure Ni coatings before, during and after fretting tests in the solution, are shown in Figure 8. The sliding tests were done at the normal forces of 5 N (Fig. 8), at 1 Hz fretting frequency with displacement amplitude of 200 μm, for 10000 cycles.

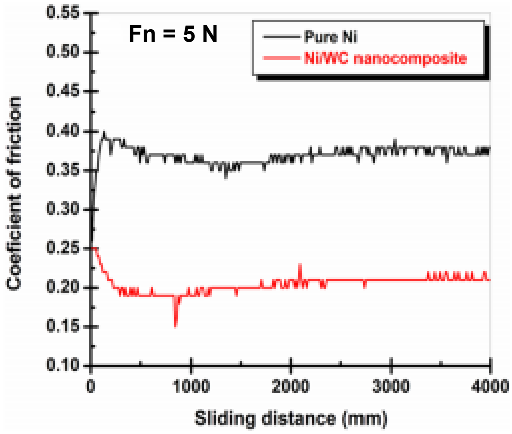


Fig. 7. *Evolution of the friction coefficient of Ni/nano-WC composite and pure Ni coatings during fretting wear tests performed in wet conditions (solution with LiOH and H_3BO_3) at 1 Hz, 200 μm, 10000 cycles for the normal force of 5 N [L. Benea and group - unbublished work]*

In the time interval preceding the starting of corrosion-wear fretting, a large increase of the open circuit potential for both Ni/nano-WC composite and pure Ni coatings is noticed. It can be observed also that before the start of the sliding tests, the potential of the samples is quite stable. Also it should be mentioned that the potential measured before sliding tests of Ni/nano-WC nanocomposite coatings is more ennobled as compared to pure Ni coatings, thus leading to a significant increase of anticorrosion effect. This means that an ennoblement of the coating surface occurs when WC nanoparticles are incorporated into the electrodeposited nickel layer.

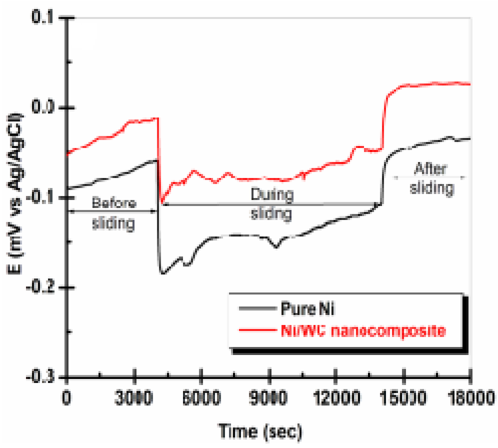


Fig. 8. *Evolution of the open circuit potential recorded before, during and after sliding tests of Ni/nano-WC composite and pure Ni coatings in the boric acid and lithium hydroxide solution at 1 Hz, 200 μm, 10000 cycles for the normal force of: 5 N [L. Benea and group - unbublished work]*

4. Experimental results for surface modified Ti-6Al-4V alloy used in biomedical applications

Working on enhancing corrosion and tribocorrosion properties of biomaterials in simulated body fluids solution, two electrochemical methods for surface modification of Ti-6Al-4V were applied. in this way it could be offered to biomedical applications improved biomaterials surfaces for longer and more efficient use.

The evolution of the friction coefficient versus sliding distance (mm) measured on untreated and surface modified Ti-6Al-4V alloy by nanoporous TiO_2 film growth and hydroxyapatite electrodeposited into nanpores is shown in Figure 9.

The fretting corrosion sliding test was performed against corundum counterbody in Fusayama - Mayer artificial saliva solution at a normal load of 800 mN, sliding frequency of 1 Hz for sliding cycles of 1000.

From Figure 9, it appears that for each separate surface the lowest coefficient of friction was recorded for hydroxyapatite electrodeposited into nanoporous TiO_2 layer surface formed on Ti-6Al-4V alloy. The coefficient of friction for untreated Ti-6Al-4V alloy exhibits strong oscillations during the sliding test compared to the anodized one and electrodeposited hydroxyapatite films. These oscillations could be attributed to the formation, accumulation and ejection of third - body particles (wear debris) [3].

