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Graded viscoelastic behavior of human enamel by nanoindentation

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ABSTRACT

Graded viscoelastic behavior of enamel, including displacement deviation (Δh) between single- and multi-cycling nanoindentations and energy dissipation, is studied using nanoindentation. The imposed load of emerging Δh is about 160 μN, 90 μN and 40 μN, and the maximum value of Δh is 1.86 ± 1.50 nm, 2.21 ± 0.55 nm and 4.15 ± 2.85 nm from the outer to the inner layers of enamel, respectively. In all three enamel sections, the hysteresis loop areas emerge from the third loading cycle, but perform different appearance. Due to the graded structure of enamel, a graded increasing viscoelasticity was found, which is essential to dissipating energy, and preventing cracks from propagating in enamel, therefore retaining the structural integrity of the tooth. The possible mechanism of enamel graded viscoelastic behavior might be the reversible unfolding and refolding of the protein layer.

1. Introduction

Enamel is the hardest tissue in the human body and could sustain physiological damage under mastication stress, due to its graded structure [1] and mechanical properties [2,3]. Enamel is composed of rods measuring 5 μm in diameter and 1-μm-wide rod sheaths. Enamel rods align regularly in the same direction, relatively straight and parallel to each other in the outer and middle enamel, while decussated and curled in different directions near dentin-enamel junction (DEJ) [4]. The mechanical properties of enamel are closely related to its complex graded structure. Hardness and elastic modulus decrease gradually from the enamel surface to the DEJ (from 5.7 GPa to 3.6 GPa for the hardness and from 104 GPa to 70 GPa for the elastic modulus) [2]. The fracture toughness of the outer enamel layer (1.157 ± 0.13 MPa m(1/2)) is lower than that of the inner enamel layer (1.22 ± 0.26 MPa m(1/2)) [5]. Graded mechanical properties contribute to minor deformation of outer enamel under biting force and retain the structural integrity of enamel [3].

The study of the stress-strain response [6] and graded creep of enamel [7] showed the importance of organic sheath in energy dissipation. Enamel has considerable creep and recovery characters, indicating the viscoelastic property of enamel. The viscoelastic properties of teeth are primarily measured using quasi-static single- and multi-cycling indentation [7]. In multi-cycling nanoindentation, the reloading curve may not overlap with the previous unloading curve demonstrating the hysteresis loops, which could be used to study the viscoelastic behavior of materials [8]. The graded enamel microstructure difference might determine its graded viscoelastic behavior, which assists energy dissipation and prevents crack propagation [9,10]. Therefore, it is necessary to investigate the graded viscoelastic properties of enamel using single- and multi-cycling nanoindentations.

2. Materials and methods

The study was approved by the Research Ethics Committee, Sichuan University, China. Six noncarious human third molars from 19 to 23-year-old patients were extracted under informed consent. The crowns were embedded in epoxy resin (EpoFix, Struers, Denmark) and mounted with only one cusp exposed. The exposed cusp was grounded flat with 800, 1200, 2400 and 4000 grit abrasive papers (Struers, Copenhagen, Denmark) under water irrigation, and polished with 3 μm alumina suspension slurry for 5 min and with 0.04 μm OP-Nondry for 10 min on a polishing machine (Struers, Copenhagen, Denmark). The tooth specimens were ultrasonically cleaned for 15 s Tests were conducted in three regions: the outer enamel, at a distance within 100 μm from the occlusal surface; the middle enamel, the midpoint area between the occlusal surface and DEJ; and the inner enamel, at a distance within 100 μm from the DEJ [3] (Fig. 1a and b). Nanoindentations were applied using Hystron triboindentator (Hystron Inc., Minneapolis, USA) equipped with a Berkovich diamond tip (radius ~150 nm). The outer enamel samples were tested first, and then ground to the middle and inner enamel, repeating the same test

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[1] Represents the same contribution to the paper.

