UNCERTAINTY ANALYSIS OF AREA CALIBRATION IN NANOINDENTATION

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Abstract

Nanoindentation instruments are commonly used to study hardness, elastic modulus and other related mechanical properties at the nanoscale. They are a popular tool for the study and design of thin film systems, nanocomposites and other nanostructured materials and devices. Metrological issues such as uncertainty analysis and traceability are often treated only briefly. In many cases only the statistical variability is taken into account although the uncertainties of the method itself are in the range of percent.

It is well known that the correct determination of the area function of the indenter plays a crucial role. However, this can also be a difficult step in the traceability measurement chain.

The probably simplest and most widely used calibration procedure for area function is by measuring a well-characterized reference sample. We analyze the uncertainties related to this procedure and compare the impact of different sources of uncertainties, such as quality of reference sample, data acquisition, data processing and choice of fitting procedure. The uncertainty analysis is performed using NIGET, an open-source software for general nanoindentation data processing.

Keywords: Nanoindentation, uncertainty analysis, area

1. INTRODUCTION

The determination of mechanical properties using nanoindentation is a well-established technique which has been used for decades for material assessment and quality control. In these tests an indenter with well-defined shape is pressed into the material and the response of the material in terms of the depth of the imprint and its area are converted into properties characterizing stiffness and hardness.

With the improvement of nanoindentation instruments, the reduction of noise, improvement of thermal stability, the field of application broadened from the classical metals, ceramics and glasses to thin films, nanoparticles, nanocomposites as well as polymers and biological samples e.g. [1-6]. It has also been used to map surface properties across a sample surface, e.g. [7].

Uncertainty analysis is often left aside. This is partly due to the fact that for many samples the variance due to surface roughness and/or sample heterogeneity is much higher than the expected uncertainties due to noise of the instrument. As noise has been largely reduced in modern instruments, the uncertainty of the contact area becomes an important source of uncertainty.

The determination of the contact area itself remains a problematic aspect of nanoindentation measurements. This is a crucial quantity and the effects of incorrect determination of area when pile-up or sink-in occur are well-known [1, 8]. The recommended solution is to measure the indent using atomic force microscopy (AFM). For large numbers of measurements which are necessary for, e.g. mappings of nanocomposites or depth dependences of thin films this is hardly feasible.

Methods which determine area function of an indenter tip at once are better suited for large numbers of measurements and automation. The tip can be either directly measured using AFM [9], this can be made easily traceable but is time consuming and the need to move the tip between the instruments presents a certain risk of damage. On the other hand, the calibration standard method [10] requires no tip manipulation, however, traceability is problematic. Firstly, not all standard reference samples which are used for nanoindenters, such as fused silica or BK7 glass, are shipped with an appropriate calibration certificate for the elastic modulus. Secondly, the calibration procedure involves a large amount of highly complex data processing. This is usually performed within the instrument software and thus the user is limited by the details of the documentation and the variability of the software.

In this work we will focus on the aspect of uncertainty propagation from the noise of the nanoindenter and uncertainty of the elastic modulus of the reference sample to the area function. The effects of the choice of the function to fit, the covariance model, the number of data, the uncertainty of the elastic modulus of the reference sample are analyzed and compared. The area function itself is rarely of interest, in the vast majority of cases the interest is in the hardness and elastic modulus whose uncertainties are also studied. Special attention is paid to very shallow indentations, since they are especially sensitive to numerical effects.

1.1. Experimental

Experimental data were measured using a Hysitron TI 950 nanoindenter and a BK7 reference sample. The Hysitron indenter has a very low noise floor below 30 nN in load and 0.2 nm in displacement with a resolution of 1 nN in load and 0.02 nm. The thermal drift was less than 0.05 nm/sec, the machine compliance was around 1 nm/mN.

BK7 is known for its highly homogenous mechanical properties, and good time stability. In this case the certificate provided by the manufacturer stated an uncertatinty of indentation hardness of 4 %. The uncertainty of Young's modulus was not stated, however, based on long term experience we assumed an analoguous value of 4 %. Young's modulus itself is stated as 90.47 GPa.

The tip was a standard diamond Berkovich tip. Two tips were used, with a nominal tip radius of 50 nm and 100 nm, respectively.

