Thin coatings and films hardness evaluation

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Abstract. The existing thin coatings and films hardness evaluation methods based on indentation with pyramidal indenter on various scale levels are expounded. The impact of scale factor on hardness values is performed. The experimental verification of several existing hardness evaluation methods regarding the substrate hardness value and the “coating – substrate” composite hardness value is made.

1. Introduction
Surface coatings and films are widely used in micro- and nanoelectronics. Usually, the indentation with pyramidal indenter is used for evaluation of coatings’ hardness, because it is known as one of the most simple and high efficient methods. Frequently, the indentation is the only possible method of thin coatings mechanical properties control, because thin films and coatings are not suitable for preparing the proper samples for other types of mechanical tests. However, the hardness determination of thin items, including the thin coatings, requires the observance of the condition, concerning the relation \( \varphi \) between item thickness \( H \) and indent depth \( h \) (\( \varphi = H/h \)). According to the international standard documents, regulating the hardness evaluation procedure, at least \( H/h = 10 \) should be provided for correct hardness estimation. The violation of this condition inevitably leads to the substrate properties influence on the obtained hardness value. Moreover, for example, for 1 \( \mu \text{m} \)-thickness coating the indent depth \( h \) should not exceed 0.1 \( \mu \text{m} \) or 100 nm which corresponds to nanoscale level (in accordance with ISO 14577-1:2002). However, it is known that the hardness values sufficiently increase with the indentation load (and along with that, the indent depth) reduction due to scale factor impact [1,2]. Therefore, hardness values, determined on the nanoscale level, may be greatly higher, than even on micro scale level and even more on macro scale level.

If it is not possible to provide the required value of \( \varphi \), (that is \( \varphi \) sufficiently less than 10), the substrate will exert the influence on coating hardness test result. In this case, it is necessary to evaluate the hardness of substrate thereafter the hardness of “coating - substrate” composite system and after which calculate the coating hardness using the obtained results. The equations for calculation of coating hardness for such cases were proposed in several papers, for example [3,4].

2. Experimental section
This paper is focused on the research of scale factor impact on thin coatings hardness, evaluated with pyramidal indenter on macro and micro scale levels with the observance of \( \varphi \geq 10 \) conditions. Hardness tests were made at TiN-based 23…25 \( \mu \text{m} \) thickness coatings with carbon steel (0.45 %C) substrate. Hardness was evaluated using the Instron Tukon 2500 and Buehler MicroMet 5124 Vickers hardness testers with the various indentation loads \( F \) (from 0,049 N (5 gf) up to 2,94 N (300 gf)).
The results of tests are performed in Table 1. Indents depths $h$, diagonals $d$, volumes $V_i$, strained volumes beneath the indents $V_{eps}$ as well as hardness values for different indentation loads $F$ are submitted.

### Table 1. Indents depths $h$, diagonals $d$, volumes $V_i$, strained volumes beneath indents $V_{eps}$ as well as hardness values of the TiN-based 25 $\mu$m coating for indentation with different loads $F$.

| No. | $F$ (gf) | $N$ (kgf) | $d$, $h$, $\varphi$, $d$, $h$, $\varphi$ ($\mu$m) | $V_i$, $\mu$m$^3$ | $V_{eps}$, $\mu$m$^3$ | $HV$, N/mm$^2$ | Indentation scale range |
|-----|----------|-----------|-------------|-----------|-------------|----------------|-----------------|
| 1   | 5        | 0.049     | 1.8, 26    | 1.43$\cdot$10$^4$ | 1.37$\cdot$10$^4$ | 28 057 | 2 860 | micro |
| 2   | 10       | 0.098     | 2.6, 68    | 4.14$\cdot$10$^4$ | 3.97$\cdot$10$^4$ | 25 722 | 2 622 | micro |
| 3   | 15       | 0.147     | 3.4, 51    | 9.61$\cdot$10$^4$ | 9.23$\cdot$10$^4$ | 24 064 | 2 453 | micro |
| 4   | 20       | 0.196     | 4.0, 44    | 1.51$\cdot$10$^5$ | 1.45$\cdot$10$^5$ | 22 740 | 2 318 | micro |
| 5   | 25       | 0.245     | 4.5, 38    | 2.24$\cdot$10$^5$ | 2.15$\cdot$10$^5$ | 22 563 | 2 300 | micro |
| 6   | 50       | 0.490     | 6.6, 26    | 7.00$\cdot$10$^5$ | 6.72$\cdot$10$^5$ | 20 758 | 2 116 | micro |
| 7   | 100      | 0.980     | 9.8, 18    | 2.24$\cdot$10$^5$ | 2.15$\cdot$10$^5$ | 19 159 | 1 953 | micro |
| 8   | 200      | 1.960     | 14.3, 12   | 7.04$\cdot$10$^5$ | 6.76$\cdot$10$^5$ | 17 952 | 1 830 | micro |
| 9   | 300      | 2.940     | 18.4, 10   | 1.49$\cdot$10$^6$ | 1.43$\cdot$10$^6$ | 16 128 | 1 644 | macro |

