[1.]Jozef PETRÍK, [1.]Peter BLAŠKO, [2.]Juraj ĎURIŠIN,
[1.]Andrea VASILŇÁKOVÁ, [3.]Anna GUZANOVÁ

THE ORIENTATION OF THE GRAINS AND INDENTATION SIZE EFFECT

[1.]Technical University of Košice, Faculty of Materials, Metallurgy and Recycling, Letná 9, Košice, SLOVAKIA
[2.] Institute of Material Research, Slovak Academy of Sciences, Watsonova 47, Košice, SLOVAKIA
[3.]Technical University of Košice, Faculty of Mechanical Engineering, Letná 9, Košice, SLOVAKIA

Abstract: The aim of the submitted work is to study the relationship between the orientation of the grains and the Indentation Size Effect (ISE). The micro–hardness was measured by tester Hanemann with applied loads ranged between 0.09807 N to 0.9807 N. The micro–hardness was measured in the middle of in the center of each of the four grains of the sample – the copper of semiconductor quality (99.995 %). The influence of the load and the orientation of the grains on the micro–hardness and the ISE was evaluated by the Analysis of Variance (ANOVA), it is statistically significant for both factors. The presence and character of ISE were evaluated using Meyer's index n, PSR method, and Hays – Kendall approach. The relationship between the crystallographic orientation of the grain and ISE was confirmed.
Keywords: copper, grain orientation, micro–hardness, Indentation Size Effect (ISE)

1. INTRODUCTION

The micro–hardness test is a common procedure for determining mechanical properties of a small volume of materials. Several authors such [1] found that the principle of Vickers micro–hardness method is identical to macro–hardness test, except for test load lower than 1.691 N (200 g). The micro–hardness can be used for the measurement of the hardness of small parts, thin layers and also to identify the phases in metallography. The advantage of the Vickers test is the independence of the applied load. The shape of the Vickers indentation is geometrically self–similar at all test loads. It is therefore expected that the value of the hardness is constant within the statistical precision over a very broad load range as long as the tested sample is reasonably homogeneous.

If a very low load is used, the measured hardness is usually high; with an increase in test load, the measured hardness decreases. Such a phenomenon is referred to as a **"**normal" indentation size effect (ISE). According to [2] using a load–dependent hardness in material characterization may result in some unreliable conclusions.

The ISE may be caused by the testing equipment (the device for the measurement of indentation diagonals, the determination of the applied load belongs in this group [1, 2, 3], intrinsic properties of the tested material (work hardening during indentation, load to initiate plastic deformation, indentation elastic recovery, elastic resistance of the materials [1,3,4], the method of preparing the tested samples (grinding, polishing and the resulting residual stress) and possibly other factors (indenter/sample friction, lubrication and also corrosion [2, 5**]**.

In contrast to **"**normal" ISE, a reverse (inverse, RISE) type of ISE, where the apparent micro–hardness increases with increasing test load, is also known. It mostly takes place in materials in which plastic deformation is predominant. As demonstrated [4] reverse ISE can be explained regarding the existence of a distorted zone near the crystal–medium interface, effects of vibration and bluntness of indenter, the applied energy loss as a result of specimen chipping around the indentation and the generation of the cracks.

In the literature, there are many examples, which reveal that the **"**normal" ISE occurs in brittle materials. On the other hand, the literature is scarce regarding reverse ISE. It is reported mainly for materials undergoing plastic deformation [1].

In addition to the above–mentioned ISE resources, other possible sources are also mentioned in the literature, such as different grain orientation. Zhang and Bradt [6] confirmed the effect of crystallographic grain orientation on ISE parameters in the study of MgF_2 and also TiO_2 and SnO_2 crystals. As far as metals are concerned, Feng and all [7] dealt with this relationship for copper single crystal specimen with an <111> orientation using Berkovich nanoindentation test and Britton and all [8] for α–titanium polycrystal, also by Bekovich test. The influence of crystallographic orientation on ISE has also been confirmed for these metals. At the same time, it should be borne in mind that the above results were obtained by experiments in the field of nano–hardness.

The aim of the experiment was to study the influence of grain orientation and applied load on the values of the micro–hardness and then on the indentation size effect of the copper.