One hundred nanoindentations were performed in a force-controlled regime with a linear loading period of 5 s, a hold period at the maximum load for 2 s and a linear unloading period of 5 s. The maximum load varied from 0.1 mN to 11 mN.

1.2. Data processing

The standard Oliver-Pharr model [10] was used to determine the contact depth and contact area for each loading curve. We assume that the noise of depth and force sensors are Gaussian and independent. Since the contact depth and the contact area are determined from the same dataset, we expect them to be correlated. The contact areas from different measurements are correlated through the modulus of the reference sample

The uncertainties and correlations were determined using the General Uncertainty Framework as described in GUM [11]. The uncertainties of the fitted variables were estimated as the square of the Jacobian of the fitted function [12]. All loading curves were processed using the open-source software NIGET www.nanometrologie.cz/niget. A fixed range of 20 -98 % of the maximum load was used as the fitting interval.

The resulting depth-area data were fitted with a standard Oliver Pharr polynomial [10]

$$A(h) = c_2h^2 + c_1h + c_{1/2}h^{1/2} + c_{1/4}h^{1/4} + c_{1/8}h^{1/8} + c_{1/16}h^{1/16}$$

This form is used only for its mathematical convenience, apart of the first coefficient which is given by the geometry of the Berkovich tip there is no physical meaning to the coefficients. Usually, not more than 5 coefficients are used, the number, however, should be adjusted to the situation. Some parameters can also be fixed. This is often used for the quadratic term, which is known from theory for an ideal tip. For a Berkovich tip, the coefficient is 24.5. The fitting method used does not guarantee the area to be nonnegative. This is of course just a mathematical artifact due to fitting, which is manifested only at minimal depths, and should not affect any measurement.

A weighted least squares algorithm with the composite covariance of both depth and area as weight was used for the fitting procedure [13,14]. The software package ODRPACK [15] was used for this.

In order to assess the uncertainties both the uncertainty estimates based on the Jacobian as given by ODRPACK and a Monte Carlo approach as described in [16] were used. In the Monte Carlo approach a large number approx. 1000 of area curves are generated according to given distributions. Each of these area curves is fitted by a polynomial. A probability distribution of the coefficients is thus obtained. From this the uncertainty of the area can be calculated, and also the uncertainty of the modulus or hardness.

The depth and area data were assumed to have normal distributions with a variance given by the uncertainty determined from the corresponding loading curve. Three different models for the correlation of the data were used: in the first case no correlation was taken into account, in the second case the correlation between each depth-area pair was taken into account, in the third case both the correlation between the depth-area pairs and among the area values was taken into account.

2. RESULTS

A typical area calibration curve for the sharp tip with 50 nm radius is shown in **Figure 1**. The full covariance matrix was used, i.e. both correlations between depth-area couples and between each area-area pair were taken into account.

