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Original paper

The procedure of evaluating the practical adhesion strength of new biocompatible nano- and micro-thin films in accordance with international standards

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The possibilities of using newly developed nano- and micro-thin films in biomedicine are intensively studied at the present time. Many research institutions are looking for ways to evaluate mechanical properties of these films. One of the most important and frequently studied characteristics is practical adhesion. A very important method for evaluating the practical adhesion strength is scratch test. Often, however, the research teams use a method based on the disunity evaluation of adhesion of biocompatible surface layer. This makes the quantitative comparison of research results impossible. We designed and tested new evaluation method and procedure based on international standards in order to eliminate these problems. This article is aimed at showing the new possibility of using established standards for evaluating adhesion of nano- and micro-thin biocompatible films and at showing the application of the standards to evaluate the often studied DLC biocompatible layers. The thickness of the film was 470 nm. As a substrate a titanium alloy Ti6Al4V was used.

Key words: *practical adhesion, strength, nano-layer, micro-layer, international standards, scratch test*

1. Introduction

Nano- and micro-thin layers are widely used in many branches of industry, including medicine. One of the most important mechanical properties of such a layer is its practical adhesion [1]–[3]. Practical adhesion is very important in each application because after detachment of thin layers from basic material it does not serve its purpose and the remains of the layer can cause additional damage. In the case of implants, the remains can cause additional damage to surrounding tissue in living organism. There is a number of techniques to measure the adhesion strength of biocompatible film/substrate interfaces. However, a practical adhesion studies are rooted in empirical data analysis. The primary difference between practical and basic adhesion lies in the fact that the sample geometry and testing method can change the value of practical adhesion strength, where basic adhesion strength is a fundamental quantity affecting two materials. The practical adhesion strength is useful as a design parameter, and the basic adhesion strength broadens the scientific understanding of why materials stick together.

The practical adhesion strength, i.e., the work done to separate the thin film from the substrate (or one film from another film), is very sensitive to the test methods and the mechanical effects, such as the strain rate, residual stress, thickness and mechanical properties of the layers. Deriving intrinsic adhesion properties of the interfaces, which are independent of such parameters, from the practical adhesion strength measurements is an important task.

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The practical adhesion strength is an important parameter for every coating and we can use more methods for its determination, and one of them used most frequently is scratch test. Nowadays, micro- and nano-scratch tests supplement the classic (macroscopic) scratch test, which is well described in literature. In current research on nano- and micro-biocompatible films, the problem lies in the fact that although the measurement of practical adhesion is highly influenced by the conditions of measurement, institutes conducting research do not apply widely used uniform measuring conditions and procedures.

Currently there are already in use internationally recognized industrial standards for evaluating adhesion of macro- and micro-layers. The scratch test procedures are described in the national/international standards, which are usually enforced also in the European, American, Japanese or other national standards [1]–[4]. The standards are designed for a wide range of different coating materials. The most important standards for quantitative single point scratch testing are as follows:

International:
- ISO 1071-3: Test method for adhesion and other mechanical failure modes of thin advanced technical ceramics.
- ISO 20502: Test method for adhesion of fine ceramics (advanced ceramics, advanced technical ceramics).
- ISO N269 (Working Draft): Test method for adhesion of fine ceramics coatings.
- ISO 1518: Test method for adhesion of paints and varnishes.

USA:
- ASTM C1624: Test method for adhesion strength and mechanical failure modes of ceramic coatings.
- ASTM D7027-05: Test method for evaluation of scratch resistance of polymeric coatings and plastics.
- ASTM D7187: Test method for measuring mechanistic aspects of scratch/mar behaviour of paint coatings by nanoscratching.

Japan:
- JIS R 3255 (1997): Test method for adhesion of thin films on glass substrate.

The standards applicable to testing thin films in biomedical engineering are: ISO 1071-3, ISO 20502, ASTM C1624-05 and because of the possibility of testing nano-films also the ASTM D 7187-05. At the required thickness and load the measuring conditions recommended by standard ISO 20502 and standard ISO 1071-3 are the same. Table 1 presents the most important conditions for the evaluation according to these standards. The standards are designed mainly for macro-scratch testers, as these are commonly used to measure the nano- and micro-layers.

