DRY LUBRICATION EFFICIENCY: FROM GROUND TO SPACE

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ABSTRACT

Finding dry lubricant appropriate to space applications remains of great challenge as it implies lubricate mechanism in several atmospheres successively.

To understand and *in fine* predict the influence of environment on dry lubrication, two MoS_2 based coatings (MoS_2 and MoS_2+Ti) exhibiting opposite tribological behaviours are studied under four different environments (ultrahigh and high vacuum, dry N_2 and humid air).

Thanks to an original experimental procedure coupling physicochemical and mechanical measurements and analyses, and using common instrumentations, the study notably showed and explained why:

- N_2 does mimic neither ultrahigh vacuum (UHV) nor high vacuum (HV),
- HV does not mimic UHV.
- Contamination drives lubrication efficiency and is needed, even for MoS_2 under UHV.

The results consequently give some recommendation on the characterisation tests' realisation. They also pave the way to the development of new coatings for long life space application.

1. INTRODUCTION

Feedbacks from former applications and parametric studies often give recommendations on how to use dry lubricants but, based on those elements, it is difficult to predict how they will behave and for how long lubrication can be ensured, in particular for space applications where a lubricant is expected to operate in 2, or even 3, environments successively and where very long life is required.

Up to now, no dry lubricants provides an ideal tribological behaviour (friction coefficient and wear life) in all encountered environments. The most commonly used lubricant, MoS_2, provides low friction and long life in vacuum, yet provides a high friction and short life in air [1,2]. When tested in dry N_2, the friction coefficient is equivalent to the one obtained in vacuum but the wear life is longer [2,3].

One remedy to the limitations of MoS_2 in air was the co-deposition of MoS_2 with metals (Cr, Au, Ag, Ti…) [4,5]. The aim was to ensure a better resistance to oxidization in air and to provide a better load carrying capacity while still ensuring good lubrication in vacuum. Due to its good tribological behaviour under air and N_2, MoS_2+Ti coatings appeared to be the most promising. Unfortunately, in vacuum, the inclusion of Ti in MoS_2 dramatically changed the MoS_2 behaviour giving it extremely poor lubrication properties [6]. However, for financial and time reasons, numerous tribological characterization tests are done in air or in dry N_2, the latter globally considered vacuum equivalent. But both MoS_2 and MoS_2+Ti demonstrate that it is not the case. Apart Gardos [3] and Buttery [7], no concrete warning about such a statement exist.

To understand and *in fine* predict the influence of environment on dry lubrication, two MoS_2 based coatings (MoS_2 and MoS_2+Ti) are chosen for this study because of their antagonistic lubrication behaviours. Contrary to MoS_2, which provides low friction and long wear life in both vacuum and dry nitrogen environments, MoS_2+Ti provides low friction and long wear life in both dry nitrogen and air environment. Such an antagonism is positively used to decipher the origins of their tribological behaviours in relation with the physicochemical interactions between the environment and the dry lubricated contact.

The results presented in this paper are extensively described and explained in two recently published papers [8,9]. They are here summed up.

2. EXPERIMENTAL DETAILS

2.1. Materials

2.1.1 MoS_2

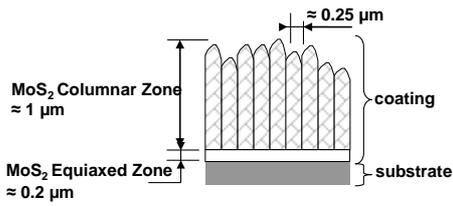The MoS_2 coating (Fig. 1) is a 1µm thick dense crystalline columnar coating. It is deposited by RF-PVD.

Figure 1. Scheme of the dense columnar MoS_2 coating.