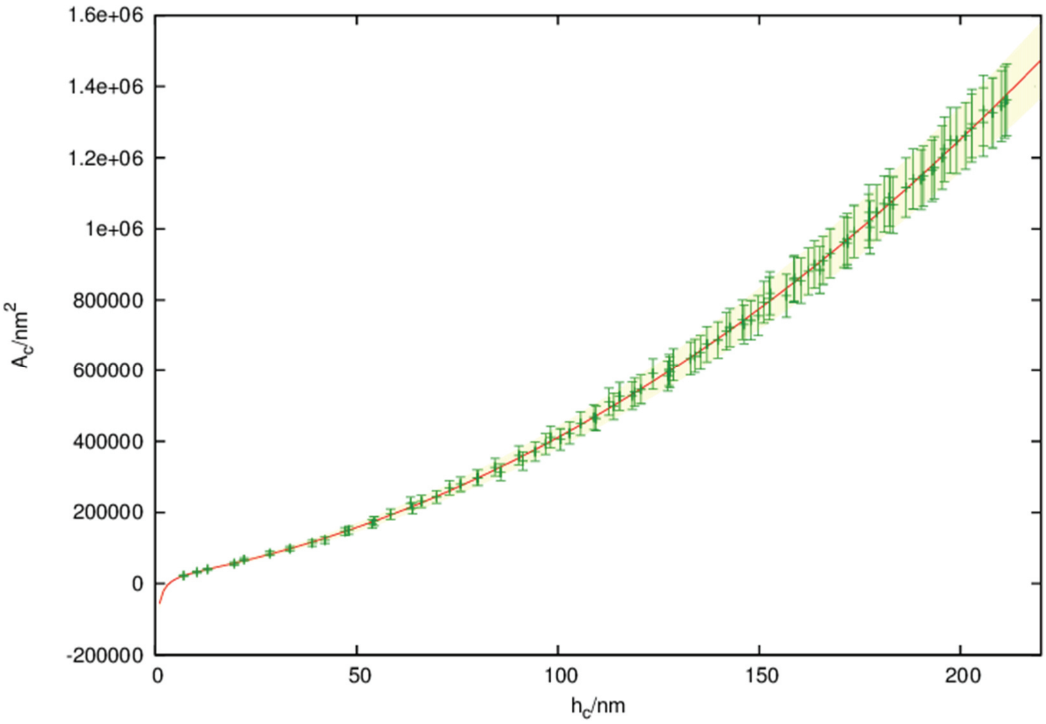

Figure1 Area calibration data (green) and the fitted tip area function (red), the area covered by the uncertainties of the area function is shown in yellow

Note, that the area becomes negative for extremely shallow depths. This is a typical mathematical artifact for these functions. In fact in almost all cases, the area function does not vanish as the contact depth goes to zero but diverges either to positive or negative values. No problems should arise as long as the area function is evaluated within the interval covered by the calibration. Since we set the coefficient for the quadratic term to a fixed value we do not expect any artifacts of this kind for large depth values. The uncertainty of the evaluated area function is approximately the same as the uncertainty of the measured area values.

The number of polynomial coefficients to be fitted should be chosen such that the residual sum of squares is minimized. The residual sum of squares decreases up to n = 5, no significant improvements could be achieved for higher values of n. Visually, the different area curves are barely distinguishable. The mathematical stability tends to deteriorate with growing n. Notably, the behavior outside the fitted interval tends to be worse for higher values of n. The computational demands grow with the number of coefficients as well as the relative area uncertainty growth rate. These are all reasons to keep the number of coefficients as low as possible. In the following the numerical examples use the value n = 5 although the general trends are the same for all values. For the coefficients we obtained uncertainties between 23 - 30 % and correlations around 0.99. In the end, when calculating the total area the uncertainties from the different terms cancel due to the very high correlation. The resulting area uncertainty is much smaller: around 7.5 % for depth values within the interval [50, 250] nm which was used for the calibration and decreasing to 6.5 % for 500 nm to around 2 % for 2000 nm. The uncertainty decreases since the quadratic term which has a fixed coefficient becomes more important.

The difference between the different covariance models manifests itself mainly within the fitted interval. However, it doesn't exceed 0.05 %. Not taking the correlation into account can decrease the relative uncertainty can from 7 % to only 2 %. The most notable trend when including the correlation between the individual area values is that the relative uncertainty of the area becomes less depth dependent. This reflects that ignoring the correlation between area values is over optimistic and leads to an underestimate of the uncertainty.

It must be stressed that the quality of the whole calibration depends dramatically on the low depth data. In **Figure 2** we show a comparison of two calibrations. Two things should be noted. Firstly, with growing order of the polynomial the behavior at very shallow depths values becomes more and more unphysical, as can be seen for values below 18 nm for tip A where only a polynomial with n=3 is usable. Secondly, this can be reduced if data for very shallow depths were included in the calibration procedure as is obvious when compared to tip B, which has a reasonable behavior also for n=4.

