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# The first indenter-sample contact and the indentation size effect in nano-hardness measurement

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**Abstract**

The problem of appropriately defining the First Indenter-Sample Contact (FISC) is known to be a major problem in hardness measurement and particularly in nano-indentation. Another difficulty arises from the determination of the real indentation area when a piled up occurs near the indentation print. This phenomenon is known as the Indentation Size Effect (ISE). The indentation piles up can be particularly heavy for low loads which affects the nano-indentation measurements. The Loading Displacement Curves (LDC) with ISE can be modelled by the PSR law (Proportional Specimen Resistance). We show that an error on the FISC validates PSR law even if no ISE occurs meaning that it is illusory to quantify the ISE with only one loading curve. To overcome this difficulty, a statistical protocol is proposed that consists in simultaneously analysis a fixed number of loading curves. With this approach, a new equation of LDC is proposed that allows determination of Hardness, ISE coefficient and the FISC position on each curve (98 LDC are recorded from a fused silica standard). The uncertainty on each coefficient is determined by a bootstrap protocol.


*Keywords:* Nano-hardness; Fused silica; Loading displacement curves; Indentation size effect; First indenter-sample contact

## 1. Introduction

The problem of appropriately defining the First Indenter-Sample Contact (FISC) is known to be a major problem in hardness measurement and particularly in nano-indentation that induces a false origin of the load-displacement curves from which mechanical properties will be evaluated. An additional difficulty arises from determining the real indentation area when a piled up occurs around the indentation print [1]. In this paper, we show that these two major problems are strongly linked in the mathematical formulation of the hardness evaluation. In this work, we focus upon two laws in the hardness measurement: the Kick's [2,3] and the PSR laws (Proportional Specimen Resistance) [4].

## 2. Materials and method

Nano-indentations were performed using a Nano Indenter XP® (MTS Nano Instruments Oak Ridge, USA). Sample is fused silica that is commonly used as the calibrating material. Indentations are performed using a diamond Berkovich indenter at room temperature. Experiments are performed using the Continuous Stiffness Measurement (CSM) at constant loading rate (0.05 $s^{-1}$) until a maximum indentation depth of 2500 nm. The applied load $F$, contact stiffness $S$ and indentation depth $h$ are continuously measured. 98 indentation measurements are recorded and shown on Fig. 1 (left).

## 3. Pre-treatments of experimental indentation curves

To calculate indentation curve parameters, three pre-treatments have to be completed. In a first time, the loading part must be extracted from the experimental curve. At the end of loading, curves present a crossover (slow decrease of $\partial^2 F/\partial h^2$) on around 0.85 $F_{max}$ where $F_{max}$ is the maximal load. As a consequence, we only retain points of the loading curves that are under this value. The second treatment consists in transforming the penetration value $h$ as i.i.d (independence and identical distribution) to avoid statistical artefacts in the future statistical regression. Then, load data are averaged each 20 nm leading to i.i.d $h$ values. The depth $h$ measured during the indentation includes the elastic deformation of the sample around the indentation in addition to the contact depth $h_c$. The depression of the sample around the indentation ($h_s = h - h_c$) is caused by elastic deformation and must be subtracted