2. EQUIPMENT, MATERIAL AND METHODS

The sample used in the experiment was the copper of semiconductor quality (99.995 wt. % of Cu). After cutting by cooled diamond saw the *sample was* embedded in the resin (dentacryl) and gradually ground by the silicon paper 80 ANSI/CAMI grit with intensive water–cooling. Fine grinding with papers in the sequence 220, 240 and 3000 ANSI/CAMI grit was followed. The metallographic surface was mechanically polished with the water suspension of Al_2O_3 (400 ml H_2O/ Al_2O_3) to a mirror finish and finally polished with the diamond paste (0.5 μm) moistened with kerosene. Finally, to make the grain boundaries visible, the surface was etched using a solution of: 30 ml of HCl, 5 g of $FeCl_3$ and 100 ml of H_2O. Four grains

(No. 1, 2, 3 and 4) with a diameter of 8 to 22 mm were made visible by etching. Prior to the micro–hardness measurement, was studied by X–ray diffraction (XRD) method. X–ray patterns were carried out on a Philips X'Pert Pro diffractometer equipped with Cu cathode at operating parameters of 40 kV and 50 mA. The samples were scanned from 40–150 º 2θ using a scan rate of 0.5º/min and a step size of 0.03342º 2θ.

Micro–hardness was measured with manually – operated tester Hanemann, type Mod D32 fitted to microscope Neophot–32. A reference block – CRM (certified reference material) with specified hardness H_c = 195 HV0.05 and standard uncertainty u = 4.0 HV0.05 was used for the calibration of the tester. The average mico–hardness was 193.88 HV0.05, relative repeatability r_{rel}, = 2.72 %, relative error of tester E_{rel} = –0.57 % and relative expanded uncertainty of calibration U_{rel}= 5.56 %, the tester meet the requirements of the standard ISO 6507–2:2018 [9].

Table 1 – Characteristics of the individual grains

grain No.	$\overline{HV}$	s	p_1	outliers	HV0.05	U_{rel} (%)	p_2	H_{PSR}		
								a_2	c_2	A_1
1	55.73	6.10	0.0091	0	60.32	24.47	1.65E–9	59.0	30.3	56.1
2	52.18	2.81	0.7333	0	53.62	27.37	2.21E–5	51.8	40.1	51.2
3	55.99	3.00	0.0855	0	53.51	27.46	2.54E–6	52.9	35.4	53.0
4	56.45	2.74	0.2680	0	56.60	25.90	3.03E–5	52.0	41.6	53.0

The same operator measured the micro–hardness of selected areas on the metallographic surface of the sample according to ISO 6507–1:2018 [10]. The applied loads P were 0.09807 N, 0.24518 N, 0.49035 N and 0.9807 N. Five indentations were made at each load with the load duration time 15 seconds. The result of the measurement was a "cluster" of 20 indentations approximately in the middle of each of the four grains. The average value of the micro–hardness of individual clusters $\overline{HV}$, its standard deviation s, the micro–hardness HV0.05, and its relative expanded uncertainty U_{rel} are in Table 1. Average speed of the indenter's penetration into the sample was calculated by method, described in [11], it ranged between 1.86 and 2.35 μm s^{-1}.

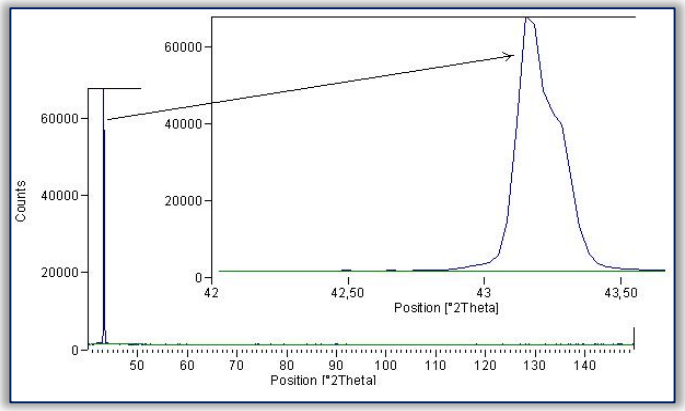

Figure 1 – Grain No. 2: the relationship between the angle incidence of the radiation and its intensity

The values of the experimental error in the form of relative expanded uncertainty U_{rel} (coverage factor k = 2) calculated according to 6507–2:2018 [10] are listed in Table 1. The uncertainty of the reference block was not taken into account since there is a big difference between the hardness of the reference block, made of iron, and copper. Listed values of the U_{rel} are overestimated and therefore have only informative character.

