Journal of Optoelectronics and Advanced Materials Vol. 2, No. 4, December 2000, p. 317 - 325

THIN FILMS INDENTATION SIZE EFFECTS IN MICROHARDNESS MEASUREMENTS

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Microhardness measurements of various thin films (e.g., Cu, Ta, Al, Ge), deposited by evaporation and magnetron sputtering techniques or Diamond-Like Carbon (DLC) films produced by plasma enhanced chemical vacuum deposition (PECVD), were carried out. Microhardness measuring equipment consists of a standard tester with flat field optics and a diamond stylus probe. Vickers-indentation measurements were performed with constant loads of: 4, 5, 10, 20, 30, 50, 100, and 200 g, in a continuous pressure. After indentation, the obtained stamps were examined by means of optical and scanning electron microscope techniques. It was found that the microhardness values of the above mentioned thin films, strongly dependent on the methodological principles of the measuring technique. Generally, there is a strong relation between the Indentation Size Effects (ISE), following an indentation, and the obtained microhardness, due to unsuitable mechanical load. As a result, various defects, e.g. stratifications, cracks, flaws, etc. are generated causing erroneous microhardness values. In this paper, a novel method for microhardness measurement of thin films is suggested. This method is based on an iteration approach, used for choosing of the correct load value, in order to avoid ISE. These measurements are of a non-destructive type, and the obtained microhardness values are not influenced by the mechanical load and the thickness of the film, but only by thin film properties. According to this method, every indentation must be accompanied with a structural control of the stamp quality.

(Received September 15, 2000; accepted November 20, 2000)

Keywords: Microhardness, Size effects, Thin metallic films, Diamond like carbon

1. Introduction

In previous papers, microhardness characteristics of polycrystalline materials, e.g. steels, alloys, metalloceramics etc., were intensively studied. The microhardness equipment and measurement methods for these materials are standardized, and numerous data was obtained. However, in microhardness measurements of thin solid films, several problems are not yet resolved. In these measurements, the microhardness becomes a dominant parameter, influenced by various factors such as: film density, film strength, film adhesion, surface morphology, bonding length, and microcrystalline orientation. Therefore, major considerations should be taken in choosing the suitable microhardness measuring technique. This technique should become reproducible, in absolute physical values, as in other measuring methods. Microhardness measurements of thin films, as well as microhardness instrumentation, have so far, not been standardized. The leading approaches in microhardness measurements of thin films such as *empirical approach* [1-13], the *cavity model of the indentation hardness* [14] and the *model of composition hardness* [14,15] were analyzed and evaluated for the preparation of this paper.

Various attempts have been made to create new original measuring systems [1, 5], in addition to the conventional and methods, such as the: Knoop, Vickers, and Rockwell indentation. The range

of the applied loads in the empirical measurements was found to be extremely wide: from 3.5 mg for Vickers indentation of Al-Zr-O [5] films, up to 150 kg for Rockwell indentation of diamond coatings [13].

Much interest was dedicated to the development of various theories or conceptions, which may be used as the basis for thin film's microhardness determination [13 - 17]. For instance, a cavity model of indentation hardness of a coated substrate, based on an analytical solution for the stress and strain fields around a pressurized coated spherical cavity, as well as, the elastic and plastic deformation, were studied [14, 16]. Another conceptually different method for the determination of hardness from residual stress and strain parameters, by using the sub-band-gap optical absorption of polycrystalline thin films was suggested in [18]. The main deficiency of this approach is that the evaluation of mechanical properties and microhardness, is based only on mathematical correlation with broadening of the absorption band tail. This indirect measuring technique is not suitable for obtaining precise hardness results, but only rough estimation. The indirect results were compared with those obtained in another paper for the AlN and diamond-like films from direct measurements [18].

Constant pressure blister test, is another approach to analyze the mechanical properties (adhesion) of thin film systems, e.g. metallization layers [18]. In this test the characteristic blisters were growing at constant applied stress and this process was accompanied by an increase of the maximum meridional and radial stresses in the film membrane. The authors of [18] used the scanning electron microscope before and between experiments, in order to analyze the characteristics of deformation and failure mechanism. It was found that the active failure mechanism depends upon the mechanical properties of the film, substrate and interface, and may vary with environmental changes. It means that direct measurements of thin film mechanical properties, together with methodological improvement, have priority before creating a theoretical model.