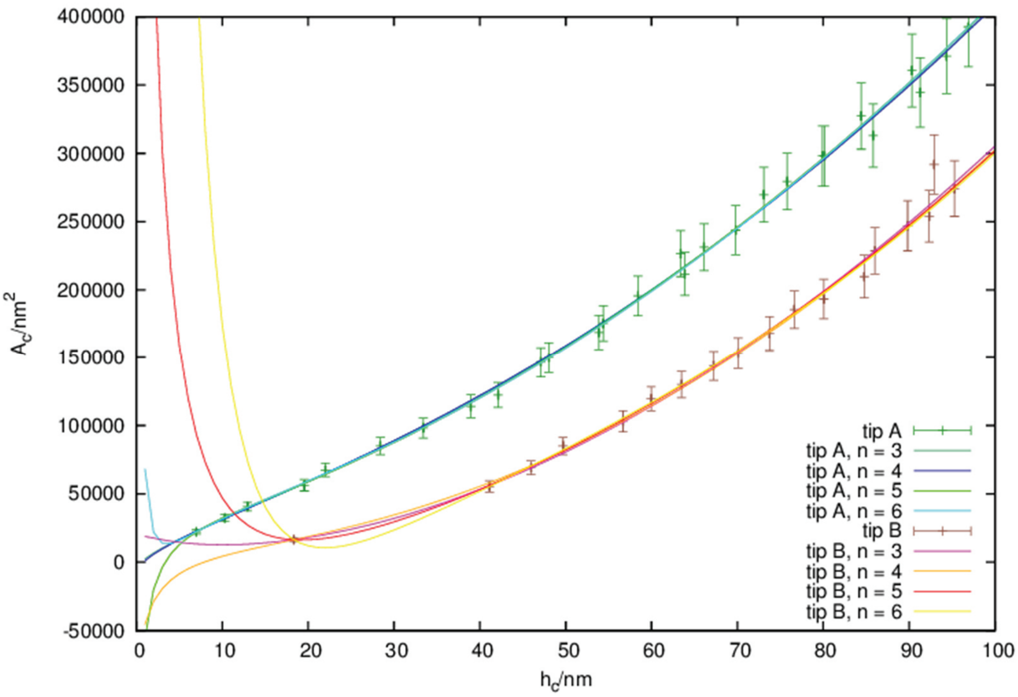

Figure 2 Tip area function for two different tips and different choices of the order of polynomial

An interesting question is the relation between the quality of the fit and the amount of data. We tested this by comparing area calibrations obtained from datasets where a certain number of points where removed. Extrapolated area values are affected the most, especially those for very shallow depths. Within the fitted interval even keeping only every 10th point introduces a difference in area value as low as 2 %. For larger depths, e.g. up to 500 nm the difference grows up to 5 % with growing trend. The relative uncertainty of the area shows a similar behavior.

Usually, we are not interested in the area function itself, but in the mechanical properties such hardness or Young's modulus. Therefore, it is interesting to note the individual contributions to the Young's modulus determined from the same loading curves. The contributions coming from the uncertainty of the slope, uncertainty of the contact depth, uncertainty of the polynomial coefficients and the correlation between the slope and contact depth are shown in **Figure 3**. It can be seen that the area calibration, i.e. the polynomial coefficients are the dominant contribution. Only for very low depths are other contributions significant. The relative uncertainty is approx. 3.7 %, i.e. slightly lower than the value of the reference sample. For the indentation hardness we find a similar behavior, the relative uncertainty is twice as large approx. 7.5 %. On other samples the effect of noise should be comparable and thus the area calibration should form the major contribution to the overall uncertainty. The uncertainty due to the choice of the contact point, however, has not been taken into account in this work. Based on preliminary results we expect the choice of the contact point to significantly affect the results, especially on rough samples where the contact point is more ambiguous.