2.1.2 MoS_2+Ti

The MoS_2+Ti coating (Fig. 2) is a 1µm thick amorphous coating. It is deposited by DC CFUBMSIP (Direct Current Closed Field UnBalanced Magnetron Sputter Ion Plating). Ti is dispersed inside the MoS_2. Thus, the structure is not multi-layered.
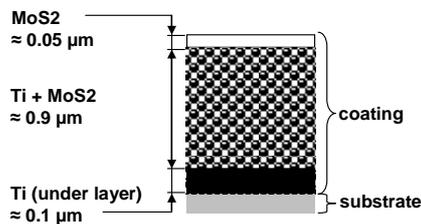

Figure 2. Scheme of the amorphous MoS_2+Ti coating.

2.2. Contact conditions

The contact used in this study is a pin-on-plate pure sliding reciprocating contact. Only the plate is coated. The pin is barrel shaped to get an ellipse contact of around 1.5mm². Schemed on Fig. 3, one cycle of sliding is defined as A+B. The contact is loaded in position 1. After n cycles of sliding, the contact is unloaded in position 2.
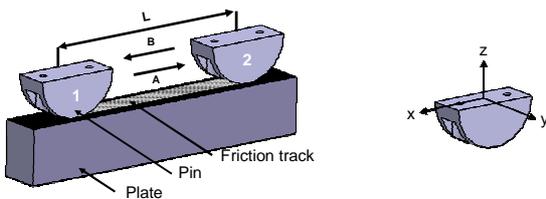

Figure 3. Contact scheme.

Kinematics	Pure sliding Reciprocating: ±20mm
Temperature	Room temperature
Environments	UHV 10^{-6} Pa
	HV 10^{-3} Pa
	Dry N_2 10^5 Pa, 0%HR
	Air 10^5 Pa, 50%HR
Max Hertz contact pressure	1 GPa
Sliding velocity	10 mm/s

Table 1. Contact conditions.

Contact conditions are summed up in Tab. 1. As the contamination time of surfaces depends on the ambient

pressure value, the two vacuum levels allow studying the impact of such a change on the tribological behaviours of the contact.
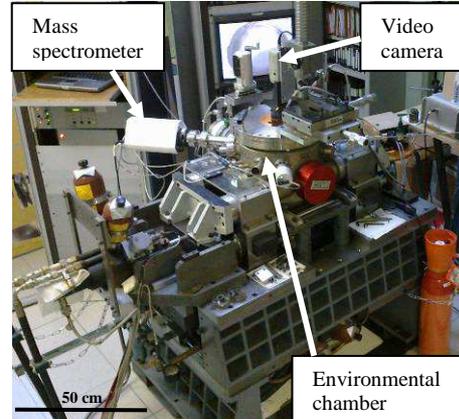
2.3. Experimental set up

Figure 4. Equipped environmental tribometer.

Experiments are conducted on a fully equipped environmental tribometer (Fig. 4). Besides the classical force measurements with a 3D piezoelectric force sensor, the tribometer is equipped with:

- a mass spectrometer allowing to control the atmosphere before, during and after experiments. The continuous measurement allows determining the species desorbed and adsorbed by the contact, and which body is the main stressed (the substrate, the coating or the 3rd body). The mass spectrometer can only be used under UHV and HV.
- a video camera to visualise one extremity (extremity 1, Fig. 3) of the friction track during the experiment. It allows studying the morphological changes occurring in the track under friction and the distribution of the 3rd body inside and outside the friction track. Dynamically, it helps determining the 3rd body flows inside the contact.

2.4. Experimental procedure

All the procedure and the analysis methodology have been clearly described in a previous paper [9].

During experiments real time mechanical and physic-chemical measurements are done thanks to tribometer's equipment. After experiments, the specimens are studied by SEM and EDX analyses in order to study the friction track (substrate, coating, and 3rd body) morphology and composition. The velocity accommodation modes (V.A.M.) are at this time determinable. Confronting real time and post-test information, the contact life can be reconstituted and

understood mechano-physico-chemically. In specific case, other physicochemical post-test analyses are conducted by ToF-SIMS (Time of Flight Secondary Ion Mass Spectrometry) which allows studying at the molecular level the extreme surface of the bodies in contact. The 3rd body concept is used in the reconstitution and description to conserve the dynamic of the mechano-physicochemical mechanisms governing the tribological behaviour. Once reconstituted, they can be understood and explained.