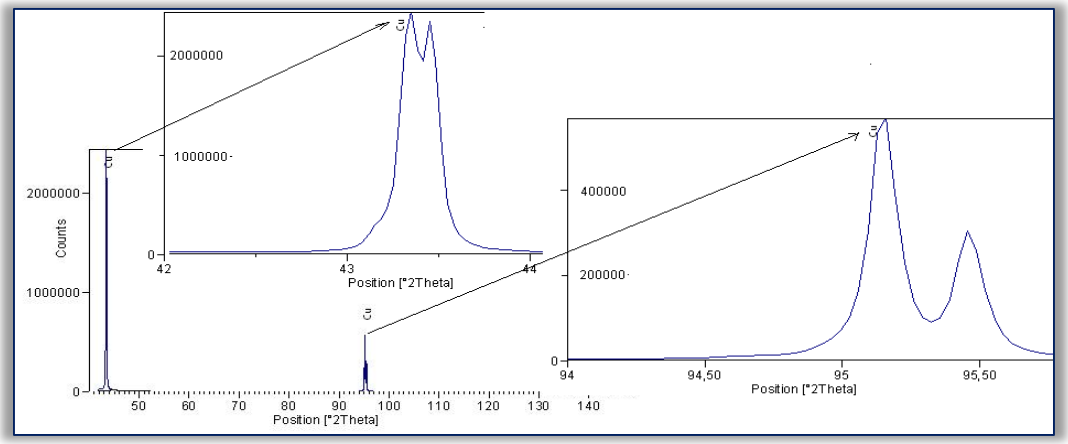

Figure 2 – Grain No. 3: the relationship between the angle incidence of the radiation and its intensity

Grubbs' test (significance level α = 0.05) was used for detection of statistical outliers. Their presence would indicate a measurement process suffering from special disturbances and out of statistical control. The normality was determined by Freeware Process Capability Calculator software (Anderson – Darling test, p_1 >0.05 (significance level α) for normal distribution). The normality and the outliers were determined for files involving values of one "cluster". Because the distribution of the values measured on the grain No. 1 was different from the normal distribution, non–parametric tests were used, or caution should be exercised when interpreting the results obtained by parametric test or by the Analysis of Variance (ANOVA).

The crystallographic orientation of the four grains considered was only qualitative – thus, whether or not there is a difference in grain orientation. Figures 1 (grain No. 1) and 2 (grain No. 2) show the relationship between the angle of incidence of radiation and intensity and confirm their different orientation. The smallest difference in orientation is between grains No. 2 and 4.

As for the values of the micro–hardness, according to two–way ANOVA with replication, the grain (p = 3.64E–13) and the load (p = 6.48E–25) both have a statistically significant effect on the measured value of the micro–hardness. If the p value is less than the significance level α = 0.05, the factor has a statistically significant effect. The interaction between both factors is also statistically significant (p = 4.09E–12).

The the examples of the diffraction analysis results presented in Figure 1 and 2 shows that the crystallographic orientation of the individual grains varies. It is believed that due to the homogeneous chemical composition of the sample, the measured micro–hardness as well as the Meyer's index n and PSR parameters will be the same in each grain. The aim of the measurement is to determine whether the grain orientation affects these indicators.

The paired t–test confirmed a statistically significant difference between the mean micro–hardness values for grains No. 2 and 1 (p = 0.0012), No. 2 and 3 (p = 0.0) and also No. 2 and 4 (p = 0.0). Based on the values of micro–hardness we can be 98.93% confident that medians are different using non–parametric Mood's median test. The significance level α = 0.05 for both tests.

Table 2 – Meyer's index and parameters of PSR model

grain No.	n	A_{ln}	a_1	a_2	c_0	c_1	c_2	W	A_1	a_1/a_2	c_1/c_2
1	2.1503	6.1903	–0.4507	311.69	–0.1845	10.9800	160.00	0.0060	296.92	–0.001450	0.068625
2	2.0395	5.7514	0.0966	274.19	–0.0766	4.8547	211.98	0.0089	270.94	0.000352	0.022902
3	2.0032	5.6999	0.5526	279.97	–0.1087	7.4945	187.07	0.0198	280.26	0.001974	0.040063
4	1.9639	5.5774	0.7528	275.05	–0.0619	4.8021	220.08	0.0192	280.29	0.002737	0.021820

3. RESULTS

Meyer's Power Law and proportional specimen resistance (PSR) are two principal approaches to describe ISE quantitatively [3]; the simplest way to describe the ISE is Meyer's Law:

$$P = Ad^n \tag{1}$$

The parameters n and A are determined by exponential curve fitting to indentation diagonal d (mm) versus applied load P (N) or n and A_{ln} from straight line graph of ln (d) versus ln (P). Meyer's index n or work hardening coefficient is the slope, and coefficient A_{ln} is the y–intercept of the line. The index n < 2 for "normal" ISE, n > 2 for reverse ISE. If n = 2 the micro–hardness is independent of the load and is given by Kick's Law. The values of n and A_{ln} are in Table 2. The values of Meyer's index are close to value 2 for grains No. 2, 3 and 4, thus falling within the validity area of Kick's law. The differences between the grains are minimal. The exception is grain no. 1 with significantly reverse ISE. The values of Meyer's indices correspond to reality. As with most ductile metallic materials, even in the case of copper, ISE has reverse behaviour in nature [12].

Using the single–factor ANOVA, the statistical significance of the applied load effect on the measured micro–hardness values for all grains (value p_2 in Table 1) was evaluated even in the case of grains No. 2 and 3, whose Meyer's index is close to 2.