In this paper we analyze various approaches to the microhardness measurements of thin films, in order to find the most suitable technique, to avoid different ISE, and to improve the existing methodological approaches.

2. Experimental details

Microhardness was studied in films obtained by sputtering and thermal deposition techniques. The Cu, Al, Ta, Ge and Sn films were prepared using a laboratory vacuum station VUP-5M, whereas, the Cu, Ge, and Ta films were obtained using DC magnetron sputtering and the Al and Sn films were deposited using thermal evaporation. The distance between the metal target in the sputtering chamber and the film's substrate was about 53 mm; The background pressure was approximately 4×10^{-5} Torr. Argon pressure in the DC magnetron sputtering was varied between 1.5 to 2 Torr. The applied target voltage for the Cu films was 315 V. For Ta, the applied target voltage was 215 V and for Ge, 2 kV. The micro-structural investigation of the obtained films was done using an optical microscope Nikon-Optiphot 100 and scanning electron microscope, Stereoscan 430 of Leica, operating at 20 keV. The film's composition was analyzed by an Energy Dispersive Spectrometry (EDS) accessory, mounted on the SEM. Film's thickness was measured using a profilometer system Alphastep 100. Microhardness measurements of the investigated films were performed using a standard tester with a flat-field microscope at a × 500 magnification. The indentation time was 15 s. Five to ten indentations were taken for each specimen. An extended description of this apparatus is given in [20]. Microhardness calculations were carried out according to [18-23]:

$$H = 1854 \times P/C^2, \tag{1}$$

where: H is the microhardness, P is the applied mechanical load, and C is the diagonal magnitude [μm] of the indention stamp. This diagonal magnitude was determined according to the following equation:

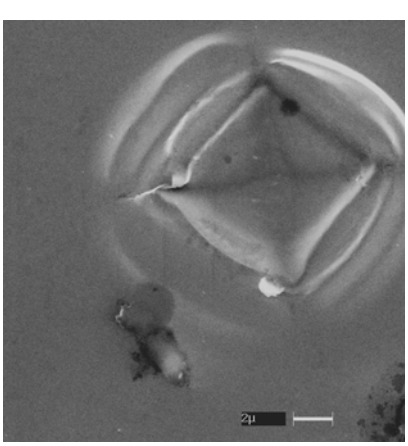
$$C = E \times N, \tag{2}$$

where N is the number of the drum steps on the tester, determined by the indention stamp size, and E is the length of one step (in μm).

3. Results and discussion

According to microhardness measurements principles [18-22], the obtained results with various weights should had been independent on the applied mechanical loads. This principle holds as long as structural effects or substrate influences are avoided. Hence, with a load increase on the measured sample, the dimension of the diagonal of the indentation stamp increases as well. Therefore, the conclusions of [2, 6, 12] concerning the influence of the testing loads on the microhardness results, seem to be wrong. The real goal of a microhardness measurement is to obtain film parameters in physical units, Pascal or kgf/mm^2, as described in references [3, 7, 8, 9], and not in non-dimensional relative units, as described in [12]. In this respect, it is of a great importance to choose the correct values of the applied loads in order to obtain the relevant indentation stamps without having indentation size effects (ISE), e.g., cracks or adhesive failure flaking within the visible substrate, as described in [3, 10, 11]. Due to the great difficulty in estimating the real microhardness values in cases where defects appear in the film, special theories for the composite hardness models were developed [14, 15, 17-19]. Jonsson and Hogmark's model [15] uses a geometrical approach to combine the hardness of the film and of the substrate according to the *Area Mixture* model. However, Ford [14] has reported that the model is unsuitable for indentation depths shallower than the film thickness. This is due to the film cracking which transmits a mechanical load directly into the substrate over the central part of the contact area. Burnett and Rickerby [14,15] developed the *volume mixture model* where the hemisphere under the indenter is related to the plastic volumes in a spherical cavity model, and is affected by the elastic and plastic properties of the measured material. However, this model was extended empirically to allow for the ISE, whereby the hardness depends on indentation depth, even for uncoated substrates. The most recent development of the model was the introduction of a more realistic geometry, whereby the indenter is allowed to penetrate into the substrate. This approach was used to interpret a wide range of experimental data. I. J. Ford [14] summarized that available models of the composite hardness of a coated substrate are not soundly based in theory**.**