3. RESULTS

Fig. 5 to 7 present the main results obtained for both MoS_2 and MoS_2+Ti coatings.

3.1. Vacuum environments

The results obtained for both materials in both UHV and HV show important differences:

- The mass spectra (Fig.5) show that, whereas only desorption occurs in UHV, i.e. there is only release of gas species from the contact under tribological stress, there is adsorption (and consequently consumption) of water by the contact in HV. The consumption is observable when both a decrease of water detection at the beginning of the test and an increase of water at the end of the test. In parallel, the detection of the other species sharply increases at the beginning and sharply decreases at the end. The complementarity between the evolutions of desorption and adsorption shows that chemical reactions occurs inside the contact between the stressed bodies and the environment,

- The friction coefficient is twice lower in HV than in UHV for MoS_2 (Fig. 5) and there is no change for MoS_2+Ti (Fig 6),
- There is a lower wear (3rd body particles definitively ejected from the contact) and a lower coating consumption in HV than in UHV for MoS_2 (Fig. 5) and no change for MoS_2+Ti (Fig. 6),
- For MoS_2 (Fig. 5), the 3rd body layer created from the agglomeration of particles detached from the coating is continuous and cohesive in both UHV and HV,
- The friction track morphology changed for MoS_2+Ti. Indeed, in UHV, the resulting contact area on the pin is rectangular whereas in HV, it is rectangular in the middle with both the extremity of an ellipse at both extremities of the rectangular area. However, the 3rd body morphology is the same as in UHV,
- There is a chemical segregation in HV. Indeed, for MoS_2, the composition of the 3rd body layer is heterogeneous. There are two phases, a Mo+O+S phase similar the 3rd body layer obtained in UHV and a phase containing a high concentration of C and O. For MoS_2+Ti, Ti is only present in the rectangular area and Mo, S, and O are distributed in the ellipse extremities and in the ejected 3rd body.

Such changes are the results of the physicochemical interactions of the 3rd body and the contaminants coming from both the coating (internal contamination) and the surrounding environment (HV). Indeed, in UHV
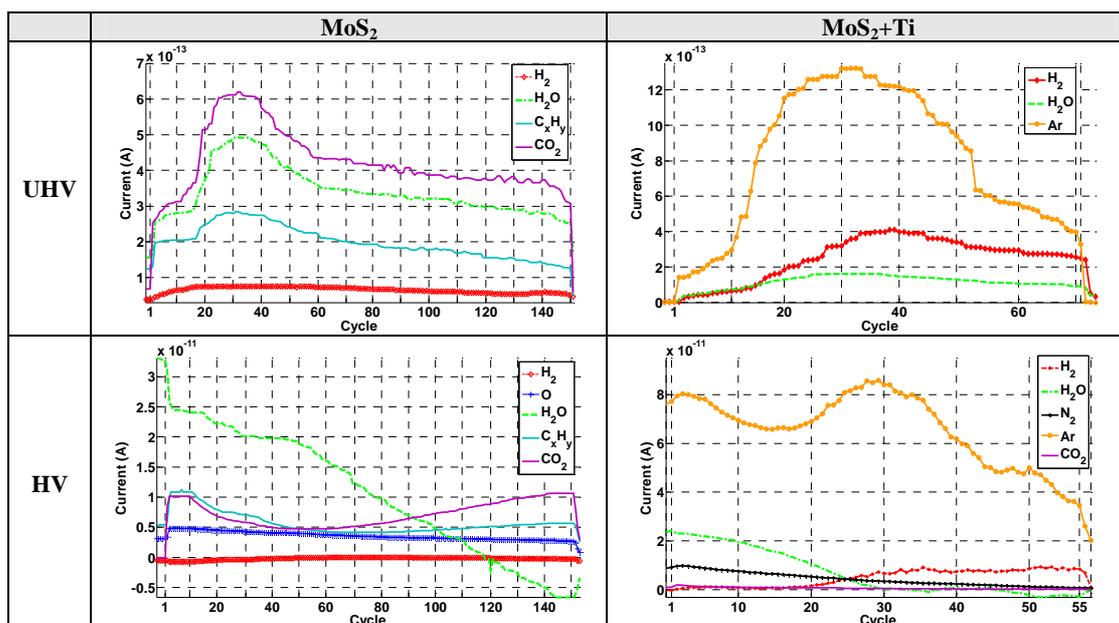