Table 1. Measuring conditions in accordance with appropriate international standards designed for nano- and micro-films testing

	ISO 1071-3	C 1624-05	D7187
Film material	ceramic coating and other	ceramic coating	paint coating
Film thickness	to 20 μm	from 0.10 μm to 30 μm	to 500 nm
Preload	1 N	1 N for $L_{max} < 10$ N; 5 N for $L_{max} > 20$ N;	0.1 to 1 mN
Progressive loading scratch test mode – loading rate	100 N/min	10 N/min for $L_{max} < 20$ N; 100 N/min for $L_{max} > 20$ N; minimum 5 N/min	5 to 200 mN/min
Progressive loading scratch test mode – displacement speed	10±0.1 mm/min	10±0.1 mm/min	0.5 to 10 mm/min
Constant load scratch test mode – loading rate	20% of L_{max}	20%, 40%, 60%, 100% of L_{max}	–
Constant load scratch test mode – displacement speed	10 mm/min	10±0.1 mm/min	–
Multi-pass scratch test mode – loading rate	50% of L_{max}	–	–
Multi-pass scratch test mode – displacement speed	–	–	–
Stylus geometry	Rockwell C	Rockwell C	spherical, 1 to 100 μm
Surface roughness	$Ra < 0.5$ μm	$Rq < 1$ μm	–
Film hardness	–	$HV > 5$ GPa	–
Min. number of test operations	5×	5×	3×
Optical microscope	100 to 500×	100 to 500×	–
Temperature	22±2 °C	20±5 °C	23±2 °C
Relative humidity	50±10%	50±10%	50±5%

All scratch tests (table 1) consist in pulling a diamond stylus over the surface of a sample under a normal force. The recommended shape of stylus is spherical Rockwell C or a smaller one. Rockwell C is a conical diamond intender with an included angle of 120° and with its spherical tip of 200 μm [3]. It is generally accepted that three–five test operations are suitable for coatings of the thickness ranging from 0.1 to 20 μm. The maximum load (L_{max}) should be selected to produce the desired maximum level of coating damage, but without markedly exceeding that load, which can cause excessive stylus wear [3].

For the scratch tests, three modes of measurement were proposed: progressive loading scratch test mode (PLST), constant load scratch test mode (CLST) and multi-pass scratch test mode (MPST). The CLST and MPST are the modes defined only for some standards. The progressive loading scratch test mode is defined for all the standards mentioned. The load on the indenter increases linearly as the indenter moves across the test surface at a constant speed and failure is observed. The normal load at which this happens is called the *critical normal load*. PERRY [6], [7], STEINMANN and HINTERMANN [8], VALLI [9] suggest a use of the progressive load scratch test for coating adhesion measurements. Today, the method is widely used by the coating industry and development laboratories. Its usefulness as an adhesion and quality assessment method has been discussed [5], [10]–[14]. The scratch test is generally accepted as a good and an efficient method for the quality assessment of coated surfaces, but its use for coating-to-substrate adhesion assessment has been criticised by several authors [14], [15].

The type of the failure which is often observed in the scratch test depends critically on the properties of substrate and coating. The material response to loading conditions has been divided into three independent phases by HOLMBERG [16]. The first phase represents the ploughing of a stylus in the substrate material. The substrate material is deformed by plastic or elastic deformation and a groove is formed. The second phase represents the bending and drawing of a free-standing coating. The bending movements cause stresses and stress release in the coating when drawn between the surfaces. In this phase, the work done for overcoming friction is considered. The third phase represents pulling and spalling the coating from one point on the surface when its other part is fixed. The increasing pulling force results in cracks at the place of maximum tensile stress. The formation of cracks in the groove of a scratch tester has been shown by e.g. in [5], [14], [16]. They can typically be described as angular cracks, parallel cracks, transverse semi-circular cracks, coating chipping, coating spalling and coating breakthrough.

Although the mentioned standards are designed and subjected to professional criticism, unfortunately they are not usually and widely used for testing and developing new thin layers, even if they are suitable for measuring emerging nano- and micro-films in medical applications.

Most of research findings are not comparable because authors do not describe the main measuring conditions or they apply nonstandard conditions (table 2, [18]–[23]). Only a few papers give limited information about the measuring conditions.