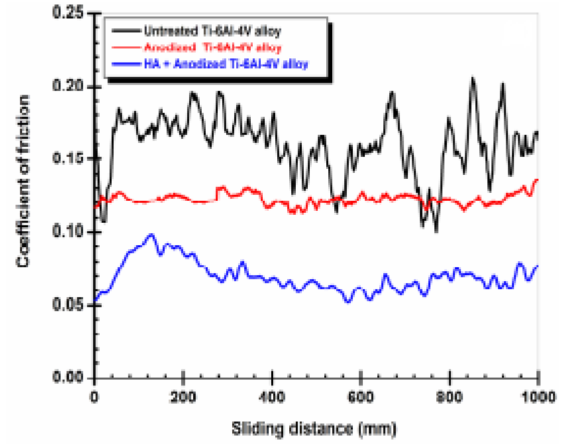
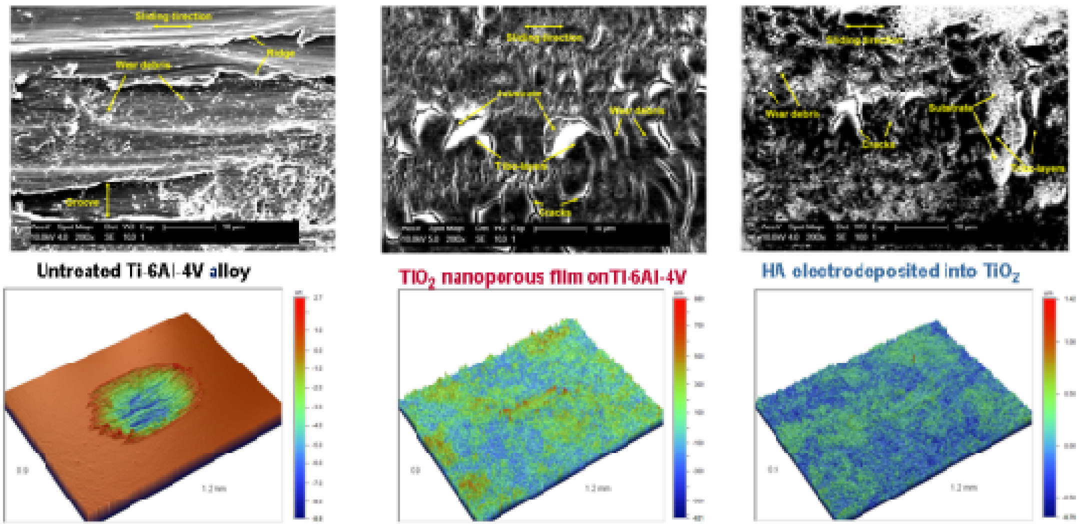


Fig. 9. *Evolution of the friction coefficients versus sliding distance for (▬) Untreated Ti-6Al-4V alloy surface; (▬) Anodic nanoporous TiO$_2$ film surface and (▬) hydroxyapatite electrodeposited into nanoporous TiO$_2$ layer, at normal forces of 800 mN, sliding frequency - 1 Hz and sliding cycles - 1000*

The corresponding scanning electron micrographs of the wear tracks and the 3D views obtained by non-contact profilometry of the wear tracks generated after reciprocating sliding tests (F_N=800 mN, displacement amplitude 500 µm, 1 Hz and 1800 sliding cycles) in Fusayama - Mayer saliva solution are shown in Figure 10 for (a) untreated Ti-6Al-4V surface, (b) nanoporous TiO$_2$ film surface and (c) HA electrodeposited layer into nanoporous TiO$_2$ film formed on titanium alloy.

The 3D morphologies of the wear tracks, the were track depth profiles and the volumetric material loss in the wear tracks was further investigated by non-contact white light profilometry.

The wear track of untreated Ti-6Al-4V sample reveals that it has suffered a major damage due to the plastic deformation and worn of wear debris provided by the fretting action of the alumina ball. For untreated Ti-6Al-4V alloy, wear debris are found spread out over the entire wear track surface [3]. The third bodies are obtained by the accumulation of wear debris particles detached because of the mechanical action between the first (metallic substrate) and second (hard inert counter piece) bodies. The third body particles which are not ejected from the contact can adhere to both first and second bodies and so contributing to the formation of a transfer film or can be fragmented in smaller particles being spread on the first body [3]. These third bodies trapped at the interface contribute to an enlarged real contact area.



(a) (b) (c)

Fig. 10. *SEM images of the wear tracks in the central region and 3D views obtained by non-contact profilometry of the wear tracks after reciprocating sliding tests (F_N=800 mN, displacement amplitude 500 µm, 1 Hz and 1800 sliding cycles) performed in artificial saliva for: (a) untreated Ti-6Al-4V alloy surface; (b) Anodic nanoporous TiO$_2$ film surface and (c) hydroxyapatite electrodeposited into nanoporous TiO$_2$ layer surface*

Since anodic nanoporous TiO_2 film surface and hydroxyapatite electrodeposited into nanoporous TiO_2 layer surface do not reveal wear debris in a significant amount, the dimension of the wear tracks corresponding to these surfaces is less than the dimensions of the untreated Ti-6Al-4V sample. As shown in Figure 10, untreated Ti-6Al-4V is characterized by a very high wear rate, which is in good agreement with the morphology of the worn surface. The irregular surface profile of the wear track in case of untreated surface points out the presence of adhered wear debris in the track indicating again the abrasive wear mechanism, observation also supported by SEM micrographs of the wear tracks developed on the same surface. The wear tracks of the anodic formed nanoporous TiO_2 film surface and hydroxyapatite electrodeposited into nanoporous TiO_2 are very shallow in depth and narrow in width compared to the untreated Ti-6Al-4V surface. Electrochemical methods applied for surface modification of Ti-6Al-4V alloy led to the lowering of wear depth and hence improved wear resistance compared to the untreated Ti-6Al-4V surface.

5. Conclusions

► Tribocorrosion process of materials degradation is present in many fields of materials, biomaterials, nanomaterials or coatings applications.

► New knowledge about tribocorrosion process and its synergy is needed in order to develop improved materials/coatings for industrial and biomedical applications.

► Accelerated tribocorrosion tests are possible for industrial problems based on mechanistic understanding, and determination of rational criteria for materials selection.

► New advanced nanocomposite coatings could be developed by electrochemical methods to improve the friction coefficients and wear-corrosion resistance in specific environments.

► Successful electrochemical methods could be applied to improve surface properties and biocompatibility of biomaterials for longer and more efficient use.

Acknowledgments

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