Figure 5. Mass spectra of both MoS_2 and MoS_2+Ti under UHV and HV

Friction coefficient				
Environment	**UHV**	**HV**	**Dry N₂**	**Air**
Plate	1ˢᵗ body film — 2mm	1ˢᵗ body film — 2mm	1ˢᵗ body film — 2mm	1ˢᵗ body film — 2mm
Pin	2 mm · 300 µm	2 mm · 60 µm	2 mm · 200 µm	2 mm · 100 µm
Contact Schemes	3ʳᵈ body layer [Mo+S+O] · 1ˢᵗ body film [Mo+S+O]	3ʳᵈ body layer [Mo+S+O] et [C+O rich.] · 1ˢᵗ body film [Mo+S+O]	3ʳᵈ body layer [Mo+S+N+O] · 1ˢᵗ body film [Mo+S+O]	3ʳᵈ body layer [Mo+O+S] · 1ˢᵗ body film [Mo+S+O]
V.A.M	*No rolling*	*No rolling*	*No rolling*	*No rolling*
	Shearing at the interface between 3ʳᵈ body layer and the 1ˢᵗ body film	Shearing at the interface between 3ʳᵈ body layer and the 1ˢᵗ body film *	Shearing at the interface between 3ʳᵈ body layer and the 1ˢᵗ body film	Shearing at the interface between 3ʳᵈ body layer and the 1ˢᵗ body film
	Shearing in the volume of the 3ʳᵈ body layer and 1ˢᵗ body film *	Shearing in the volume of the [Mo+S+O] 3ʳᵈ body and 1ˢᵗ body film	Shearing in the volume of the 3ʳᵈ body layer and 1ˢᵗ body film	*No shearing in the volume*

