Available online at www.sciencedirect.com

ScienceDirect

Journal of the European Ceramic Society 30 (2010) 1967–1969

www.elsevier.com/locate/jeurceramsoc

Commentary

Some comments about "Comment on hardness definitions", by J. Malzbender [J. Eur. Ceram. Soc. 23 (2003) 1355–1359]

Gelson Biscaia de Souza[a,*], Alexandre Mikowski[b,c], Carlos Maurício Lepienski[b], Carlos Eugênio Foerster[d]

[a] Departamento de Ciências, Universidade Estadual de Maringá, CEP 87020-900, Maringá, PR, Brazil
[b] Departamento de Física, Universidade Federal do Paraná, CP 19044, CEP 81531-990, Curitiba, PR, Brazil
[c] Centro de Engenharia da Mobilidade, Universidade Federal de Santa Catarina, CP 246, CEP 89219-905, Joinville, SC, Brazil
[d] Departamento de Física, Universidade Estadual de Ponta Grossa, CEP 84030-900, Ponta Grossa, PR, Brazil

Received 25 November 2009; received in revised form 21 February 2010; accepted 17 March 2010

Abstract

Malzbender has suggested a model to determine hardness and elastic modulus as a function of the mechanical energies involved during tip penetration in instrumented indentations tests. However, the values obtained with these expressions are not consistent with the ones determined by the well-accepted Oliver and Pharr method. After revision, based on Malzbender's study itself, equations were rewritten and then, the obtained indentation hardness (H) for soda-lime glass was in agreement with the literature data. However, the reduced elastic modulus (E_r) was still about 20% higher than the values in the literature. Developing Malzbender's proposal by the inclusions of additional mechanical energy assumptions, a new expression for E_r is now suggested. Using the new expression, the hardness and reduced elastic modulus agreed very well with the Oliver and Pharr method.

Keywords: Surfaces; Hardness; Mechanical properties; Glass; Instrumented indentation

Malzbender's paper[1] discusses different definitions on hardness obtained by indentation tests with conical indenters. In particular, the ratio between indentation hardness (H) and Martens hardness (HM)[1,2] is presented as

$$\frac{HM}{H} = \left[\sqrt{\frac{\alpha}{\pi(\tan\gamma)^2}} + \frac{\varepsilon}{\beta}\sqrt{\frac{\alpha\pi}{4}\frac{H}{E_r}}\right]^{-2}, \tag{1}$$

where $\gamma = 70.3°$ is the half-angle of the conical indenter, which presents the same contact area under a particular load for ideal Vickers and Berkovich indenters[1,3]; $\varepsilon = 0.75$ corresponds to the geometric constant for paraboloid indenters; $\beta = 1.034$ is a correction factor used to compensate the lack of symmetry in pyramidal tips[4]; E_r is the reduced elastic modulus; and $\alpha = \pi \tan\gamma\sqrt{1 + (\tan\gamma)^2}$ corresponds to the proportionality between the area and the maximum depth in the Martens hardness (for a Berkovich indenter, $\alpha = 26.44$). The term H/E_r in Eq. (1) is also present in another relationship, previously proposed by Malzbender and de With,[5] for the deformation energies involved during an indentation test:

$$\frac{W_p}{W} = 1 - \frac{W_e}{W} = 1 - \left(\frac{\varepsilon}{2} + \frac{\beta}{\pi\ \tan\gamma}\frac{E_r}{H}\right)^{-1}, \tag{2}$$

where W_e, W_p and W correspond to the elastic, plastic and total mechanical energies, respectively, which are calculated from loading–unloading curves.

Based on the statements described above, Malzbender[1] proposed a hardness equation, dependent on the mechanical energies:

$$H = \frac{W}{h^3}\frac{3(\tan\ \gamma)^2}{\pi}\left[1 + \frac{\varepsilon}{2W_e/W - \varepsilon}\right]^2, \tag{3}$$

where h is the maximum tip penetration depth.

However, following step by step Malzbender's proposition,[1] it was verified that Eq. (3) is incorrect and, actually, it needs to

* Corresponding author. Tel.: +55 44 3522 1851; fax: +55 44 3261 4828.
E-mail addresses: gbsouza@uem.br, gelbsou@yahoo.com.br (G.B. de Souza).

0955-2219/$ – see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jeurceramsoc.2010.03.013

be written as

$$H = \frac{W}{h^3}\frac{3}{\pi(\tan\gamma)^2}\left[1 + \frac{\varepsilon}{2W/W_e - \varepsilon}\right]^2. \quad (4)$$

In the same paper, Malzbender[1] also derived an energy-dependent equation for the reduced elastic modulus E_r:

$$E_r = \frac{3}{\beta}\frac{W_e}{h^3}\left(\frac{\varepsilon W}{W_e} + \frac{2\,\tan\gamma}{2 + \varepsilon W/W_e}\right). \quad (5)$$

This equation is also incorrect. By combining Eqs. (1) and (2) or by substituting Eqs. (4) in (2), it can be verified that the reduced elastic modulus also needs to be corrected to:

$$E_r = \frac{3}{\beta}\frac{W_e}{h^3}\left[\frac{(W/W_e)^2}{\tan\,\gamma(1 - \varepsilon W_e/2W)}\right]. \quad (6)$$