Table 2. Measuring conditions used to study DLC films in research works

Author	Substrate	Film thickness	Stylus geometry	Displacement speed	Loading rate	Critical force
NAKAO et al. [18]	Si	161 nm	spherical $R = 5$ μm	–	progressive load mode 31.93 mN/mm	26.2 mN
FUNADA et al. [19]	glass	0.5 μm	Spherical $R = 0.2$ mm	10 mm/min	constant load scratch test mode	45 mN
HORIUCHI et al. [20]	steel	0.9 μm	–	10 mm/min	progressive load mode 100 N/min	20 N
Center for Tribology Inc. – Application note [21]	NiP	6 nm	spherical $R = 0.4$ mm	10 mm/s	progressive load mode	0.9 N
ZANG, HUAN [22]	40 CrNiMo	50 nm	Berkovich	10 μm/s	progressive load mode	114 mN
VERCAMMEN et al. [23]	steel	1.39 μm	–	–	–	28 N

The second problem lies in a lack of specific information about the critical force measured [19]–[22]. The normal force at which failure occurs is called the *critical normal force* L_c. In a scratch, a number of consecutive coating-failure events may be observed at increasing critical normal force values. Failure by cracking through the coating usually occurs at lower normal force than the detachment of coating. In general, a series of failure modes can be observed and used to study the mechanical behaviour of the coated surface, where the onset of the *n*-th failure mode defines the critical normal force L_{cn}. Several different methods are in use for evaluating scratches and for the determination of critical normal forces, but only the microscope examination of the scratch is able to reliably differentiate between different failure modes and enable L_{cn} values to be attributed to specific modes of failure [1]–[4]. To assist the users of the scratch test in the standardized reporting of scratch test results, an atlas of scratch test failure modes is given in the standards mentioned [1]–[4].

2. Material and method

We compared the standardized and recommended measuring conditions and found out the overlapping measuring conditions and procedures (table 1). All three standards recommend a film thickness from 0.10 μm to 0.50 μm, and two of them from 0.10 μm to30 μm. Micro- and nano-thin films should be measured by standard method if their thickness ranges from 0.10 μm to 0.50 μm. The roughness or waviness of the surface should be $Ra < 0.5$ μm because this range is mentioned in two standards. A surface roughness value Ra of 0.50 μm may lead to oscillations of 0.1 N [1]. Film hardness should be $HV > 5$ GPa because this range is mentioned in the C1624-05 [3]. It is necessary to guarantee these conditions in the case of specimens for laboratory testing or samples intended for application in clinical practice.

The preload should be 1 N if $L_{\max} < 10$ N for measurement according to two standards and from 0.1 to 1 mN for measurement according to D7187 standard. We recommend and used the preload of 1 N because most macro- and even micro-scratch testers can work in this range. The loading rate of the progressive loading scratch test mode is 100 N/min for two standards, and in the third standard the rate to 200 mN/min is recommended. The loading rate of 100 N/min should be used because most macro- and micro-scratch testers can work in this range. The loading rate of 10 N/min could be used if $L_{\max} < 20$ N. The displacement speed of the progressive loading scratch test mode is 10 ± 0.1 mm/min for all three standards, though the D7187 recommends the speed from 0.5 to 10 mm/min. The displacement speed of 10 ± 0.1 mm/min is used because all the standards recommend the same speed. The two standards define stylus geometry shape of the Rockwell C. The stylus geometry of the Rockwell C is a spherical tip of 200 μm. The D7187 defines spherical tip of 100 μm. The Rockwell C is recommended and used because in most macro- and micro-scratch testers this spherical tip is applied. Two standards as a minimum number of test operations recommend five operations and the D7187 recommends three ones. It is generally accepted that the three tests are suitable for coatings. All three standards recommend the temperature ranging from 20 to 24 °C and a relative humidity of 50 ± 10%. The magnification for optical observation is between 100:1 and 500:1.

The loading rate of the constant load scratch test mode is 20% of $L_{\max}$ (can be measured by progressive loading scratch test mode) for two standards, though the C1624-05 recommends also 40%, 60%, 100% of $L_{\max}$. The D7187 does not define the constant load scratch test mode. The two standards define 10 mm/min displacement speed of constant load scratch test mode. Only the ISO 1071-3 defines the multi-pass scratch test mode. The loading rate is 50% of $L_{\max}$ and the displacement speed is 10 mm/min. Hence, we can only recommend the constant load scratch test mode (loading rate of 20% of $L_{\max}$ and displacement speed of 10 mm/min) because only the ISO 1071-3 defines conditions for the multi-pass scratch test mode. Based on the above, we designed and recommended standard measuring conditions (table 3).