*Figure 6. MoS₂. *: the main VAM of the VAM exhibited.*

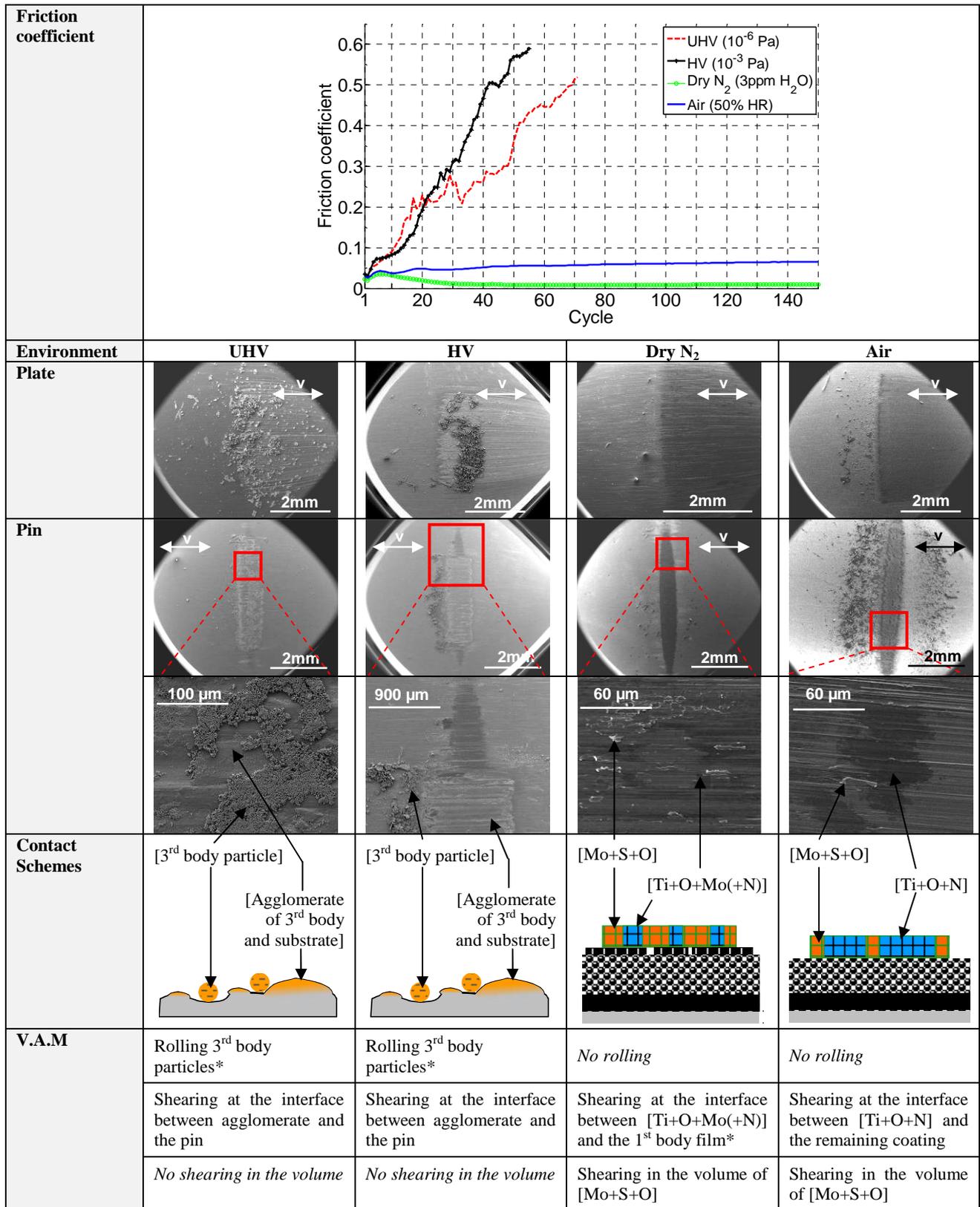|	UHV	HV	Dry N₂	Air
Friction coefficient	UHV (10^{-6} Pa) — HV (10^{-3} Pa) — Dry N_2 (3ppm H_2O) — Air (50% HR)			
Environment	UHV	HV	Dry N₂	Air
Plate	v 2mm	v 2mm	v 2mm	v 2mm
Pin	v 2mm 100 µm	v 2mm 900 µm	v 2mm 60 µm	v 2mm 60 µm
Contact Schemes	[3rd body particle] [Agglomerate of 3rd body and substrate]	[3rd body particle] [Agglomerate of 3rd body and substrate]	[Mo+S+O] [Ti+O+Mo(+N)]	[Mo+S+O] [Ti+O+N]
V.A.M	Rolling 3rd body particles*	Rolling 3rd body particles*	*No rolling*	*No rolling*
	Shearing at the interface between agglomerate and the pin	Shearing at the interface between agglomerate and the pin	Shearing at the interface between [Ti+O+Mo(+N)] and the 1st body film*	Shearing at the interface between [Ti+O+N] and the remaining coating
	No shearing in the volume	*No shearing in the volume*	Shearing in the volume of [Mo+S+O]	Shearing in the volume of [Mo+S+O]

Figure 7. MoS₂+Ti. *: the main VAM of the VAM exhibited.

the internal contaminant of MoS₂, i.e. the species desorbed under friction (Fig. 5) are reactive with the freshly created 3rd body and form a Mo+S+O phase able to deform itself to accommodate velocities. In HV, the supply of external water emphasises the chemical rearrangement by inducing a segregation of C initially

contained in the MoS_2 coating. Besides the creation of the $Mo+S+O$ phase, a phase mainly comprised of C is thus created leading to the following V.A.M.: shearing at the interface between the 3rd body layer (made of both phases) and the 1st body film and shearing in the volume of the $Mo+S+O$ phase.