The proportional specimen resistance model of Li and Bradt (PSR) may be considered a modified form of the Hays/Kendall approach to the ISE [2]. Several authors as [1,2,3,13] have proposed that the ISE may be described by the (2):

$$P = a_1 d + a_2 d^2 \tag{2}$$

Li and Bradt pointed out that the parameters a_1 (N mm^{-1}) and a_2 (N mm^{-2}) of (2) are related to the elastic and plastic properties of the material, respectively [5,13].

The parameter a_1 characterizes the load dependence of micro–hardness and describes the ISE in the PSR model. It consists of two components: the elastic resistance of the test sample and the friction resistance developed at the indenter facet/sample interface [1,3]. The parameter a_2 is directly related to load–independent micro–hardness sometimes referred to as "true hardness" H_{PSR} [2].

$$H_{PSRa2} = 0.1891 \cdot a_2 \tag{3}$$

Equation (2) may be rearranged in the form:

$$P/d = a_1 + a_2 d \tag{4}$$

The parameters a_1 and a_2 of (4) may be obtained from the plots of P/d (N mm^{-1}) against d (mm). Equation (5) can be regarded as a modified form of the PSR model.

$$P = c_0 + c_1 d + c_2 d^2 \tag{5}$$

The parameters c_0 (N), c_1 (N mm^{-1}) and c_2 (N mm^{-2}) of (5) may be obtained from the quadratic regressions of P (N) against d (mm). Parameter c_0 is associated with residual surface stress in the sample and parameters $c_1 \approx a_1$ and $c_2 \approx a_2$ are related, respectively with the elastic and plastic properties of the sample [1,2].

The ratio c_1/c_2 (mm) is a measure of the residual stress due to machining and polishing. The values of the indices obtained by modified PSR are given Table 2. It is assumed that the differences should not be large, all grains are grind and polished

as well. As can be seen from the c_1/c_2 values shown in Table 2, these differ, despite the assumption. It is possible that this factor may be influenced to some extent by grain orientation. In this regard, further experiments are appropriate to confirm or rebut this assumption. The numerical values of of c_0 (N) is proportional to residual stresses in the sample. A literature survey [1] reveal expected relationship between c_0 and c_1/c_2, this fact confirms Figure 3.

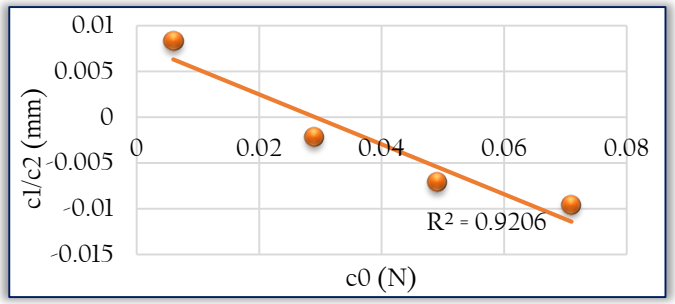

Figure 3 – The relationship between values of c_0 and the ratio of c_1/c_2

Meyer's index n decreases with increasing of average micro–hardness HV. Inverse relationship between the micro–hardness and n was observed for CRMs made of iron or heat treated steel with micro–hardness between 195 HV0.05 and 519 HV0.05, heat – treated carbon steel and aluminum alloy EN 6082 or technically pure metals (Al, Zn, Cu, Fe, Ni, Co), all with reverse ISE [14,15]. Given examples were not, except for grinding and polishing, deformed.

Hays and Kendall proposed the existence of a minimum test load W (N) necessary to initiate plastic deformation. Below it only elastic deformation occurs. In that event, the load dependence of hardness is expressed:

$$P = W + A_1 d^2 \quad (6)$$

Where A_1 (N mm^{-2}) is a constant independent of load. The values of W and A_1 may be obtained from the regressions of P (N) against d^2 (mm) [1]. The values of the indices obtained by modified PSR are in Table 4.

The "true hardness" by analogy to a_2 can be calculated as $H_{PSR}c_2$ using c_2 or $H_{PSR}A_1$ using A_1 in equation (3). Calculated "true hardness" is listed in Table 1.

4. CONCLUSIONS

- Different crystallographic orientation of grains was confirmed by diffraction.
- The crystallographic orientation statistically significantly affects micro–hardness and subsequently ISE behaviour.
- The paired t–test confirmed a statistically significant difference between average micro–hardness of some grains.
- Despite expectations, values c_0 and c_1/c_2 are affected by grain orientation. On the contrary, the expected relationship between these values has been confirmed.
- "True hardness" calculated using c_2 it is significantly underestimated and therefore unreliable.
- Further research will focus on determining the repeatability of measured data as well as on the possible relationship between grain orientation and c_0 and also c_1/c_2 indices.

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