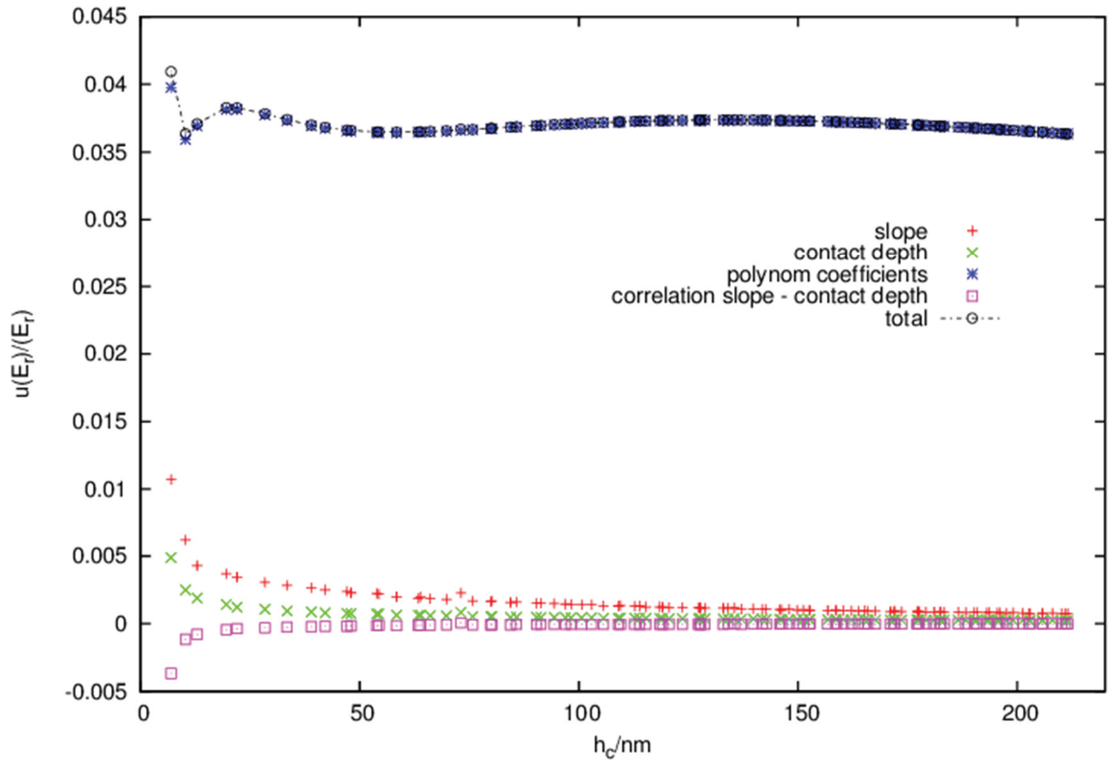

Figure 3 Comparison of the different sources of (relative) uncertainty of the reduced modulus

Obviously, the uncertainty of the Young's modulus of the reference sample which was used for the calibration is crucial. Reference samples made of fused silica which reach an uncertainty below 0.5 % are commercially available. Unfortunately, it is not practical to acquire a set reference samples with a different uncertainty. Therefore, we compared the data to a hypothetical sample with a lower uncertainty of 0.1 %, 0.5 % and 1 %. As expected, the dominance of the contribution from the polynomial coefficients decreases with decreasing uncertainty of the reference sample's modulus. However, even for a fictitious sample with only a 0.1 %

uncertainty in modulus it remains the dominant term, forming around 75 % of the total uncertainty. For realistic reference samples and shallow depths its importance decreases as noise becomes the dominant source of uncertainty.

When using the Monte Carlo method it is necessary to check that the results have converged with respect to the number of iterations. We found that the necessary number of iterations grows with the number of coefficients fitted and the complexity of the covariance model. Without correlation between area values even values as low as 1000 iterations were sufficient. When the correlation was included 50000 iterations were necessary. This corresponds to a computational time of tens of minutes in the worst cases.

3. CONCLUSION

We developed a new tool for the calibration of the nanoindenter area function and the evaluation its uncertainty budget. We used a Monte Carlo method for the determination of uncertainties and included the correlations between each depth-area pair and between the different area values. This model can be easily modified to include other sources of uncertainties. The relative uncertainty was found to vary only little for values which lie within the depth interval in which the calibration was performed with values between 6-7 %. For larger depth values the relative uncertainty diminishes. For smaller depth values, the area function often gives unphysical results. It was found that the uncertainties of the coefficients of the area function are the dominant source of uncertainty for the reduced modulus and hardness. For reference samples with lower uncertainty in elastic modulus this contribution can be decreased. The procedure is only weakly sensitive to the number of data points used for fitting.

ACKNOWLEDGEMENTS

This work is supported by the Czech Science Foundation project GA15-17875S "Lokální mikrostrukturní změny vyvolané statickou a dynamickou indentací nanostrukturovaných a nanolaminovaných povlaků".

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