What seems to be missing in the development of these models is a clear reference to the analytical work on spherical cavity analysis, which has been applied to the indentation of bare substrates. Therefore, further calculations, based on the volume mixture or area mixture model**,** can predict the hardness values of the measured depths. The reason for this is that the soft coatings are not deformed under indentation in the manner assumed in the model; instead, they flow outwards and upwards around the indenters and produce pile-ups. According to I. J. Ford [14], yielding of a harder substrate does not occur until the indenter has penetrated the film, causing the film material to deform and to produce a bulge at the surface. We clearly observed such bulges on the outer contour of the indentation stamp during SEM-investigation (Fig. 1). In such cases the final microhardness was directly obtained [2]. However, it must be emphasized that for ISE with upper layer destruction, the indenter does not penetrate into the substrate, but comes into contact with the lower subsurface layers of the film (Fig. 2). This fact was proven by SEM X-ray investigations (Fig. 3), in which it is clearly seen, that the composition of the lower surface is the same tantalum as the film itself. Hence we may conclude that the films tested, made of different materials, demonstrate more strength and elasticity, than the expected values.



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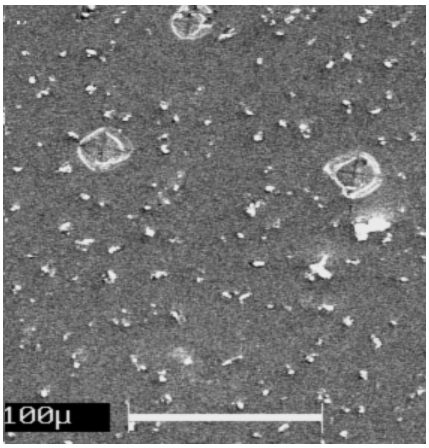

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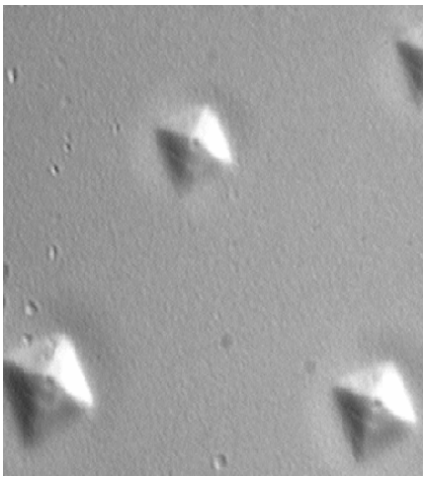
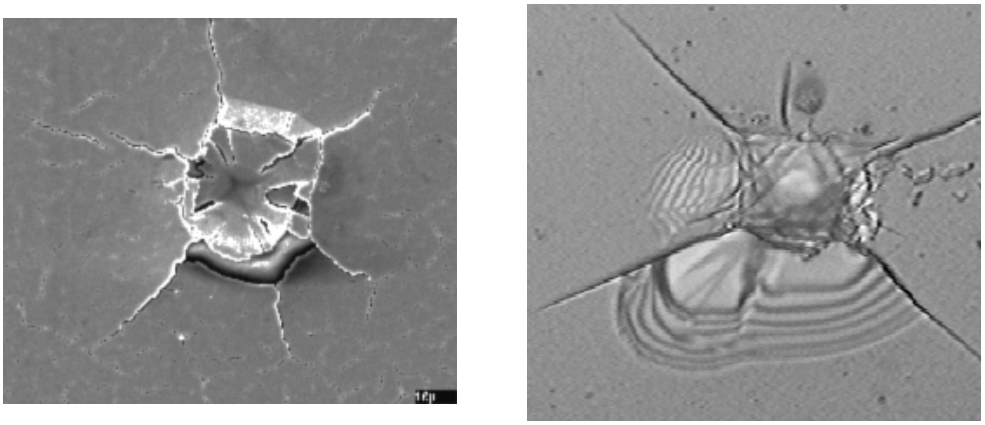


c

Fig. 1. Plastic deformation without Indentation Size Effects (ISE).
a – tantalum film on glass substrate; b – copper film on glass substrate;
c – germanium film on glass substrate

These assumptions were proven in our microhardness analysis, obtained on: Cu, Al, Ta, Sn, Ge and DLC - thin films, produced in the Technological Academic Institute - Holon, thin films laboratory. Measurement results are presented in Fig. 4. Indentation stamp views are shown, using an optical and SEM- images (Figs. 1, 2, 5). It should be noted that all measurements were taken on homogeneous plain films with no initial cracks and discontinuities after indentation, as demonstrated in Fig. 1. These homogeneous plain films enabled us to make the microhardness calculations, using Eqs. (1,2). Investigation by SEM revealed that ISE in sputtered Ta films appeared with small cracks (Fig. 2a). Another type of ISE in Ge films using higher loads, was obtained as a pattern of relaxation of internal stresses (Fig. 2b).