Such hardness and elastic modulus calculation can be interesting for materials pile-up or sink-in during indentations, when the contact area cannot be correctly determined.[4]

We used experimental data in order to compare Eqs. (3)–(6) with the Oliver and Pharr[3,4] definitions for indentation hardness and reduced elastic modulus, given respectively by

$$H = \frac{P_{max}}{24.5h_c^2 + \sum_{i=1}^{8} C_i h_c^{[2^{-i+1}]}} \quad (7)$$

$$E_r = \frac{\sqrt{\pi}}{2\beta}\frac{S}{\sqrt{A(h_c)}}. \quad (8)$$

In these equations, P_{max} corresponds to the maximum applied load, h_c is the contact depth and C_i are numerical coefficients necessary to correct the tip rounding effect. S represents the contact stiffness, and $A(h_c)$ the projected contact area. In both equations the contact depth is $h_c = h - \varepsilon(P_{\max}/S)$.

The reference material chosen for the indentation tests was a commercial soda-lime glass (SiO_2 71%, Na_2O 13.2%, CaO 10%, Al_2O_3 0.7%; MgO 4% in weight), since it is an isotropic material with well-known mechanical properties.[3,6,7] The surface finishing for the received samples was in a good condition and they were cleaned only with acetone in an ultra-sound bath. In order to reduce the pre-existing residual stresses, annealing treatment at 540 ± 1 °C during 1 h in air was carried out and cooled to room temperature at a rate of 1 °C/min. The instrumented indentation tests employed a Berkovich-type diamond tip. Ten indentation tests were made with an applied load of 100 mN. This load allowed the tip to reach a penetration depth of (1015.5 ± 3.0) nm.

Table 1 shows indentation hardness values calculated using Oliver–Pharr as well as Malzbender's (original and corrected equations) methods. According to Oliver–Pharr (Eq. (7)), the hardness of the soda-lime glass is (6.4 ± 0.1) GPa, which is in agreement with the literature data.[3,8–10] On the other hand, the hardness is overestimated $(5.5 \pm 0.3$ TPa) when it is calculated using the Malzbender's Eq. (3). However, by determining the hardness with the revised Malzbender's equation (Eq. (4)), the obtained value drops to (6.3 ± 0.1) GPa. This value is now in agreement with the one determined by the Oliver and Pharr and the accepted literature data.

Table 1
Indentation hardness (H) and reduced elastic modulus (E_r) for soda-lime glass calculated using the Oliver–Pharr method,[3,4] Malzbender's method[1] and the revised Malzbender's equations. The value that corresponds to the E_r solution, here proposed, is also shown.

Method	H (GPa)	E_r (GPa)
Oliver–Pharr (Eqs. (7) and (8))	6.39 ± 0.08	74.5 ± 0.5
Malzbender (Eqs. (3) and (5))	5533.01 ± 288.15	151.1 ± 1.5
Revised Malzbender (Eqs. (4) and (6))	6.30 ± 0.05	91.9 ± 0.5
Proposed E_r (Eq. (10))	–	70.1 ± 0.4

Table 1 also shows the reduced elastic modulus that was calculated using the Oliver–Pharr (Eq. (8)), the original (Eq. (5)) and the corrected (Eq. (6)) Malzbender's equations. As expected, the reduced elastic modulus according to the Oliver–Pharr method $(74.5 \pm 0.5$ GPa) is in good agreement with the literature.[3,7] Otherwise, reduced elastic modulus obtained by the Malzbender's suggestion (Eq. (5)) is overestimated $(151.1 \pm 1.5$ GPa). By using the revised form of the corresponding Malzbender's equation, the reduced elastic modulus decreases to (91.9 ± 0.5) GPa. This value still remains about 23% higher than the value obtained by the Oliver–Pharr method (see Table 1, revised Malzbender).

As can be seen in Eq. (2), the involved mechanical energies are connected to the H/E_r ratio, which is a material intrinsic property. According to Cheng and Cheng,[4,11,12] this same ratio can also be described by

$$\frac{W_e}{W} = \frac{1}{\kappa}\frac{H}{E_r}, \quad (9)$$

where κ is a function of the indenter half-angle. For a set of ceramic and metal materials, κ^{-1} is approximately 5.33.[1,4,11] Since H and E_r, from the Malzbender's proposition,[1] are derived from Eq. (2), we used indentation data of the soda-lime glass and the Oliver–Pharr method in order to verify the consistency of this equation as well as Eq. (9). The measured elastic and total mechanical energies for soda-lime glass were $W_e = (17.42 \pm 0.17)$ nJ and $W = (36.39 \pm 0.17)$ nJ, respectively. The H/E_r ratios, calculated using these energies in Eqs. (2) and (9), were compared to the ratio obtained directly by the Oliver–Pharr method, as shown in Fig. 1a. The difference between the value obtained by the Oliver–Pharr method and the Cheng–Cheng's proposal was 4.5%, while with the Malzbender–de With's proposal (Eq. (2)), it was 20%.

