Determination of Elastic Modulus of Dentin by Small Spherical Diamond Indenters

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A nano-indentation test was applied to determine elastic modulus (E) and hardness (H) of dentin. Three spherical indenters with nominal radii of 20, 5 and 1 μm were used and load/partial-unload cycles were repeated. Each cycle provided E and contact pressure or Meyer's hardness. The plot of contact pressure versus penetration depth was converted into a normalized indentation stress-strain relationship, which was used to select the optimum maximum indentation force for each indenter. The results were compared with those determined by the conventional triangular pyramidal (Berkovich) indenter technique. The comparable E value, irrespective of radius, was 19.5–20.9 GPa and the 1 μm indenter was able to provide E values of peritubular (28.0 GPa) and intertubular dentin (14.9 GPa). The H values generated with the Berkovich indentation technique were comparable to those determined by the spherical indenter technique at indentation strains of 0.9 and 0.5 for the 1 and 5 μm indenters.

Key words: Dentin, Elastic modulus, Spherical indenter

INTRODUCTION

Teeth deform elastically and return to the original configuration under normal function. Determination of elastic modulus (E) of teeth is thus as important as, if not more, that of fracture strength and fracture toughness, for simulation or mathematical modeling of the performance of restorations in teeth. Cutting a tooth to testing dimensions for conventional tests is difficult, especially when the specimen is prepared to evaluate enamel, dentin or cementum separately. Nano-indentation testing, which is similar to micro-indentation hardness testing, offers an alternative way of determining E , as the force-displacement relationship determines the stiffness of a material¹⁾. The test also eliminates possible errors associated with the optical measurement of indentation size, as it uses a precision depth-sensing device and the force-displacement response is used to determine the size rather than after the material has undergone its elastic recovery^{1,2)}. Nano-indentation testing has generally used a triangular pyramidal (Berkovich) indenter tip to determine E and hardness of enamel and dentin^{3–5)}. Use of a spherical indenter with a unique load/partial-unload cycle to determine the elastic-plastic properties of materials has recently been reported^{6,7)}. The objective of the present study was to determine E and hardness (contact

pressure) of dentin with small spherical indenters.

MATERIALS AND METHODS

A clinically sound lower premolar from a female patient aged 18 years was disinfected in Milton's solution for 24 h. The tooth was then stored in deionised water (4°C) for 2 weeks until it was embedded in an epoxy resin (Epofix Kit, Struers, Copenhagen, Denmark). The embedded tooth was cross-sectioned to expose dentin at the buccal cusp using a low speed saw (Isomet, Buehler, Lake Bluff, USA). The newly created surface was first ground with grit #400 and #600 silicon-carbide paper. The following polishing was made using an automatic polishing system with 9 and 1 μm diamond suspensions (DP Suspension, Struers, Copenhagen, Denmark) and then finally with 0.04 μm alumina particle mixed with lubricant (OP-S Suspension, Struers, Copenhagen, Denmark).

The polished specimen was positioned on the traveling stage of an ultra micro-indentation system (UMIS-2000, CSIRO, Sydney, Australia). Areas to be investigated were displayed on a monitor through a microscope with an overall magnification of either 600 or 1200 times. Three diamond cones having nominal spherical tips of 20, 5 and 1 μm radii (Synton BA, Lyss, Switzerland) were used (hereafter 20-, 5- and 1 μm -indenters, respectively). The layout program enabled to select measuring spots and to record them in a file⁶⁾. When the traveling stage was translated to the indenting position, the indenter automatically positioned at the starting spot by the program and then lowered until a contact force of 0.01 mN (0.001 g) or 0.1 mN was detected, depending on the maximum measuring force, less than or more than 50 mN. The specimen was automatically translated to the next spot until all selected spots were measured. A Berkovich indenter (Synton BA, Lyss, Switzerland), a diamond pyramidal tip with an equilateral triangular base of 65.3° face angle to the axis, was included for comparison. The whole system was stabilized in an anti-vibrating chamber and under ambient conditions of 23±1°C and 50±10%RH.