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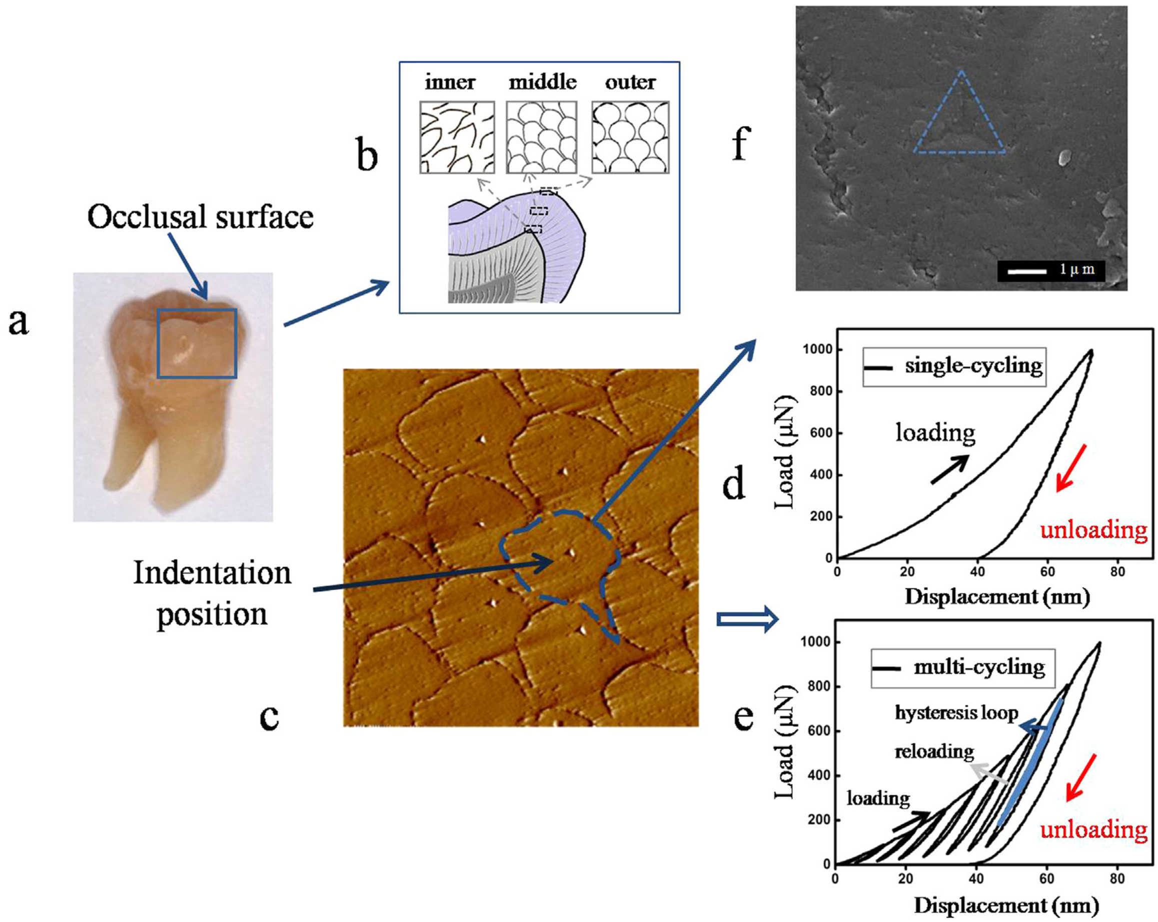

Fig. 1. An illustration of test procedure. (a) Noncarious human third molar; (b) three regions for schematic drawing of test in a buccal cusp; (c) typical in-situ SPM image of nanoindentation; (d) the typical load-displacement curves of single- and (e) multi-cycling nanoindentations; (f) a SEM image of nanoindentation.

procedure respectively. The indenter was calibrated by conducting the single-cycling nanoindentation on fused quartz sample. A maximum load of 1000μN was selected for single-cycle nanoindentation and the loading/unloading rate of 400 μN/s (Fig. 1d) [8]. Six samples of each specimen were obtained and 10 indentations were performed on each sample. Multi-cycling nanoindentation tests were conducted with an incremental load of 10 cycles [8]. During each cycle, the samples were loaded to the cycle's peak load, partially unloaded by 90% of the load, and immediately reloaded to the next peak load (Fig. 1e). The thermal drift rate was less than 0.2 nm/s. The Δh and the hysteresis loop area were analyzed using a one-way Analysis of Variance (ANOVA) with significant differences identified at $\alpha = 0.05$.

3. Results and discussion

From the outer to the inner enamel, the emerging of Δh shifted from the fifth to the third cycle, the corresponding imposed load (as shown in black box in Fig. 2(a–c) was about 160 μN, 90 μN and 40 μN, respectively. The Δh of the outer enamel was similar to that of enamel occlusal surface investigated by Jia and Xuan [8]. At the beginning of single- and multi-cycle nanoindentations (when the load is less than 200 μN), enamel displayed an elastic response and the curves of both nanoindentations overlapped without Δh (Fig. 2a–c). The contact stress (equivalent to nanohardness calculated by computer automatically) of Δh emergence is about 1.9 GPa, 0.54 GPa and 0.063 GPa for 160 μN, 90 μN and 40 μN, respectively. Enamel deformation of HAp would occur when stress reaches 56 GPa [11], thus under the small imposed load (< 200 μN), deformation was limited to the protein component. The shear strain in protein layer between HAp crystals was calculated using the equation $\gamma_p = ((h_p + h_m)/h_p)\tan\theta$ according to He's theory, where θ represents the equivalent angle of Berkovich indenter 19.7°, the shear stress $\tau_p = G_p\gamma_p$ [11]. The organic content increases with the depth of enamel layers [2] (Fig. 3(c–e)). The h_m/h_p is about 50, 26 and 12.5 for the outer, middle and inner enamel layers, respectively [8,11], and the shear stress was 1.82 GPa, 0.966 GPa and 0.483 GPa, respectively. The graded shear stress might contribute to the decreasing graded force of emerging Δh.

Δh increased with the increasing cycle (Fig. 2d), with a maximum value of 1.86 ± 1.50 nm, 2.21 ± 0.55 nm and 4.15 ± 2.85 nm from the outer to the inner layers of enamel, respectively, suggesting gradually increasing graded viscoelasticity similar to the graded increase in creep response [2]. The increase in graded Δh may relate to the differential response of enamel protein layer under single- and multi-cycle nanoindentations. Enamel exhibited elastic and plastic response with single-cycle nanoindentation but elastic, viscoelastic and plastic response with multi-cycle nanoindentation [12]. The protein layer reacted to the response by the reversibly unfolding under loading and refolding upon un-