a b

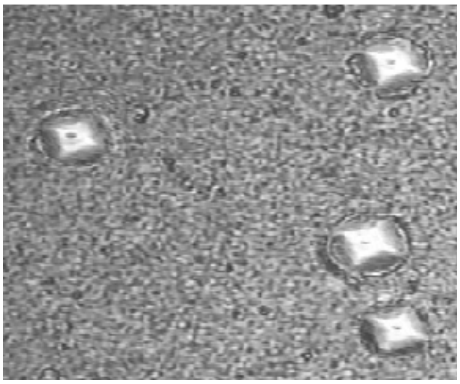


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Fig. 2. Indentation size effects without penetrating into the substrate.
a – tantalum film on glass substrate; b – germanium film on glass substrate; c – diamond - like - carbon (DLC) film on silicon substrate

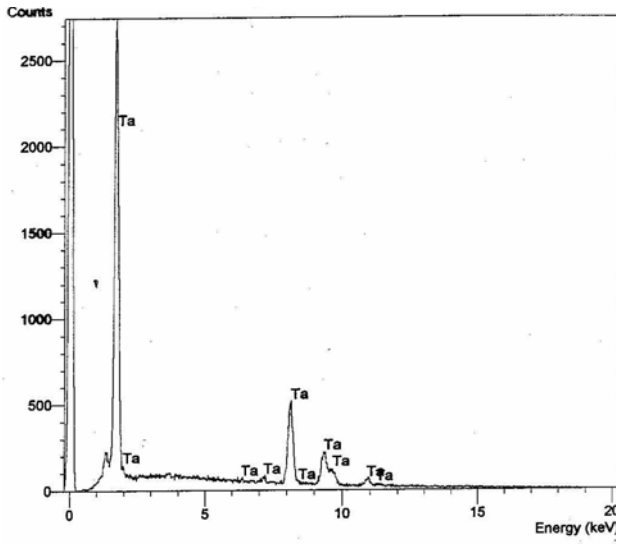
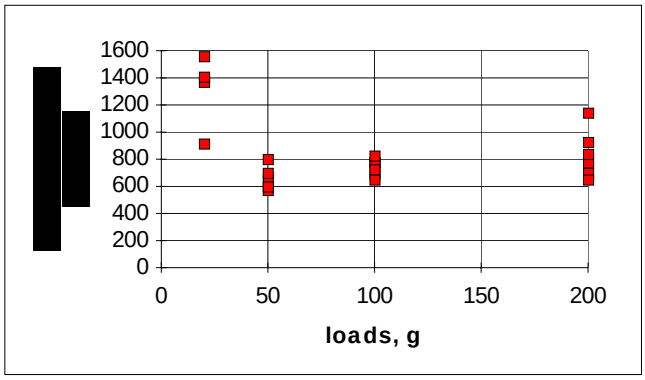


Fig. 3. Spectrometer analysis of lower Ta-film layer after microhardness measurement with ISE.

That leads to some non-uniformity of the final microhardness value (Fig. 4a, b) and to its reduction (Fig. 4c) under higher loads of 200 g. Maximum dispersion of the results is obtained under

20 g loads (Fig. 4a). This dispersion is explained by the small size of the indentation stamp diagonal. The differences in the diagonal size can sometimes lead to a false estimation in the microhardness readout of about 800 kgf/mm^2. Best results with minimum dispersion were obtained in measurements under loads of 50 and 100 g (Fig. 4). Microhardness values for evaporated Cu films varied between 600 to 800 kgf/mm^2. The microhardness values of sputtered Ta films varied between 800 and 1300 kgf/mm^2 for 50g and 100g weights (Fig. 4b). The most uniform results were obtained for thermal evaporated Sn films, with microhardness varying between 700 to 1000 kgf/mm^2, for 50 g and 100 g loads.