As for MoS_2+Ti, the internal contaminants (Ar mainly, Fig. 5) are not reactive with the 3rd body. This fact added to the structure which does not allow the creation of 3rd body particles small enough to be trapped inside the contact, do not allow the MoS_2+Ti coating to provide low friction and long wear. To dissociate the importance of the coating microstructure from the Ti role in the generation of big particles, tests were conducted in UHV with an amorphous MoS_2 (Fig. 8). The microstructure is the main reason for short wear life. In HV, however, the external contamination improves the tribological behaviour in the sense that a smaller surface, i.e the rectangular part of the contact area on the pin, is strongly damage. A segregation phenomenon also occurs inducing the Ti to stay inside the damaged area. Unfortunately, the mecano-physico-chemical interactions inside the contact between the 3rd body and the HV environment do not suffice to improve the tribological behaviour as it is the case with MoS_2.
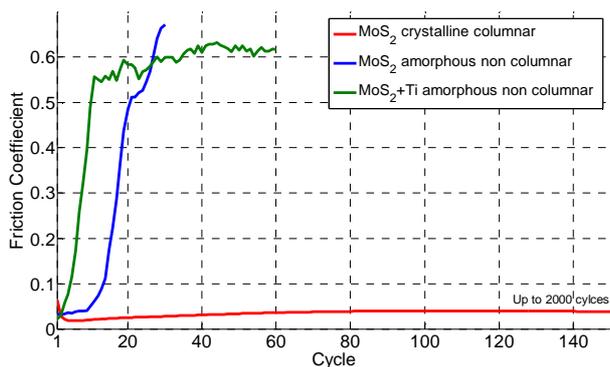

Figure 8. Friction coefficient of crystalline columnar MoS_2 vs both amorphous non columnar MoS_2 and MoS_2+Ti

3.2. Dry N_2 environment

MoS_2 tribological behaviour presents:

- A friction coefficient similar to the one obtained in UHV (Fig. 6),
- Both the wear and the coating consumption are lower (Fig. 6). The behaviour is consequently similar to the one obtained in HV,
- The 3rd body morphology appears to be the same as the one obtained in UHV (Fig 6). However, the EDX analysis shows that there is no chemical segregation as encountered in HV and no difference in O distribution inside and outside the track as encountered in UHV

(lower O content in the track than outside it).

There is consequently a certain ambiguity on whether the tribological behaviour is equivalent to the UHV or to the HV… Gardos [3] explains the good friction behaviour of MoS_2 under N_2 by the easier adsorption of N_2 than H_2O on the oxidization sites. To validate the incorporation of N_2 in the 3rd body during experiment, a friction test starting in dry N_2 and finishing in UHV has been conducted. The mass spectra obtained in UHV shows that the desorbed species from the 3rd body are (in order of importance): H_2O, CO_2, O & N_2, and H_2. This detection lasts as long as the 3rd body stays inside the contact and is stressed. Consequently N_2 has effectively been incorporated in the 3rd body while under friction in dry N_2. N_2 molecule has not been dissociated during incorporation, letting think that the adsorption is a low energy adsorption which does not lead to the formation of covalent bound with the 3rd body. It is difficult to determine whether the H_2O and O contained in the N_2 are adsorbed in the 3rd body and react or if the O detected by EDX comes from the coating internal contamination whose release is blocked by the ambient N_2 high pressure (10^5 Pa).