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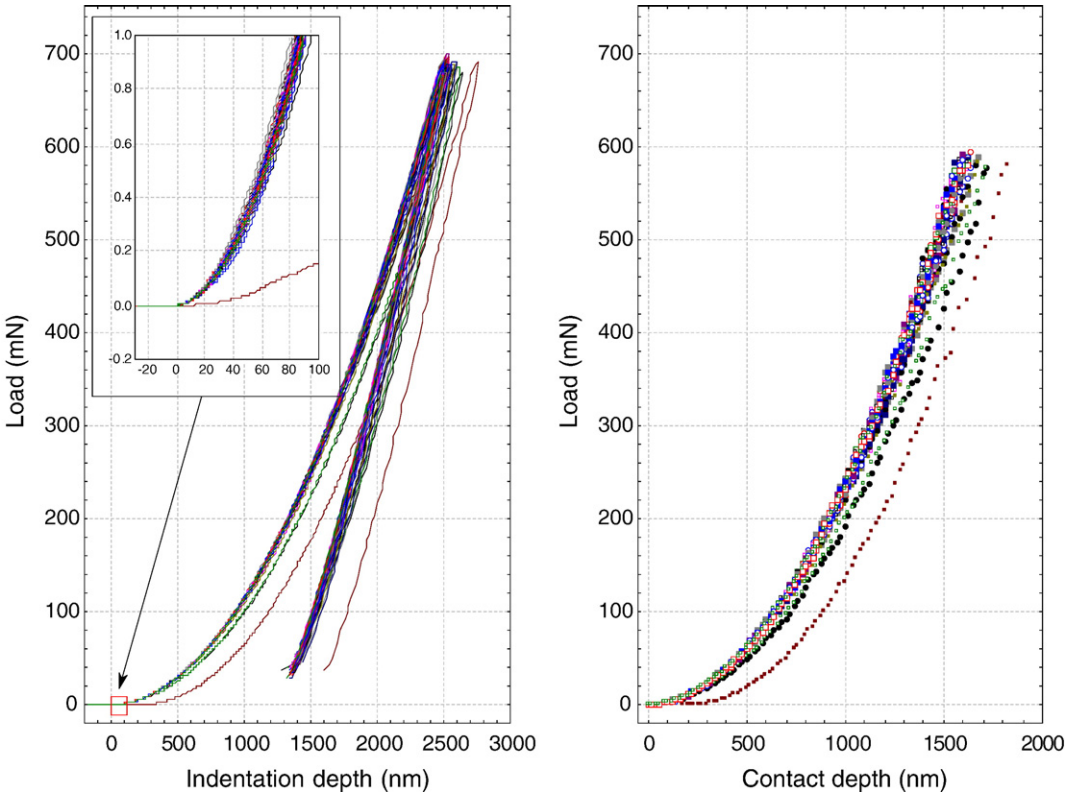


Fig. 1. 98 initial load–displacement curves on fused silica (left) and load–displacement curves after statistical pre-treatments using Oliver and Pharr method (right).

from the data to obtain the actual depth of indentation and hardness. Oliver and Pharr [5,6] developed an expression for $h_c$ at the maximum load (required for hardness calculation) from $h$, and gives $h_c = h - \varepsilon F/S$ where $\varepsilon = 0.75$ for the Berkovich indenter and $S$ is the stiffness that can be deduced from the slope of unloading curve (in our case, the stiffness is obtained by CSM). The Fig. 1 (right) represents the final loading curves, after these three pre-treatments, from which our statistical treatments are performed.

## 4. Kick's law and the PSR one

The Kick model [2,3] is given by the following equation:

$$F = ah_c^2 \tag{1}$$

This is in fact the basic definition of the hardness $H$ taking into account the spring back:

$$H = F/A \tag{2}$$

where $A$ can be related geometrically to $h_c$ with $A = 24.56h_c^2$ for Berkovich indenters. In presented units ($H$ in GPa, $h_c$ in nm, $F$ in mN) $\alpha = 25.5610^{-6}$, and then:

$$H = \alpha^{-1}F/h_c^2 \tag{3}$$

The PSR model [4] is given by the following equation:

$$F = ah_c^2 + bh_c \tag{4}$$

This model includes the well known Indentation Size Effect (ISE) when piled up occurs, i.e., when material is forced up along the sides of the indentation tip [7,8]:

$$H = H_0 + \beta/h_c \tag{5}$$

and leads to:

$$F = \alpha(H_0h_c^2 + \beta h_c) \tag{6}$$

It should be pointed out that works on the ISE are initially based on non-instrumented micro-hardness. Then several hardness tests are conducted at different loads, the diagonal print is measured and ISE is estimated by Vingsbow's law [1,7,8]. At the opposite, only one force–depth curve allows to estimate the diagonal print at different loads and then, theoretically, to estimate the ISE. However, it was shown on non-instrumented indentation tests that a large number of measurements (that increases inversely with the diagonal length of the print) is needed for a given load [1]. The uncertainty is due to both optical resolution and the heterogeneity of materials. This clearly means that it becomes illusory to quantify the ISE with only one loading curve and then, a statistical protocol must be used by simultaneously analysing several loading curves. Unfortunately, in the literature, statistical treatments are often performed with a single loading curve. Even if several loading curves are available, they are treated independently to obtain hardness characteristics from which descriptive statistics are computed [9].