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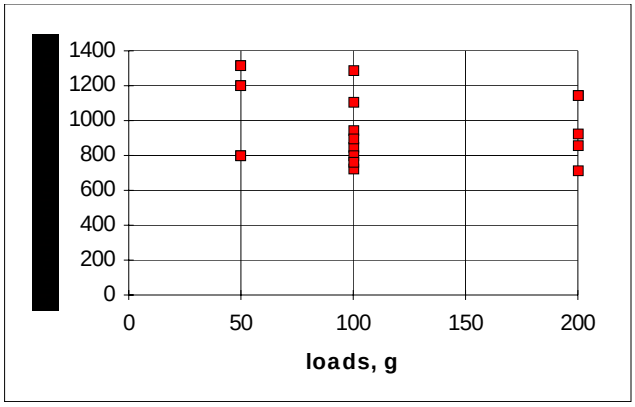

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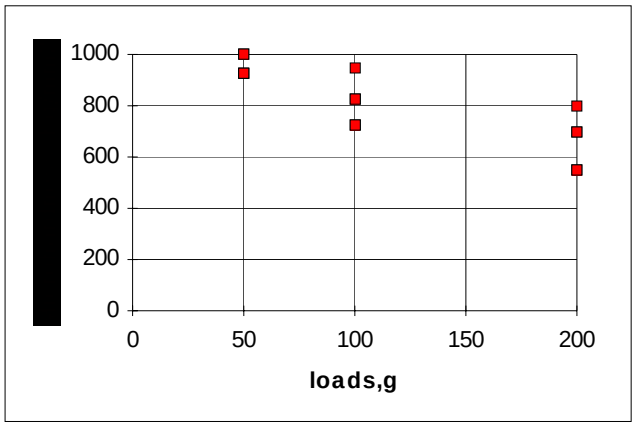
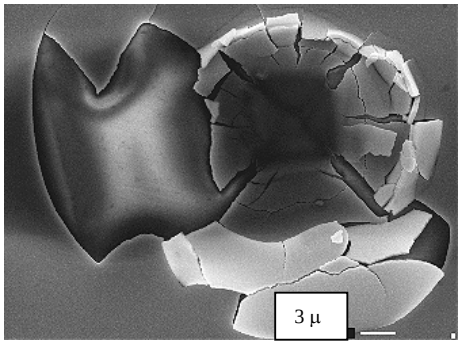


c

Fig. 4. Microhardness results influenced by indentation loads for various thin films. a - copper thin film, evaporated on glass substrate; b – tantalum thin film, sputtered on glass substrate; c – tin thin film, evaporated on glass substrate.

Doubtful microhardness results appear when ISE is accompanied with indenter penetration into substrate (Fig. 5). This penetration may give a completely brittle fracture on the thin film's surface (Fig. 5a). In this case the microhardness results are not proper, and the indentation should be repeated with an appropriate load. Another problematic case is when the indenter penetration into the substrate looks like a *double stamps* (Fig. 5b, c). Here, the smaller indentation image correlates with the substrate hardness and the larger indentation correlates with the thin film itself. The *composed microhardness* method was suggested for evaluation and calculations of these results.



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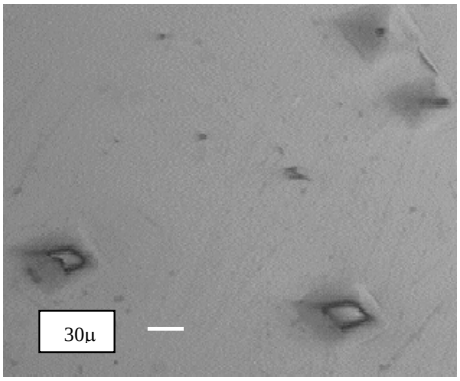

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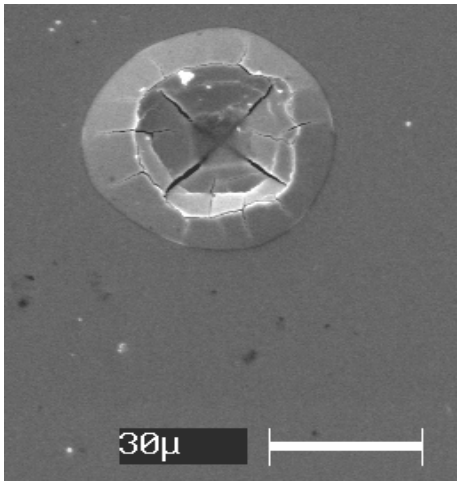
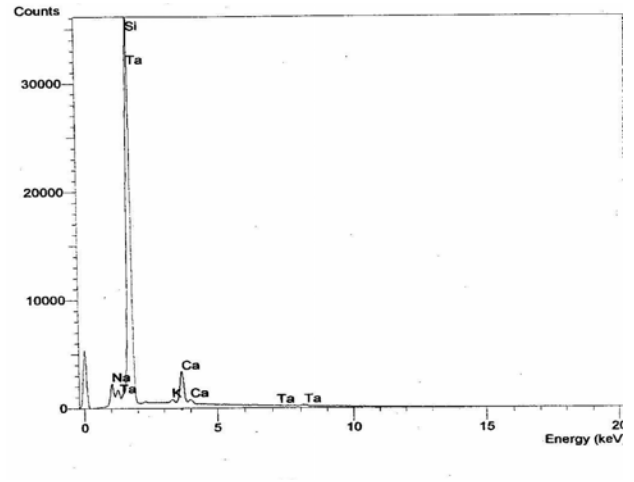