Combining the revised Malzbender's indentation hardness (Eq. (4)) with the Cheng–Cheng's relationship (Eq. (9)), it is possible to obtain another expression for the reduced elastic modulus:

$$E_r = \frac{15.99}{\pi(\tan\,\gamma)^2 h^3}\frac{W^2}{W_e}\left(1 + \frac{\varepsilon}{2W/W_e - \varepsilon}\right), \quad (10)$$

where all the terms were previously described. In Fig. 1b, the E_r values are compared according to Oliver–Pharr (Eq. (8)), the revised Malzbender's Eq. (6) and the proposed Eq. (10). As can be observed, the reduced elastic modulus determined by Eq. (10) is more consistent with the one calculated by the well-accepted

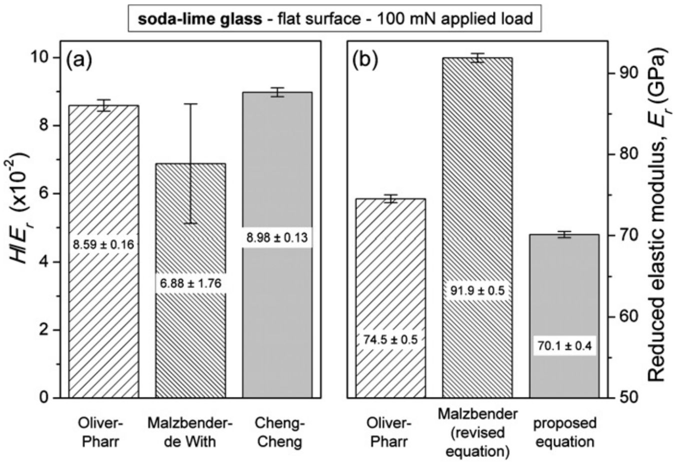

Fig. 1. (a) The H/E_r ratio obtained using the Oliver–Pharr method[3,4] (Eqs. (7) and (8)), Malzbender–de With's proposal[5] (Eq. (2)) and Cheng–Cheng's proposal[11,12] (Eq. (9)). (b) Reduced elastic modulus according to Oliver–Pharr (Eq. (8)), Malzbender after revision (Eq. (6)) and our proposal (Eq. (10)). The E_r values in the graph (b) depend on the corresponding H/E_r in the graph (a).

Oliver–Pharr method and the literature data,[3,7] despite a relative error of about 5%.

In conclusion, for instrumented indentations carried out on flat soda-lime glass, the equation based on the mechanical energies to determine the indentation hardness as proposed by Malzbender (after revision) is very well in agreement with the value obtained by the well-established Oliver–Pharr method. On the other hand, the expression proposed for the reduced elastic modulus (even after revision) does not agree with Oliver–Pharr. Reduced elastic modulus values, consistent with Oliver–Pharr and also based on the mechanical energies, were calculated using an equation derived from the Malzbender's proposal for hardness and the H/E_r ratio as described by Cheng and Cheng.

References

1. Malzbender J. Comment on hardness definitions. *J Eur Ceram Soc* 2003;**23**:1355–9.
2. European Standard EN ISO 14577-1 : 2002. *Metallic materials—instrumented indentation test for hardness and materials parameters. Part 1. Test method*. European Committee for Standarization. Brussels; 2002.
3. Oliver WC, Pharr GM. An improved technique for determining hardness and elastic modulus using load and displacement sensing indentation experiments. *J Mater Res* 1992;**7**:1564–83.
4. Oliver WC, Pharr GM. Measurement of hardness an elastic modulus by instrumented indentation: advances in understanding and refinements to methodology. *J Mater Res* 2004;**19**:3–20.
5. Malzbender J, de With G. Indentation load–displacement curve, plastic deformation, and energy. *J Mater Res* 2002;**17**:502–11.
6. Malzbender J, Toonder JMJ, Den Balkenende AR, de With G. Measuring mechanical properties of coatings: a methodology applied to nano-particle-filled sol–gel coatings on glass. *Mater Sci Eng R* 2002;**36**:47–103.
7. Míchel MD, Mikowski A, Lepienski CM, Foerster CE, Serbena FC. High temperature microhardness of soda-lime glass. *J Non-Cryst Solids* 2004;**348**:131–8.
8. Gong J, Si W, Guan Z. Effect of load-dependence of hardness on indentation toughness determination for soda-lime glass. *J Non-Cryst Solids* 2001;**282**:325–8.
9. Kese KO, Li ZC, Bergman B. Method to account for true contact area in soda-lie glass during nanoindentation with the Berkovich tip. *Mater Sci Eng A* 2005;**404**:1–8.
10. Howell JA, Hellmann JR, Muhlstein CL. Nanomechanical properties of commercial float glass. *J Non-Cryst Solids* 2008;**354**:1891–9.
11. Cheng Y-T, Cheng C-M. Relationships between hardness, elastic modulus, and the work of indentation. *Appl Phys Lett* 1998;**73**:614–6.
12. Cheng Y-T, Cheng C-M. Scaling, dimensional analysis, and indentation measurements. *Mater Sci Eng Rev* 2004;**44**:91–149.