As almost all sites are occupied by N_2, high particle mobility would be preserved giving to the trapped 3rd body layer plastic properties similar to those obtained in UHV. The quantity of O contamination inside the coating does not disadvantage plastic properties of the 3rd body layer. The V.A.M.s taking place are thus equivalent to those obtained in UHV with a higher participation of the shearing mode at the interface between the 3rd body layer and the first body film. A N_2 rich layer might be formed at the 1st body film extreme surface as it is the case with hydrogen and DLC in dry atmosphere [10]. The origin of low friction and long life in dry N_2 are consequently totally different than in both UHV and HV, even if the friction coefficient is similar to the one obtained in UHV.

MoS_2+Ti tribological behaviour presents:

- A very low friction (Fig. 7).
- A low wear (Fig. 7). The video shows that the wear (ejection of 3rd body particles) only happens between cycles 3 and 10, i.e. as soon as the consumption of Ti containing part of the coating starts and continue until a stable 3rd body layer is formed. After, cycle 10, there is no significant modification of the track.
- The 3rd body is heterogeneously distributed in the track on both the plate and the pin (Fig. 7). On the plate, the 3rd body layer is cohesive and has the ability to plastically flow inside the contact. Chemically, the detection of both N and O is more important in the track and the 3rd body layer appears to be comprised of Mo,

S and O. On the pin, there is a chemical segregation of species. Indeed, where no 3rd body layer similar to the one observed on the plate is detected, plates made of Ti, O, Mo with a small presence of N are detected (Fig 7). Those plates do not show particular abilities to flow plastically but allows an easy shearing at their surface, surely in surface complexes. The stable 3rd body layer is consequently heterogeneous chemically and mechanically.

Friction tests starting under N_2 and finishing under ultrahigh vacuum have also been conducted. The mass spectrum shows desorption of (in order of importance): H_2, Ar, H_2O, O, N, and not N_2, during the consumption of the 3rd body created under dry N_2. As all MoS_2+Ti samples only present desorption of Ar in UHV, a small amount of N_2 has chemically reacted with the 3rd body, as well as H_2O and O, all coming from the dry N_2 environment. Indeed, there 3 ppm of H_2O and 3 ppm of O, i.e. 3.10^{-1} Pa of both of them inside the "dry" environment. Consequently and similarly to the MoS_2 in HV, the V.A.M. leading to low friction and long life are both shearing in the volume of the Mo+S+O 3rd body layer and shearing at the interface between the total 3rd body layer and the 1st body film formed on the coating surface. The second V.A.M. appears to be the predominant mechanism.

N_2 dissociation into N and the presence of O in the track resulting from chemical reaction with H_2O and O show that dry N_2 environment is not neutral.

3.3. Air environment

MoS_2 tribological behaviour presents:

- High friction (Fig. 6),
- High wear (Fig. 6). The video shows that wear starts at cycle 2 and stops at cycle 70, i.e. when the substrate is visible in the track.
- Morphologically, a thin film is formed on the plate sample and is heterogeneously distributed in the track. This film is comparable to the 1st body film formed under friction at the coating top surface in vacuum and N_2 environments. On the pin, the 3rd body layer is brittle (Fig. 6). Both it and the ejected particles are comprised of Mo, S with a high proportion of O. O strongly links the small particles coming from the columns together leading to the formation of a highly cohesive and brittle 3rd body layer.
- Ejected particles are much bigger and thicker than those obtained in vacuum or N_2.

The V.A.M. is quasi-exclusively shearing at the interface between the 3rd body layer and the 1st body film formed in the track, right above the substrate.