Calculation of elastic modulus (E) and hardness (H)

The spherical indenter technique employed a load/partial-unload cycle method, where the preceding force is unloaded by half at each cycle (Fig. 1A). The cycle is repeated by increasing the indentation force by steps until a predetermined maximum indentation force (MIF) is reached. The elastic and plastic components of indentation were separated, E and mean contact pressure were calculated for each load/partial-unload cycle, and 20 data were produced from one indentation for each property. Composite elastic modulus (E^*) as a function of indentation depth can be determined by the following equation

$$\delta = (9/16)^{1/3} (1/E^*)^{2/3} (1/R)^{1/3} F^{2/3}$$

where δ is total elastic displacement and R is the radius of the spherical indenter (Fig. 2). The elastic modulus of the test material (E_m) was then calculated from the

following equation

$$1/E^* = (1 - \nu_m^2)/E_m + (1 - \nu_i^2)/E_i$$

where ν_m is Poisson's ratio of the material and ν_i and E_i is Poisson's ratio and elastic modulus of the indenter, respectively. Poisson ratios of 0.31 and 0.07 were adopted for dentin and diamond indenter respectively, and elastic modulus of 1000 GPa for indenter^{2,8,9}. Mean contact pressure (P_m), synonymous with Meyer's hardness (H_m), was calculated by

$$P_m(H_m) = F/\pi a^2$$

where F is an applied force and a is the radius of the circle of contact and related by $\delta = a^2/R$ for $\delta \ll R$. By plotting the mean contact pressure as stress against the indentation strain ($\epsilon = a/R$) as strain, a representative stress-strain curve was estimated^{7,10}.

In the general Berkovich technique, force increases in steps without unloading until a predetermined MIF is reached, and then an unloading procedure starts in the reverse way in steps to complete one load/unload cycle (Fig. 1B). Elastic modulus (E_m) of the material at maximum penetration depth in the form of $E_m/(1 - \nu^2)$ is determined using the following relationship

$$E_m/(1 - \nu^2) = 1/2k_2G$$

where ν is Poisson's ratio of the test material, k_2 is a constant and G is the gradient at the maximum force of a power law fit to the upper 60% of the unloading force-displacement data^{1,2}. Hardness (H) as a function of plastic depth can be determined by the following relationship

$$H = F/A$$

Fig. 1 Comparison of the present spherical indenter technique with the conventional pyramidal indenter technique.

A. Multiple load/partial-unload cycles (5 μ m spherical indenter, enamel).

B. A single load/unload cycle (Berkovich indenter, enamel).

Fig. 2 Geometry of the spherical indentation when loaded until an elastic/plastic zone develops, where δ is total elastic displacement, R is the radius of the spherical indenter, a is the radius of contact and F is the applied force.

where F is an applied force and A is the projected area of the indentation related to the plastic penetration (h_p) by $A=f(h_p)$. The relationship, $A=24.5 h_p^2$, was used for a perfect indenter²⁾. The form of A versus h_p was calibrated using amorphous fused silica following the procedure developed by Oliver and Pharr²⁾.

Calibration of spherical indenter shape and selection of optimum MIFs

The three spherical indenters were tested under the multiple load/partial-unload method to determine their actual effective radius with depth for the known E (72 GPa) and Poisson's ratio (0.17) of reference fused silica²⁾. MIFs of 500, 300 and 10 mN were employed for the 20-, 5- and 1 μ m-indenters, respectively. At least 5 indentations were made for each indenter. The results or so-called tip calibration tables were saved as tip calibration files. The precise value of radius at a specific depth was then used to calculate the contact dimensions that subsequently gave contact pressure. A series of trail indentations were then randomly executed under various MIFs with the dentin sample. The results were assessed to assure the penetration depth to be within the range of the tip calibration files for each indenter, and at the same time to accommodate a large contact size to average all components in the dentin structure.

Plots of E or H_m against penetration depth were used to monitor consistent data generation of the spherical indentation test with the three indenters. The normalized stress-strain relationships were also used to finally determine an optimum MIF for each indenter. Plots of hardness and penetration depth from the triangular indentation tests were also used, together with the results from the spherical indentations, to determine an optimum MIF for this indenter.

An array of 10 by 10 indentations was finally made on dentin with the optimum MIF selected for each indenter to determine E and H_m , producing 25 indentations for each indenter. All indentations were made 30 μ m apart on the selected area. Since the spherical indenter technique produced 20 E data from one indentation, the average E value was calculated for each indentation impression. However, the first few data, possibly generated at the depth less than 100 nm, may be influenced by surface

roughness and therefore they were eliminated from the calculation. Average H values were obtained in the same way in the triangular technique. All test results were subjected to a Student's t -test ($\alpha = 0.05$). The analysis was carried out by a statistical program (MINITAB 11.12, Minitab, State College, USA).