Elastic-plastic strained volumes under the indent $V_{eps}$ were determined using the equation, introduced by paper authors [5], according to which strained volume under the indent is directly proportional to indent volume $V_i$:

$$V_{eps} \approx 96 \cdot V_i,$$

where $V_i$ clearly depends on the indent depth $h$:

$$V_i = 8.167h^3.$$  \hspace{1cm} (2)

As it follows from Table 1, the indentation was made on macro and micro scale levels, and the hardness values increase sufficiently with the indentation load reduction. More clearly the scale factor impact on hardness tests results can be performed in figure 1, where the relations between Vickers hardness $HV$ and indentation load $F$ (a) and between Vickers hardness $HV$ and indent depth $h$ (b) are shown.

![Figure 1](image.png)

**Figure 1.** Relations between Vickers hardness $HV$ and indentation load $F$ (a) and between Vickers hardness $HV$ and indent depth $h$ (b) obtained for TiN-based 25 $\mu$m coating.

The reasons for scale factor impact on hardness tests results are commonly divided into physical, mechanical and instrumental [6]. One of the main reasons is the multiple reducing the strained volume of material under the indent with the transition from macro to micro scale level. In the experiment, presented above, the strained volume reduced approximately by 3 orders of magnitude (see the table 1).
Among others, the additional experiments for coating hardness evaluation providing the $\phi < 10$ conditions were made. These tests were made at TiN-based 6 $\mu$m coating with carbon steel (0.45 %C) substrate. The results of these tests are represented in Table 2.

Coating hardness values were evaluated with the regard to substrate influence. The experiments were shown that equation

$$H_f = H_s + \frac{H_c - H_s}{2C^2_h - C^2_c (H/H_s)}$$

(3)

where $H_f$ is the coating hardness; $H_s$ is the substrate hardness; $H_c$ is the “substrate - coating” composite system hardness; $C^2 = 2\sin^211^\circ$, proposed by [4], has the close approach of calculated hardness value to the hardness value, evaluated $\phi \approx 10$. Hardness values, calculated using the equation (3) are performed in Table 2.

Table 2. Indentation tests results of TiN-based 6 $\mu$m coating in case of observance and violation of the condition, regulating the $\phi$ relation.

| No. | $F$ | d, $\mu$m | h, $\mu$m | $\phi$ | $HV_s$ | $HV_f$ calculation using the equation (3) | Error $\Delta$, % |
|-----|-----|----------|----------|------|--------|--------------------------------|-----------------|
| 1   | 25  | 0.245    | 3.9      | 0.56 | 10.5   | 32 373  3 300                       | 3 120  318        | 3 300  0        |
| 2   | 50  | 0.490    | 5.6      | 0.80 | 7.5    | 28 940  2 950                      | 3 002  306        | 2 950  6.7      |
| 3   | 100 | 0.981    | 10.7     | 1.53 | 3.9    | 15 804  1 611                      | 2 708  276        | 1 611  8.6      |
| 4   | 150 | 1.472    | 16.9     | 2.41 | 2.5    | 9 614   980                       | 2 560  261        | 980  26.0       |

3. Conclusions

The following conclusions can be made by the results of experiments. The comparison of several coatings hardness values should be made not with the equal indentation loads but with the equal indent depths. It provides the equality of indent volumes, strained volumes beneath the indenter and, as a result, the similar scale factor impact. The substrate impacts on coating hardness value if $\phi < 10$, and the less the $\phi$, the more the impact. The experimental verification of different equations, suitable for coatings hardness evaluation at $\phi < 10$ with regard to the substrate hardness and the “substrate - coating” composite system hardness, performed that equation (3) can be successfully used if $\phi \geq 4$. In the case of $\phi < 4$ the error $\Delta$ starts to increase sharply (see the table 2), what can be explained by increasing the role of substrate and the impact of the scale factor.

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