MoS_2+Ti tribological behaviour presents:

- Low friction which is a bit higher than in N_2 (Fig7). During the first 10 cycles, friction evolution is the same as in N_2 but then undergoes several variation until reaching a steady state approximately at cycle 50
- Low wear which is nonetheless higher than in N_2. The wear is very similar to the wear observed in N_2 during the 10 first cycles. However, the video shows that between cycles 10 and 45, the ejected 3rd body volume alternatively decreases and increases. During those cycles, a selection on the 3rd body occurs: big particles and/or brittle agglomerates of small particles are ejected while small particles and more ductile agglomerates stay inside the contact.
- On the pin, the 3rd body layer inside the contact is heterogeneously distributed and a chemical segregation much more accentuated than in dry N_2 appears (Fig. 4). This leads to the creation of plates comprised of Ti, O, and N. Around them is formed a Mo+S+O 3rd body layer whose morphology and rheology is similar to the Mo+S+O 3rd body layer obtained in UHV with MoS_2. Ejected 3rd body particles are only comprised of Mo, S and, O and have the same morphology as the particles ejected during MoS_2 testing in air. Ti entirely stays inside the contact ellipse. No films are created on the plate, contrary to what was observed in N_2.

The 3rd body layer is not only comprised of Mo, S, Ti, but is heterogeneous mechanically and chemically. The V.A.M. is mainly shearing at the interface between the total 3rd body layer and the coating, followed by shearing in the volume of the Mo+S+O 3rd body layer.

4. CONCLUSION

Considering all experiments, those which provide low friction and long life always present:

- A 3rd body layer in which chemical rearrangement occurred between the 3rd body freshly created (comprised of the coating base material) either with internal contaminant (case of MoS_2 under UHV), or with external contaminants (case of MoS_2 and N_2 and MoS_2+Ti in N_2 and Air), or both (case of MoS_2 in HV). Ti concentrates the reactivity and thus protects the MoS_2 from excessive reaction with water as it happens in air with MoS_2.
- V.A.M. coupling shearing in the volume of 3rd

body layer and shearing at the interface between the 3rd body layer and the coating top surface either plasticised (forming the 1st body film) or not.

Experiments with MoS₂+Ti in UHV and HV and MoS₂ in air 50%HR demonstrate the requirement of a critical amount of contaminant to obtain the 3rd body able to provide low friction and long life. Below it, i.e. in the case of M$_O$S₂+Ti in UHV and HV, it does not work because the contact cannot adapt itself enough to the environment. Above it, i.e. in the case MoS₂ in air 50%HR, it does not work due to the high cohesion of the 3rd body giving it brittle properties.

The columnar structure of MoS₂ allows creating small 3rd body particles contrary to the amorphous non columnar. The slow consumption is possible in the latter case thanks to the external contamination which induces chemical segregation and detachment of small coating parts.

Consequently, contamination can be beneficial and drives the lubrication efficiency of dry lubricants. It notably modulates the 3rd body rheology and helps controlling the 3rd body generation (particle size), i.e. the coating consumption, although this is the coating microstructure that mainly controls that generation.

5. APPLICATIVE CONSEQUENCES

Direct consequences for industrial applications are:

- N₂ does mimic UHV nor HV,
- HV does not mimic UHV because it induces chemical reactions with the residual water.

Recommendations for tribological characterizations are:

- To stop considering dry N₂ and UHV equivalent. It is of no consequence for MoS₂ for ground test on flight model but could be of great importance for qualification life test (longer life under N₂) or for other materials. Characterisation tests must be conducted in UHV too,
- As UHV is difficultly reachable for full mechanisms, conducting characterisation tests in HV too is recommended if the mechanism is intended to be tested in HV once assembled,
- To use a mass spectrometer to control desorption/adsorption occurring all along a tribological test and to control the compositional evolution of the 3rd body after test, by EDX analysis for example. The experimental procedure and the related analysis method developed here have been developed to be usable in an industrial context in order to

explain the mechanical and physic-chemical phenomena controlling the tribological behaviour of dry lubricated contact (in other words friction coefficient is not sufficient!).

The study also brings qualitative information on the optimal coating for space application and notably that it:

- Should localizes 3rd body generation at its surface and be "pre-cut" to generate particle of the right size to be trapped inside the contact. It should also not be totally amorphous.
- Should contained reactive chosen *contaminants*, either solid or gaseous, whose reaction with the freshly created 3rd body and/or the surrounding environment is activated tribologically.

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