RESULTS

Figure 3 shows normalized stress-strain relationships obtained with the three spherical indenters and hardness-penetration depth relationships obtained with the pyramidal Berkovich indenter. The measurements were within the elastic limit with the 20 μm -indenter (A) for all four MIFs. A larger force was needed to create a clear elastic/plastic response with this indenter but the largest force (40 mN) was already likely to generate penetration depth beyond the tip calibration range and the second

Fig. 3 Representative indentation stress-strain curves (A, B, C) and plots of hardness versus penetration depth (D) determined for dentin under various maximum indentation forces (MIFs). The results are from three spherical indenters radii of 20 μm (A), 5 μm (B) and 1 μm (C), and a triangular (Berkovich) indenter (D). The black plots indicate the results which were finally determined as from the optimum MIF.

Table 1 Elastic modulus, Meyer's hardness at maximum depth and contact diameter at maximum force of dentin (n=25)

Indenter	Elastic modulus (GPa)	Meyer's hardness (GPa)	Contact diameter (μm)
Spherical (20 μm)	20.92 $\pm$ 2.12*	0.80 $\pm$ 0.07	8.95 $\pm$ 0.42
Spherical (5 μm)	19.53 $\pm$ 2.05*	1.06 $\pm$ 0.11 [#]	6.02 $\pm$ 0.31
Spherical (1 μm)	20.41 $\pm$ 6.94*	1.27 $\pm$ 0.43	1.04 $\pm$ 0.43
Berkovich*	25.72 $\pm$ 1.82	1.03 $\pm$ 0.11 [#]	5.97 $\pm$ 0.32

Values are mean $\pm$ standard deviation. Statistical evaluation was done with a Student's *t*-test ($\alpha=0.05$). Same affixes denote the values with no significant differences within the column.

*The values are mean $\pm$ standard deviation of elastic modulus at maximum depth, hardness at all indentation depths and the diameter of triangular impression.

Table 2 Elastic modulus, Meyer's hardness at maximum depth and contact diameter at maximum force of dentin structures determined with the 1 μm spherical indenter

Spherical indenter (1 μm)	Elastic modulus (GPa)	Meyer's hardness (GPa)	Contact diameter (μm)
Peritubular (n=8)	27.97 $\pm$ 5.60	1.74 $\pm$ 0.40	0.87 $\pm$ 0.10
Intertubular (n=10)	14.87 $\pm$ 2.39	0.93 $\pm$ 0.10	1.18 $\pm$ 0.06
Combined (n=7)	19.67 $\pm$ 4.43	1.21 $\pm$ 0.24	1.04 $\pm$ 0.11

Values are mean $\pm$ SD. Note: With optical observations, the indentations that landed on peritubular and intertubular dentin were readily distinguished and the indentations which straddled both structures were reported as combined.

largest (30 mN) was chosen as the optimum MIF. Yielding was observed with the 5- and 1 μm -indenters (B and C) and the largest MIF was selected. For the pyramidal indenter, *H* values were most consistent with 30 mN MIF.

Table 1 summarizes mean *E* and *H_m* values, and contact diameter determined with the optimum MIF. In the case of the pyramidal indenter the triangular area of contact was converted to an equivalent circular area of contact to determine the contact indenter. The mean *E* values of dentin obtained from the three spherical indenters were not significantly different. However, the 1 μm -indenter demonstrated a comparatively large standard deviation value. The mean *H_m* values increased with the decrease in indenter size. A relatively large scatter was again observed with the smallest indenter.

Separation of the dentin data with this smallest indenter for evaluation of the peritubular and intertubular dentin was possible by inspection of the visual image produced during the measurement and the results are shown in Table 2. The peritubular dentin had significantly higher *E* ($p<0.0001$) and *H_m* ($p<0.0005$) values than the intertubular dentin.

DISCUSSION

The relationship between H_m and penetration depth can be normalized into a stress-strain curve^{7,10,11}, while the transition of elastic to elastic-plastic behavior is often very difficult to pinpoint. It occurs when the mean contact pressure (H_m) is equal to 1.1 times the yield stress (δ_y) in metallic materials¹². The situation of well-developed yield depends upon the ratio of E/δ_y , as this ratio influences the size of the plastic zone about the impression and the ratio between H_m and δ_y . Optimization of MIFs is therefore an important criterion in order to reach yielding, as H values can be estimated from Meyer's hardness. The contact strain for a sphere that is the equivalent of a Vickers or pyramidal Berkovich indenter is when, $a/R \sim 0.4$ ¹¹. Therefore, at this particular strain, generated only by the 5- and 1 μm -indenters, the H_m of the material obtained with these small spherical indenters should be equivalent to that (H) of the pyramidal indenter. That is, the equivalent H value with the 20 μm -indenter is an underestimation in Table 1.

