Improving Fatigue Strength of Alumina through Surface Grading

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Abstract:

Porcelain veneered alumina crown restorations often fail from bulk fracture resulting from radial cracks that initiate at the cementation surface with repeated flexure of the stiffer crown layers on the soft dentin support. We hypothesize that bulk fracture may be substantially mitigated by grading the elastic modulus at the crown surfaces. In this study, graded structures were fabricated by infiltrating glass into *dense* alumina plates, resulting in a diminished modulus at the surface layers. The plates were then bonded to polycarbonate substrates and subjected to fatigue loading in water. Tests were terminated when fracture occurred at the cementation tensile surface or at the fatigue endurance limit (1 million cycles). Infiltrated specimens showed a significant increase in fatigue fracture loads over non-infiltrated controls. Our results indicate that controlled elastic gradients at the surface could be highly beneficial in the design of fracture resistant alumina crowns.

KEY WORDS

Alumina crowns, alumina–glass layers, modulus gradient, fatigue loading, bulk fracture

INTRODUCTION

As a restorative material, alumina has better aesthetic values than zirconia, i.e. better translucency coupled with a more natural tooth-like shade (Heffernan et al., 2002). In addition, alumina has a higher thermal diffusivity (1.0×10^{-5} m^2 s^{-1}) than that of zirconia (7.4×10^{-7} m^2 s^{-1}), which allows alumina to be more effective in dissipating heat (Swain, 2009). Zirconia, with a lower thermal diffusivity, is more prone to generating high tensile residual stresses in the porcelain veneer, leading to premature veneer chipping or fracture (Swain, 2009). However, alumina has a relatively low flexural strength, which makes alumina-based restorations susceptible to bulk fracture (Hermann et al., 2006; Kelly, 1997; Kokubo et al., 2009; Oden et al., 1998). One way to overcome this problem is to strengthen alumina by grading the material composition with a lower modulus at the tensile surfaces (Huang et al., 2007; Zhang and Ma, 2009).

Previously, we have demonstrated the feasibility of such a process, by infiltrating the surfaces of *pre-sintered* zirconia templates with a low-modulus glass of similar coefficient of thermal expansion (CTE) (Zhang and Kim, 2009). Such a gradation is able to diminish the tensile stress intensity at the plate surfaces (Jitcharoen et al., 1998; Zhang and Ma, 2009), rendering the structure less susceptible to flexural (Zhang et al., 2010) and contact (Kim et al., 2010; Zhang and Kim, 2010) damage. Here, we infiltrate a *fully sintered* and more aesthetically pleasing alumina system (relative to zirconia) with a low-modulus glass of similar CTE, creating a graded alumina-glass structure. We provide a quantitative analysis of the effects of a graded modulus on the fatigue load-bearing capacity of alumina-glass crown-like layer structures and demonstrate that a graded alumina-glass surface layer can effectively improve the fatigue strength of alumina. This novel alumina-glass material with enhanced flexural strength, aesthetics, and cementation properties may spark renewed interest in alumina-based dental restorations.

MATERIALS & METHODS

The alumina material used to produce graded structures was a dense, 99.5% pure, fine-grain alumina (AD995, CoorsTek, Golden, CO). This material has a flexural strength of 572 MPa and a CTE of 8.2 x 10^{-6} °C^{-1} (Zhang and Lawn, 2004). These properties are comparable to commercial dental alumina. A slurry containing a silicate glass composition with matching CTE was applied to the top and bottom surfaces of sintered alumina plates. The main composition (>1 wt.%) of the infiltrating glass is: SiO_2 (71.6%), Al_2O_3 (10.5%), K_2O (8.0%), Na_2O (6.9%) and CaO (1.3%). The coated plates were then heated to 1550°C for 2 hr, producing an infiltrated glass/alumina/glass (GAG) structure. Heating and cooling rates were 600°C/hr. After cooling, the surface excess glass was removed and specimens were polished to 0.5 µm finish at both the top and bottom surfaces. Twenty flat specimens each for GAG and monolithic alumina control were prepared (23 mm in diameter and 1.45 mm thick). Three specimens from each group were bisected along a diameter-line-segment; these cross-sections were polished to 0.5 µm finish. The polished cross-sections of half of the bisected plates were indented with a Berkovich diamond tip using instrumented indentation device (Nanoindenter XP, MTS Systems Corp., Oakridge, TN) with a maximum force of 500 mN. The indenter tip was loaded and unloaded with a rate of 10 mN/s. The reduced modulus was determined by the Oliver-Pharr approach (Oliver and Pharr, 1992) from the indentation curve. An array of indents was made from the specimen surface to the interior with a step size of 20 µm. The Young's modulus of the specimen was computed using the measured reduced modulus[1]. The other half of the bisected plates was carbon coated on the polished section for microstructural analysis using scanning electron microscopy (SEM, Hitachi 3500N, Japan).