## 5. Error measurements on loading curve, a new model

As pointed out by Grau [10] and Ullner [11], the problem of appropriately defining the first indenter-sample contact is quite complex. This leads to define a zero position in a scale with arbitrary selected point $h=0$. By introducing the noise $\Delta$ on $h_c$ in Eq. (1), due to error measurements on the zero level, and if the Kick's model holds i.e. $H=\alpha F/(h_c+\Delta)^2$, the following expression is obtained:

$$F = \alpha(Hh_c^2 + 2\Delta H h_c + H\Delta^2) \tag{7}$$

By neglecting the term $H\Delta^2$, Eq. (7) reduces to the PSR model with $\beta=2\Delta H$ due only to noise measurement without taking account the ISE. This means an important feature: PSR law can be validated even if only Kick's law hold with an error measurement on $h_c$. More importantly, an increase in systemic error leads to the detection of an unrealistic ISE. This unrealistic ISE increase rises artificially with the material hardness leading to false correlation between hardness and ISE. A new equation of the loading curve is introduced by taking into account both ISE ($H=H_0+\beta/h_c$) and noise in Eq. (7):

$$F = \alpha((H_0 + \beta/h_c)h_c^2 + 2\Delta(H_0 + \beta/h_c)h_c + (H_0 + \beta/h_c)\Delta^2) \tag{8}$$

that leads to:

$$F = \alpha((H_0)h_c^2 + (2\Delta H_0 + \beta)h_c + 2\beta\Delta + H_0\Delta^2 + \beta\Delta^2/h_c) \tag{9}$$

This final equation (Eq. (9)) includes both ISE and noise measurement. To integrate all curves from measurements, two

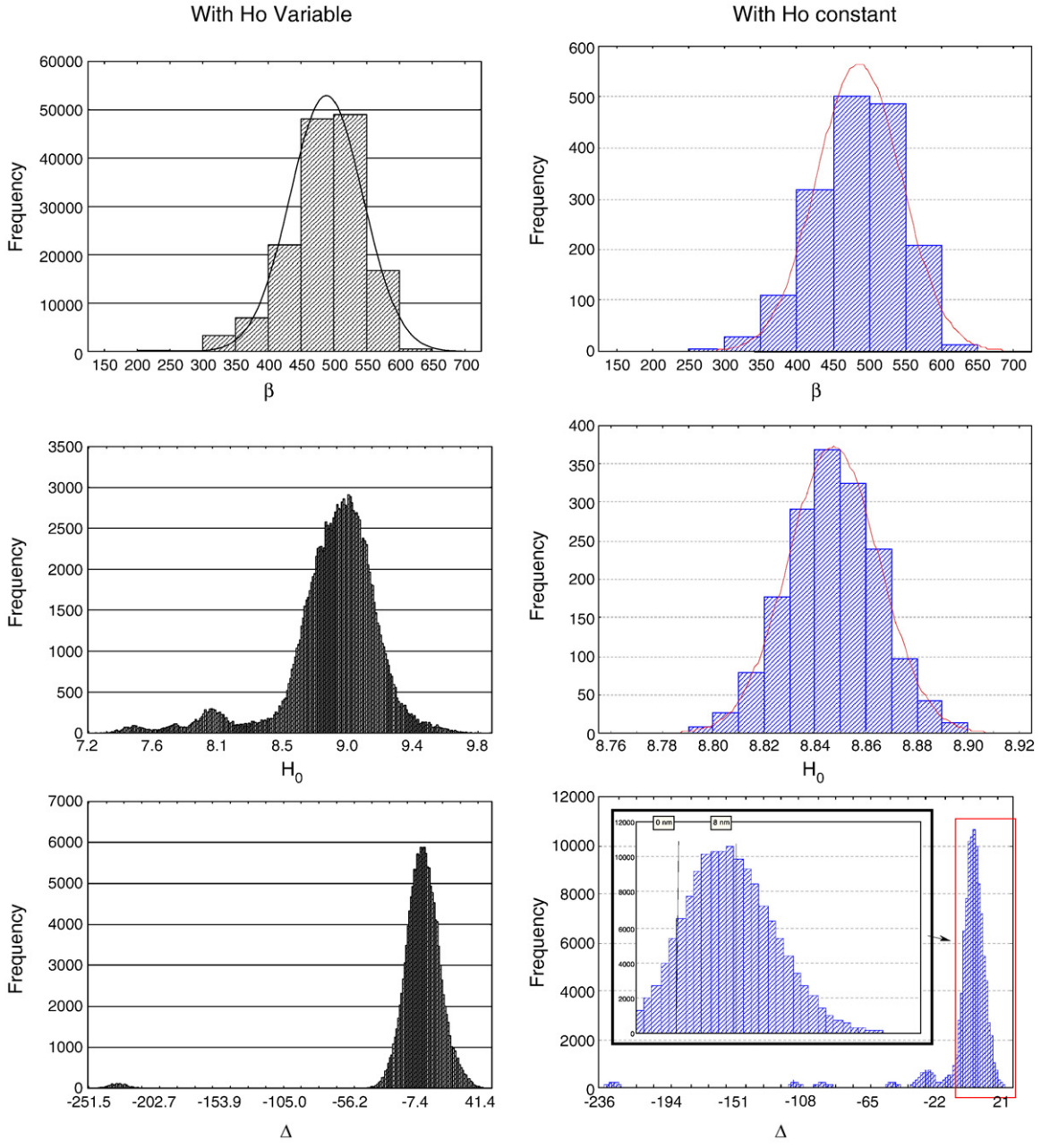


Fig. 2. Histograms of ($H_0$, $\Delta_i$, $\beta$) when they are estimated from models given by Eq. (10) (left) and Eq. (11) (right).