Table 3. Designed and recommended standard measuring conditions

Film material	ceramic coating and other
Film thickness	from 0.10 μm to 20 μm
Preload	1 N for $L_{\max} < 10$ N; 5 N for $L_{\max} > 20$ N
Loading rate PLST	10 N/min for $L_{\max} < 20$ N; 100 N/min for $L_{\max} > 20$ N
Displacement speed PLST	10±0.1 mm/min
Stylus geometry	Rockwell C
Surface roughness	$Ra < 0.5$ μm
Film hardness	$HV > 5$ GPa
Min. number of test operations	5×
Optical microscope	100 to 500×
Temperature	20 to 24 °C
Relative humidity	50±10%

Almost all academic works do not specify the identification of critical load and therefore it is necessary to follow the standards to ensure the relevance and comparability of measuring results. All three standards describe the way to identify more than one critical normal force, but academic works mention only one (table 2). We define three critical forces according to the standards for progressive loading scratch test mode. The first critical normal force L_{c1} is associated with the onset of tensile cracks, indicating cohesive failure in the coating [1]–[3]. The second force L_{c2} is associated with the onset of chipping failure or local interfacial spallation, indicating adhesive failure between the coating and the substrate. The third force L_{c3} is associated with continuous perforation of coating. It is normal to ignore the isolated failures, and critical load values generally refer to the normal load on the stylus at the beginning of cluster event. The evaluation of the described scratch test requires microscope to display the scratch and to identify the important points of scratch. In our case, the optical microscope is the part of the scratch tester [3]:

$$L_{cn}\,[\mathrm{N}] = \frac{100\,[\mathrm{N/min}]\cdot l_n\,[\mathrm{mm}]}{10\,[\mathrm{mm/min}]} + 5\,[\mathrm{N}]. \qquad (1)$$

In many measuring scratch testers, measuring conditions are freely adjustable. The problem lies in the measurement of small-size sample. The size of the sample of nano- and micro-thin films is usually few millimetres. Therefore, it is necessary to choose a faster increase of normal force of 100 N/min for small samples although the critical load $L_{\max}$ is under 20 N. The same problem is with the minimum number of test operations j because the size of sample is very small. Nevertheless, the test should be carried out at least three times (j = 3) for one coating. The average critical scratch load L_{cn} is the following:

$$L_{cn}[\mathrm{N}] = \frac{\sum_{i=1}^{j} L_{cnj}[\mathrm{N}]}{j[-]}. \qquad (2)$$

We tested the diamond like carbon (DLC) coatings under the measuring conditions described. Being a biocompatible material, diamond like carbon coatings are used in many branches of medicine for various implants. The most critical issue relating to the commercialization of DLC films concerns the improvement of the adhesion strength of the coating with the substrate [18]–[21].

3. Results

We verified the designed measuring conditions and procedures by using diamond like carbon film. Layer was chosen as an example of commonly used and developed layers for biomedical applications. The thickness of the film was 470 nm. Titanium alloy Ti6Al4V was used as a substrate. The adhesion of thin films prepared by PLD technique was evaluated by the REVETEST system (CSM co.). REVETEST CSM is a standard scratch tester. We used progressive loading scratch test mode. The conditions for film testing were: preload, 5 N; loading rate, 100 N/min; displacement speed, 10 ± 0.1 mm/min. The environmental conditions were: 20 °C temperature and 55% relative humidity. We obtained the normal force, the depth of penetration and acoustic emission by REVETEST CSM tester after every scratch test. The system is equipped with optical microscope with the possibility of making photographs. The REVETEST provided us with the high-resolution graphic information (magnification higher than 100:1). We used acoustic emission to estimate failure mode points and we visually confirmed these points by the photographs of the scratch.

Table 4. Identified critical normal forces

Number of test operations	L_{c1} (N)	L_{c2} (N)	L_{c3} (N)
1	7.8	10.5	12.2
2	7.5	9.0	14.6
3	7.7	9.6	13.1
average values	7.7	9.7	13.3

Three scratches under the same conditions were made on the sample. It was not possible to make more than three scratches because of sample size (sample diameter was 9 mm) and the whole surface of the substrate uncovered with a thin layer. We identified three types of critical normal forces in each scratch under optical microscope. We checked the penetration force, where we got through the layer to the basic substrate. There was no problem with the evaluation of DLC layer by microscopic examination and we detected acoustic emission in the three points of different kinds of film and respective critical forces (table 4 and figure 1). The macroscopic method for DLC layers gave us good results (figures 2–4). The acoustic emission method gave us only approximate results (figure 1). We could only see the changes in acoustic emission on the graph. We were not able to detect the types of critical forces, but only the dependence of a change in the slope of acoustic emission on a normal displacement. Thus, we used primarily the macroscopic method as the standards recommend.