Seventeen plates from each group were bonded to polycarbonate bases using epoxy, simulating ceramic restorations cemented to dentin structures. Hertzian indentation fatigue tests were performed on this ceramic/polycarbonate bilayer structure with a spherical tungsten carbide (WC) indenter of radius r = 3.2 mm in water using a mouth-motion simulator (Elf 3300, EnduraTEC Division of Bose, Minnetonka, MN) at 2 Hz. The specimen was mounted on an inclined block (θ = 30°, Fig. 1). Load was applied in the vertical direction, but the loading consisted of a contact–load–slide–liftoff sequence—the indenter contacting the specimen, loading to a maximum while sliding down the surface to create a wear facet, unloading, and lifting off from the specimen surface. Failure of the ceramic layer on a compliant substrate was defined when cementation (intaglio) surface radial cracks popped-in (i.e. bulk fracture). Damage sustained in fatigued specimens was examined using a 3D polarized specular reflection microscope (Edge R400, Micro Science Technologies, Marina Del Rey, CA).

The current fatigue tests were designed to determine the number of cycles to failure n_F for a range of prescribed fatigue loads 200 N to 1000 N. A minimum of three specimens were tested for each prescribed fatigue load. Damage maps were constructed for graded GAG and monolithic alumina on compliant substructures. Two-way analysis of variance (ANOVA) was used to evaluate the effect of test load and material (GAG or alumina), and their interaction on the number of cycles to failure. Cycles to failure were $\log_{10}$ transformed before analysis. Statistical tests that yielded p-levels <0.05 were considered significant.

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[1] The reduced modulus E_r is described by the relationship $1/E_r = (1-v_i^2)/E_i + (1-v_s^2)/E_s$ with E and v being the Young's modulus and Poisson's ration of the indenter (i) or tested sample (s), respectively. For a diamond indenter tip, E_i is 1140 GPa and v_i is 0.07. For alumina, glass, and graded alumina-glass materials, v_s is 0.22.

Fatigued specimens were subjected to post-mortem damage examination using optical microscopy (Kim et al., 2007a). Selected specimens were subjected to surface topography and roughness measurements across the wear crater using a 3D optical profilometer (Microphase DMC 100, The Digital Wavefront Company, Seattle WA, USA).

RESULTS

SEM analysis revealed a relatively high glass content at the graded alumina-glass surface (Fig. 2a). The glass content gradually decreased as the distance from the surface increased; eventually the graded alumina-glass layer merged with a highly crystalline alumina core (Fig. 2a). Nanoindentations confirmed a relatively low Young's modulus at the hybrid alumina-glass surface, which rose rather quickly to the bulk modulus of alumina as the distance from the surface increased (Fig. 2b). The thickness of the graded layer, according to nanoindentation, was approximately 100 µm.

Typical damage patterns of ceramic/polycarbonate bilayers after subjection to sliding-contact fatigue are presented in the optical micrographs (Fig. 3). Cementation view revealed a classical flexural radial crack pattern with multiple arms emanating from the fracture origin beneath the contact area (Fig. 3b). Occlusal view revealed that only one of the radial crack arms has extended to the top surface (Fig. 3a). In addition, a typical teardrop shaped wear track induced from the sliding contact (sliding direct from left to right of the images) was also evident from the occlusal surface (Fig. 3a). The example shown here was an alumina/polycarbonate bilayer specimen after sliding-contact fatigue at 608 N for 108 cycles. However, such fracture patterns were observed in all specimens, except for those that survived (without radial fracture) after the endurance limit (1 million loading cycles) was reached.