optimisation methods can be performed. In the first optimisation the hardness is assumed to be variable or each curve while the $\beta$ value is hold constant. Consequently, the $n$ pairs ($\Delta_i$, $H_{0,i}$) (corresponding to the $i$th loading curve with $p_i$ sampling points) and $\beta$ are obtained by solving the following functional:

$$\min_{\substack{H_{0,1}\ldots H_0,n\\ \Delta_1\ldots\Delta_n,\beta}} \sum_{i=1}^{n}\sum_{j=1}^{p_i}[F_{i,j}-\alpha((H_{0,i})h_{cj}^2+(2\Delta_i H_{0,i}+\beta)h_{cj} + 2\beta\Delta_i + H_{0,i}\Delta_i^2+\Delta_i^2\beta/h_{cj})]^2 \quad (10)$$

under constrains $H_{0,i}>0$. In the second method, $H_0$ is assumed to be constant and the functional becomes:

$$\min_{H_0,\Delta_1\ldots\Delta_n,\beta} \sum_{i=1}^{n}\sum_{j=1}^{p_i}[F_{i,j}-\alpha((H_0)h_{cj}^2+(2\Delta_i H_0+\beta)h_{cj} + 2\beta\Delta_i + H_0\Delta_i^2+\Delta_i^2\beta/h_{cj})]^2 \quad (11)$$

The main problem consists in determining the confidence intervals of all parameters of Eqs. (10)–(11). For this reason, a double bootstrap is computed to calculate the variation into each curve and between all curves. In a first time, let analyse the $\beta$ coefficient that quantifies the ISE. Both histograms (Fig. 2) are quite similar and present Gaussian shapes. The two methods lead, in a first approximation, to the same result ($\beta_{eq.\ 10}=486_{\pm 120}$,$\beta_{eq.\ 11}=484_{\pm 118}$) meaning that $\beta$ value does not depend on the heterogeneity of the hardness (initial hypothesis well posed). Similarly, the mean value by taking $H_0$ variable ($H_0=$ 8.86 GPa) is closed to the mean value by taking $H_0$ constant ($H_0=8.84$ GPa). The dispersion, characterised by the standard deviation, is more important for variable (0.33) than for $H_0$ constant (0.02).

The final question about this value: "*Is the ISE really exists*". More precisely, this assertion is verified if statistically $\beta>0$. Answer comes from the statistics calculated from the bootstrap set: from 1000 simulations, no negative or null values were found. As a consequence the probability to assert fallacy that ISE exists is less than 1/1000 and then the ISE cannot be rejected. The ISE exhibited by fused silica (previously shown by Li and Bradt in the measurement of the hardness of vitreous silica [4]**)**, was confirmed by Atkinson by introducing a new description, demonstrating a common phenomenology regardless of the different micro-mechanisms sustaining indentation [12]. Concerning the first indenter-sample contact, the histograms show the distribution of the zero-value $\Delta$. As it could be observed, a mode on histograms appears at the high value $\Delta=-225$ that corresponds to a loading curve on Fig. 1. This curve must be visually shifted of the same value on the left to be at the origin. Five curves possess "abnormally" negative values of $\Delta$ (false detection of the surface before contact), the 93 others possess a gaussian density with a standard deviation of 6 nm, characterising the error on the zero level determination. Our method has then well corrected the contact depth to obtain an accurate value of hardness and quantify the ISE.

## 6. Conclusion

We have shown that an error on the first indenter-sample contact validates PSR law even if no indentation size effect occurs meaning that more than one curve is necessary to detect the ISE. An equation of a loading displacement curve is proposed and is associated with a statistical protocol to quantify the ISE. As a result of the bootstrap, an ISE is exhibited by fused silica, its hardness is estimated to $8.84_{\pm 0.02}$ GPa and the error on the first indenter-sample contact is quantified for each loading curve to be around ±6 nm.

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