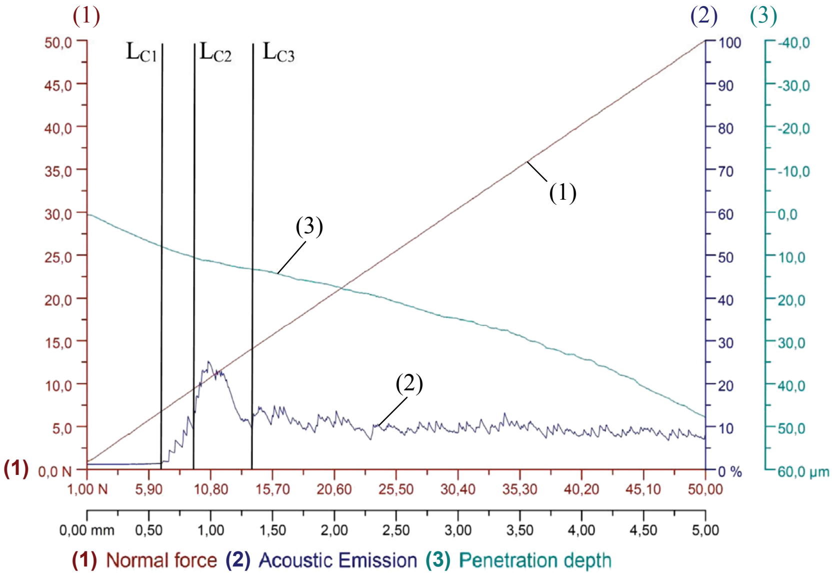

Fig. 1. Acoustic emission and penetration depth recording versus normal force in progressive load test