Sliding-contact damage sustained on occlusal surfaces of homogeneous alumina and graded GAG following 1 million cyclic loading at 480 N and 682 N, respectively, are shown for comparison (Figs. 3c and d). Remarkably, only a smooth wear crater formed in both materials after prolonged sliding-contact fatigue at such high loads. Neither cracks nor material spalling was observed.

To simulate nominal biting force (Kelly, 1999), we also conducted sliding-contact fatigue at 200 N for 1 million cycles on both alumina and GAG (Figs. 3e and f). Again, only smooth wear craters without cracks or spalling were observed in both materials. 3D optical profilometer line mapping across the center of the wear craters, along the sliding direction (indicated by white dashed lines in Figs. 3e and f) revealed that the depth of wear craters, relative to the surrounding virgin surface, was 9 ± 1 μm (n = 3) and 7 ± 1 μm (n = 3) for alumina and GAG, respectively (see pronounced peak-valley curves in Figs. 3e and f). The characteristic surface topography of the wear craters also became apparent. Material pile-up was observed at the trailing edge as well as the leading edge of the sliding indenter. The deepest valley in the wear crater corresponded to the location where maximum load occurred in each sliding cycle. Interestingly, regardless of the relatively large peak-valley topography, the average surface roughness, S_a (the arithmetic average of the absolute deviation from the mean line over a sampling area), was small throughout the wear craters in both materials, being 0.85 and 0.89 μm for alumina and GAG, respectively.

Damage maps (load-cycles-type of failure) for cementation bulk fracture in ceramic/polycarbonate bilayers due to sliding-contact fatigue loading are reported (Fig. 4). It is apparent that more cycles, by several orders of magnitude, were required to initiate the flexural radial cracks in GAG/polycarbonate bilayers compared to the alumina/polycarbonate bilayers.

ANOVA revealed a longer lifetime for graded GAG/polycarbonate than alumina/polycarbonate specimens [F(1,24) = 68.2, $p < 0.001$]. At the mean load of approximately 710 N, the mean lifetime of the graded GAG/polycarbonate specimen was estimated to be almost 400,000 cycles while that of the alumina/polycarbonate specimen was estimated to be less than 100 cycles. In addition, there was a statistically similar slope relating load to lifetime for the two materials [F(1,24) = 0.3, $p = 0.59$], estimated to be a reduction, in either material, of approximately 2 $\log_{10}$ cycles per 100 N increase. These data suggest that for a given load, graded GAG specimens will have a much longer fatigue lifespan than pure alumina specimens; similarly, for a given lifetime, graded GAG will tolerate a much greater load than homogeneous alumina specimens.

DISCUSSION

This study investigated the resistance to flexural radial fracture of functionally graded GAG materials relative to homogeneous alumina under fatigue sliding-contact using a hard sphere on inclined ($\theta = 30°$) ceramic/polycarbonate bilayers, analogous to tooth contact during mastication. Optical microscopy identified the damage sustained in alumina-based ceramic/polycarbonate systems: flexural radial crack initiated at the intaglio surface and propagated sideward and upward, resulting in catastrophic bulk fracture. These findings are consistent with clinical reports on alumina-based restorations where flexure-induced bulk fracture initiated from the cementation surface constitutes a major proportion of failures (Kokubo et al., 2009; Oden et al., 1998). Thus, despite the advantage of better translucency and a more natural tooth-like shade, alumina-based restorations have been replaced by the stronger zirconia. Here we demonstrate an effective way to improve the flexural strength of alumina by surface grading. Our fatigue study has shown that graded GAG exhibited much better resistance to long-term flexural fracture compared to homogeneous alumina. Composite beam theory predicts that the low modulus surface in graded GAG is able to effectively dissipate the maximum flexural stress and redistribute it to the bulk of the material, away from the strength limiting surface flaws (Zhang and Ma, 2009). In addition, the glass infiltrates the surface flaws of alumina, making GAG more immune to surface flaw population relative to homogeneous alumina.