c

Fig. 5. ISE views of penetration into substrate.
a – tantalum film on glass substrate; b – DLC-films on silicon substrate; c – copper film on glass substrate

However, this technique does not seem to be correct, since it represents the destructive type of testing which does not belong to microhardness method. The additional confirmations to the destructive nature of such indentation is presented in Fig. 6. The spectrometer analysis shows that there are traces of substrate's composition (glass) in the spectrum of the tantalum film following an indenter penetration into the substrate.



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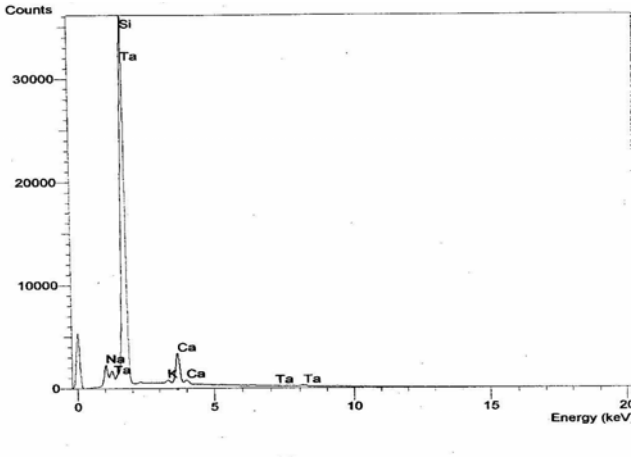

b

Fig. 6. Spectrometer analysis of lower film in the case of brittle failure and ISE with penetration into substrate.
One-and b – two different places on the lower surface of destroyed tantalum film.

Thus, a proper measuring method should consist of obtaining a regular stamp, without any kind of ISE, by applying the right load, which will vary for each film and each thickness. This way the final microhardness values should be independent on the film thickness, which was always the case in our measurements (Fig. 7).

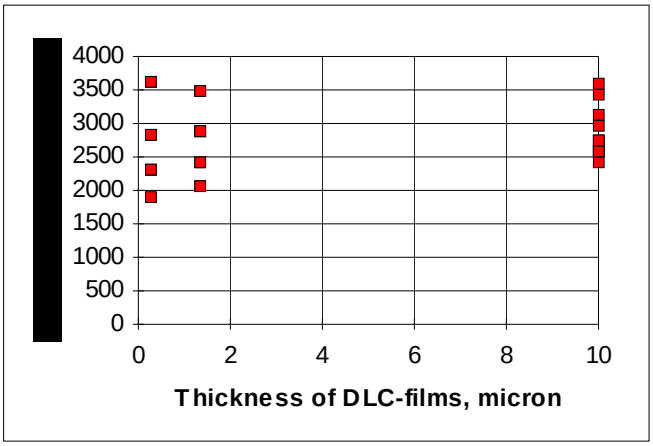

Fig. 7. Microhardness vs. thickness for DLC-films, deposited with 200 W of R.F. power.

4. Conclusions

The right choice of mechanical loads in microhardness measurements is of great importance, in order to avoid indentation size effects (ISE) in thin film systems. This can be done using an iteration method accompanied by a structural control of each indentation stamp's quality. Application of a composed hardness model is not always justified, but is suitable in the presence of indentation size effects (ISE) without indentor's penetration into substrate. For each thin film system there is an optimal value of mechanical load, in order to obtain minimal dispersion of the microhardness values. With this optimized load value there are no indentation size effects at all. The proposed method of microhardness measurements and its advantages were proven by numerous microhardness measurements of thin films, deposited by various sputtering and evaporating methods.

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