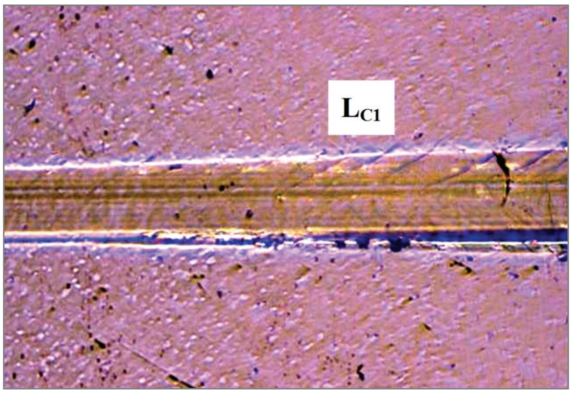

Fig. 2. Forward chevron tensile cracks at the borders of the scratch track

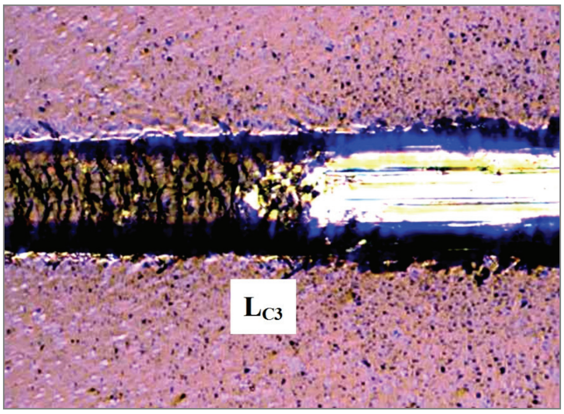

Fig. 4. Continuous perforation of coating

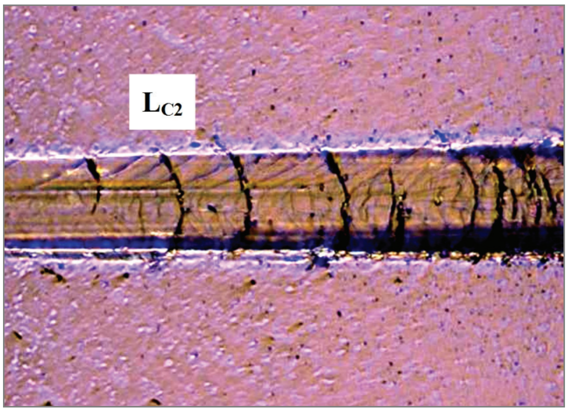

Fig. 3. Conformal-type buckling cracks with local interfacial spallation

The delamination or loss of adhesion was observed in the method and the procedure described (figures 2–4). It is normal to ignore the isolated failures, and critical load values generally refer to the normal load on the stylus at the start of cluster event. We determined the three average critical scratch loads by three test operations (table 4). The first average critical normal force L_{c1} equal to 7.7 N is associated with the start of forward chevron tensile cracks at the borders of the scratch track. Force L_{c1} indicates cohesive failure in the coating. The second average force L_{c2} equal to 9.7 N is associated with conformal-type buckling cracks and local interfacial spallation. The L_{c2} testifies to the adhesive failure between the coating and the substrate. The third average force L_{c3} (13.3 N) is associated with a continuous perforation of coating. All the critical forces represent practical adhesion but the L_{c2} or L_{c3} are primarily considered as a practical adhesion. The L_{c2} is considered as a practical adhesion in most scientific articles but some scientists mention only the L_{c1} [18], [20]–[22] because it is identified by acoustic emission as the first signal of change. Some articles mention only the L_{c3} [19] because it is associated with a continuous perforation, but sometimes it is difficult to identify this force by acoustic emission because the delamination is not observed in a graph. The behaviour of the layer is not connected only with its adhesion to basic substrate, but also with the characteristics of the layer itself. Hence, the adhesion could not be found based on the stylus behavior only, i.e., on acoustic emission. For obvious reason, evaluating all the three critical forces is vitally important in the description of the practical adhesion of thin film.

Our layers tested had a similar adhesion (table 4), measured under similar conditions [20], [23] (table 2). We cannot say whether our layers are better or not because the above mentioned research papers do not describe details or they do not follow the measuring conditions of international standards. We demonstrated and proved the application of some selected and modified measuring conditions of international standards to the measurements of practical adhesion of biocompatible nano- and micro-thin films.

4. Discussion

The application of the international standards to the measurements of practical adhesion of biocompatible nano- and micro-thin films by scratch testers offers great advantages in research area because it is a very simple way of measuring and evaluating the properties of thin layers. Unfortunately, standards are used very rarely by research teams. We designed and verified the standardized conditions and procedures for measuring practical adhesion of film whose thickness ranges from 0.10 μm to 20 μm. This range of thickness is typical of biocompatible nano- and micro-thin films. Thinner films have generally a tendency to produce higher critical scratch loads, because there are lesser residual stresses in thinner films [3], [24], [25], [28]. It is also important to follow the recommended surface roughness because higher substrate roughness decreases the critical scratch loads [3], [24].

We strongly recommend the use of scratch testers with a spherical diamond tip radius of 200 μm (Rockwell C) because the behaviour of the layer is not connected only with its adhesion to a basic substrate, but also with the probe sharpness. An increase in a tip radius increases the critical loads because larger loading area causes the lower stress applied [3], [24]–[27]. Tip materials with the lower coefficient of friction decreased the critical scratch loads [3], [24]. Diamond has the lowest coefficient of friction, compared to tungsten carbide and chromium steel [3]. These are no nano- and micro-scratch test international standards for other types of tips and thus the other tips cannot be recommended. Some systems are not equipped with the diamond Rockwell C, for example, the Hysitron TI950 has the Berkovich tip. For the measurement according to the international standards it is necessary to modify the system or to make additional comparative measurements by the Rockwell C and the other type of tip and to assess the impact of another type of tip on the results of the practical adhesion. However, the measurements do not comply with current standards.

If you do not respect the recommended and proposed basic measuring PLST conditions, the data measured will be probably irrelevant to international comparison. We recommend measuring conditions for progressive loading scratch test mode or eventually constant load scratch test mode. Loading rate should be 10 N/min for L_{max} < 20 N or 100 N/min for L_{max} > 20 N and the displacement speed should reach 10 mm/min. An increase in the load rate or a decrease in the displacement (sliding) speed increases the critical scratch load [3], [24], [25].

The above knowledge about the behaviour of a thin film and the changes in the scratch test conditions can lead to new expert systems. An expert system could be software that uses a knowledge base of expertise for problem solving. The expert system could evaluate the practical adhesion by artificial intelligence (AI), [29], [30]. The application of fuzzy logic (designed in MATLAB Fuzzy Toolbox) in estimating the practical adhesion can be an example. Inputs to the system are: coating thickness, critical load, displacement rate and loading rate. Output could be the practical adhesion described, for example, by three membership functions – poor, fair, excellent. Of course, there are other methods for the final assessment of practical adhesion, allowing us to achieve at least some relevant comparison between the adhesions measured in different laboratories and by different scratch systems.

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