Clinical research and practice have also revealed that veneer chipping or fracture induced by occlusal surface cone fracture are often observed in all-ceramic restorations (Etman and Woolford, 2010), especially in strong ceramic framework supported systems, in part due to the rare occurrence of bulk fracture. The question then arises as to what extent a glass-rich graded surface layer can withstand prolonged occlusion. Our results show that a high 682 N sliding load for 1 million cycles using a hard WC indenter only managed to leave a smooth wear track at the contact surface, indicating that GAG surfaces exhibit excellent resistance to occlusal-like sliding-contact damage. Contact mechanics analysis has shown that the low modulus glassy surface was able to reduce the maximum tensile stress in the wake of the sliding indenter and transfer it into the bulk, suppressing the formation of the sliding-induced partial cone cracks (Giannakopoulos and Suresh, 1997; Kim et al., 2010; Kim et al., 2007b; Kim et al., 2008; Suresh et al., 1999; Suresh, 2001; Zhang and Kim, 2010).

We present a new concept that improves the resistance of alumina to fatigue flexural damage while maintaining excellent wear resistance to occlusal contact damage by utilizing an alumina-glass functionally graded material. Since glass is used to infiltrate the accessible surfaces of alumina, a graded alumina-glass material possesses improved aesthetics relative to alumina. The low hardness and modulus glassy surface can prevent excessive wear of the opposing dentition. The glass-rich intaglio surface offers a potential for acid etching and silanization, thus facilitating a resin cement bond (Zhang and Kim, 2009). With a simple staining technique, the graded alumina-glass material offers an aesthetic option for damage resistant, monolithic crowns and potentially fixed partial dentures in the posterior regions.

Finally, we acknowledge that in the current GAG specimens, we gently polished away the surface residual glass layer to create a smooth graded alumina-glass surface, facilitating direct comparison with the polished homogeneous alumina surface. Our on-going fatigue studies of graded zirconia-glass structures with a thin (~20 μm) residual glass layer reveal excellent resistance to long-term sliding-contact fatigue damage. Neither spalling nor cracks have been observed on the residual glass/graded surfaces following 200 N sliding-contact for 1 million cycles in water. As all our tests were conducted on flat specimens, fatigue testing on anatomically correct crowns is next on our agenda.

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DISCLAIMER

Any mention of commercial products within this paper is for information only; it does not imply recommendation or endorsement by NIST.

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FIGURE CAPTIONS

1. Schematic of molar tooth occlusal contact. (a) Tooth-tooth contact. (b) Experimental setup for sliding-contact fatigue of ceramic restoration layer on dentin-like compliant substrate (bilayer) with an inclination angle θ. Showing cementation surface radial cracks (R). Note (b) represents the area highlighted by the gray dashed line box in (a).

2. Microstructural and mechanical properties of graded glass/alumina/glass (GAG) structures. (a) SEM image of the surface of a graded alumina-glass layer (left), 20 µm into the graded zone (center), and an alumina core (right). Note: G and A represent glass and alumina phases, respectively. (b) The Young's modulus profile across section of infiltrated GAG, plotted as function of distance z from outer surface. Note gradation of values within graded zone (I), and constant value within bulk (II). Different symbols represent separate tests on three GAG specimens.

3. Post-mortem optical micrographs showing top (occlusal) and bottom (cementation) views of ceramic/polycarbonate bilayers subjected to sliding-contact fatigue with tungsten carbide sphere of radius $r = 3.2$ mm, in water. (a) Occlusal and (b) cementation views of alumina/polycarbonate bilayer subjected to sliding-contact fatigue at $\boldsymbol{P} = 608$ N, $n_F = 108$ cycles. (c) and (d) are occlusal views of sliding-contact damage sustained in homogeneous alumina and graded GAG following 1 million cyclic loading at 480 N and 682 N, respectively. (e) and (f) are occlusal views of alumina and GAG following 1 million sliding-contact cycles at 200 N. The sliding direction is from left to right of the

images. In (e) and (f), white dashed lines indicate the position of line mapping using a profilometer. The peak-valley curves represent the surface topographic profile.