The H and E were reported to vary across dentin¹³⁻¹⁵ and randomized indentation is therefore important especially when comparative measurement could not be repeated on the same spot. The distances between indentation locations must be sufficient so that residual stress from neighboring indentations is minimal. Since the largest impression size was 8.95 μm (Table 1), a separation space of 30 μm was considered sufficient to avoid this effect.

For the different approaches in the indentation technique, the resultant E values were relatively constant compared to the H values (Table 1). The significantly higher E values with the Berkovich, can be attributed to the influence of creep at the maximum force. Creep is more likely to be reduced when the sample is unloaded to half the preceding force at every step by the rapid partial-unload cycle. Such a measuring technique produces a slightly different influence of the estimated modulus and contact pressure compared with the continuous unloading procedure with the pyramidal indenter. The latter technique leads to an overestimation of the unloading slope and results in higher E . A larger contact area and hence a lower H value is also anticipated as the effect of creep.

A broad range of E values of tooth structures has been reported by various techniques. For the Berkovich technique, 19.3 GPa for intertubular dentin³ has been reported using a linear regression line for the initial 1/3 of the unloading curve and following the method developed by Doerner and Nix¹. The linear regression line would give slightly lower E values of dentin (approximately 6%) than those estimated by the regression line from power law expression used in the present study. The E can also be influenced by location, applied force and choice of the specimen's Poisson's ratio. The difference of E by location (between dentino-enamel junction and the area next to pulp) was reported to be in the range of 2.7-3.5 GPa¹³⁻¹⁵.

The E values of dentin reported in this study were similar and comparable with the values obtained by conventional methods^{16,17}. However, the standard deviation of the E values of dentin obtained from the 1 μm -indenter was approximately 3 times

Fig. 4 Atomic-force microscopic image of dentin shows precise locations of indentation impressions of the $1\mu\text{m}$ indenter.

the values from the other 2 indenters. The result showed the scatter of the E values which was also confirmed by the scatter of the indentation size (Table 1). The spot which the indenter contacted exactly on the peritubular dentin gave a remarkably consistent high E value, 28.0 GPa, while the value obtained from the intertubular dentin was just 14.9 GPa (Table 2). Hence the standard deviation became higher since the values were obtained from different structures rather than the average value of dentin. The impressions of this smallest indenter, while they were visible during the measurement, were difficult to identify with scanning electron microscope and required atomic-force imaging (Fig. 4).

A high value of peritubular dentin, 29.8 GPa⁵⁾, compared to the intertubular dentin, 17.7-21.1 GPa^{3,5)} has been reported with the pyramidal Berkovich indenter. The properties of dentin depended on the individual structure when the small indenter was used. The large indenter would average properties of the overall contact and surrounding area which also include the dentinal tubules.

The present study described the selection of maximum indentation force (MIF) in order to have optimum indenter-material interaction within the elastic-plastic behavior and without any impairment by the resultant fracture. It has shown that small spherical indenters provide a simple reproducible method and generate acceptable values of the mechanical properties of teeth. They enable the superposition of data in the form of contact stress versus contact strain curves that eliminates the depth dependence of contact pressure irrespective of indenter radius. They also provide a better indication of the elastic-plastic behavior of dentin than that determined by the sharp pyramidal (Berkovich) indenter technique. The yield point of dentin obtained from different indenters showed very similar outcomes, and the elastic modulus (E) was 19.5-20.9 GPa for dentin. The values can be influenced by some factors such as

location, applied force and Poisson's ratio adopted in the calculation.

The smaller (5 and $1\mu\text{m}$) radius indenters generated plastic deformation and the contact pressure or equivalent hardness at a contact strain of a/R (~ 0.4 - 0.5) were 0.9 and 1 GPa for dentin, respectively. The small $1\mu\text{m}$ radius indenter enabled the hardness (contact pressure) and E of peritubular and intertubular dentin to be measured and these were 1.7 and 0.9 GPa and 28.0 and 14.9 GPa, respectively. Comparative measurements of E and contact pressure with the pyramidal indenter gave values of 25.7 GPa and 1.0 GPa for dentin, respectively.

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