4. Plot of number of cycles to failure, n_F, as a function of maximum load, ***P***, for cementation flexural radial fractures in GAG/polycarbonate (gray filled circles) and alumina/polycarbonate (open triangles) bilayers following sliding-contact fatigue. The fatigue loads ***P*** investigated are 682, 685, 701, 707, 773, 774, 793, 802, 808, 814, 879, 964, 979, 982 N for GAG/polycarbonate bilayers, and 455, 460, 480, 510, 539, 588, 608, 634, 652, 660, 669, 777, 791, 797 N for alumina/polycarbonate bilayers. Note: arrows represent specimens that survived 1 million sliding-contact cycles (the fatigue endurance limit) at prescribed loads without radial fracture.

FIGURES

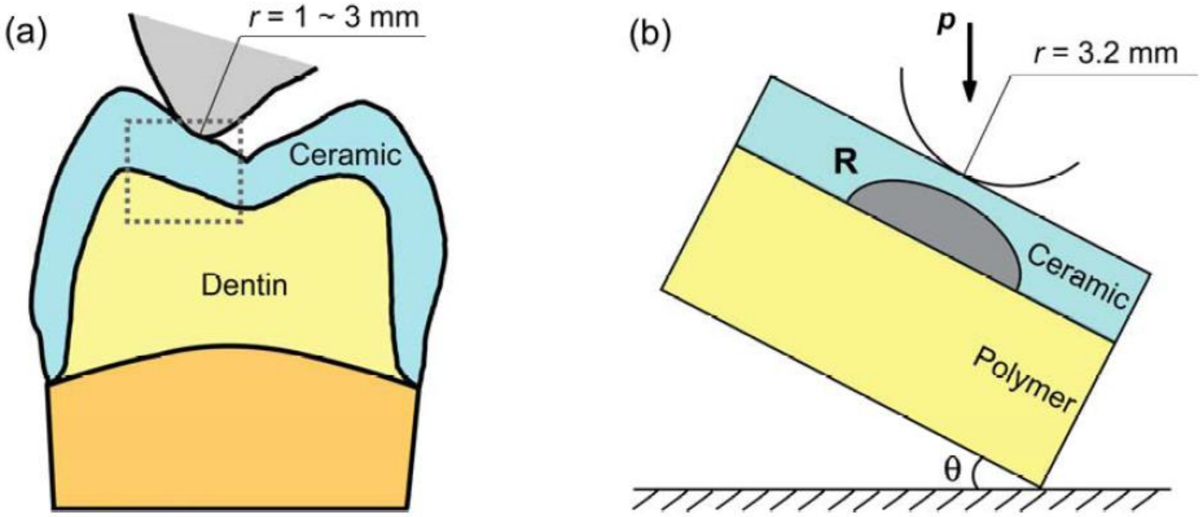
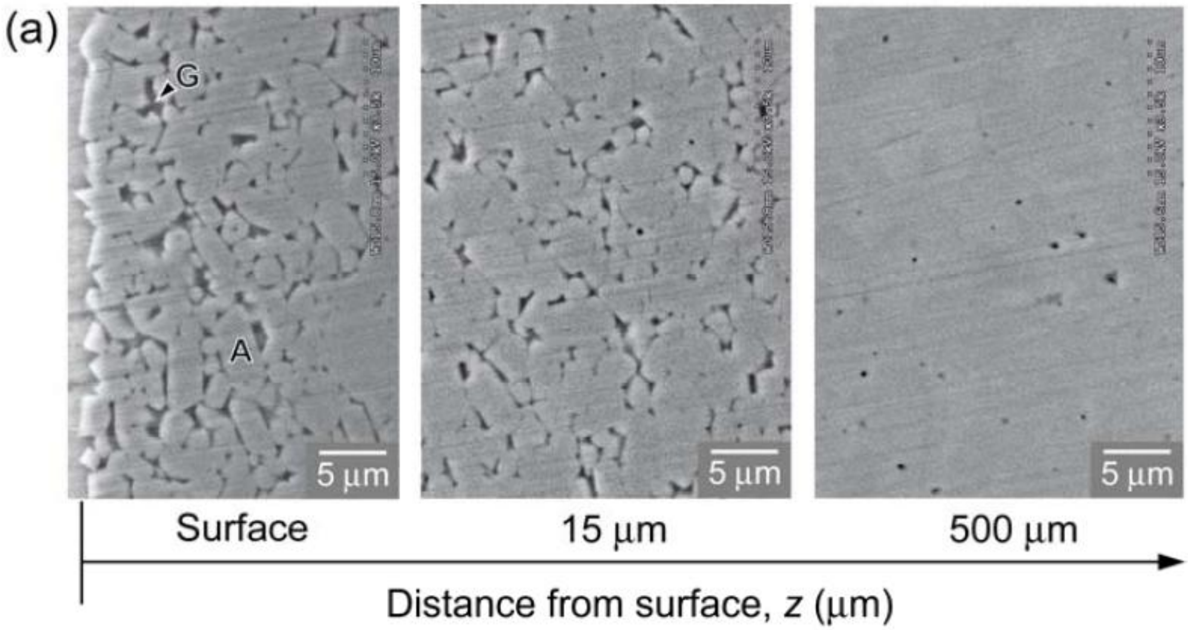


Figure 1


(a)
G
A
5 µm
5 µm
5 µm
Surface
15 µm
500 µm
Distance from surface, z (µm)

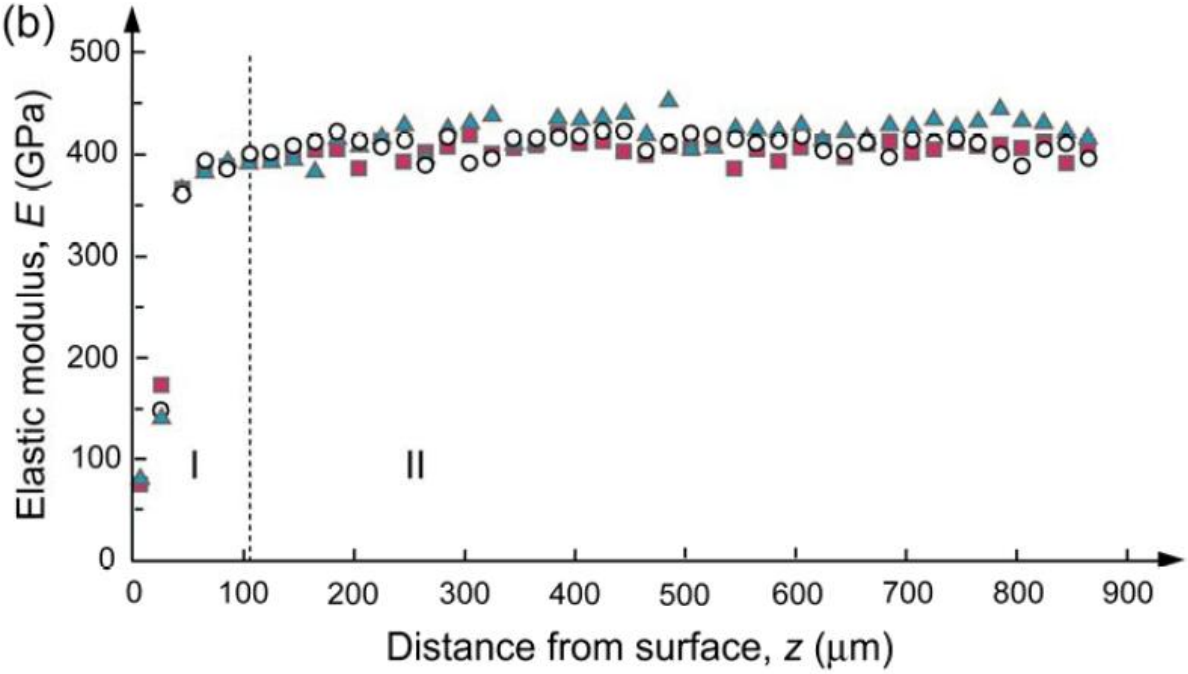

(b)
Elastic modulus, E (GPa)
500
400
300
200
100
0
I
II
0
100
200
300
400
500
600
700
800
900
Distance from surface, z (µm)

Figure 2

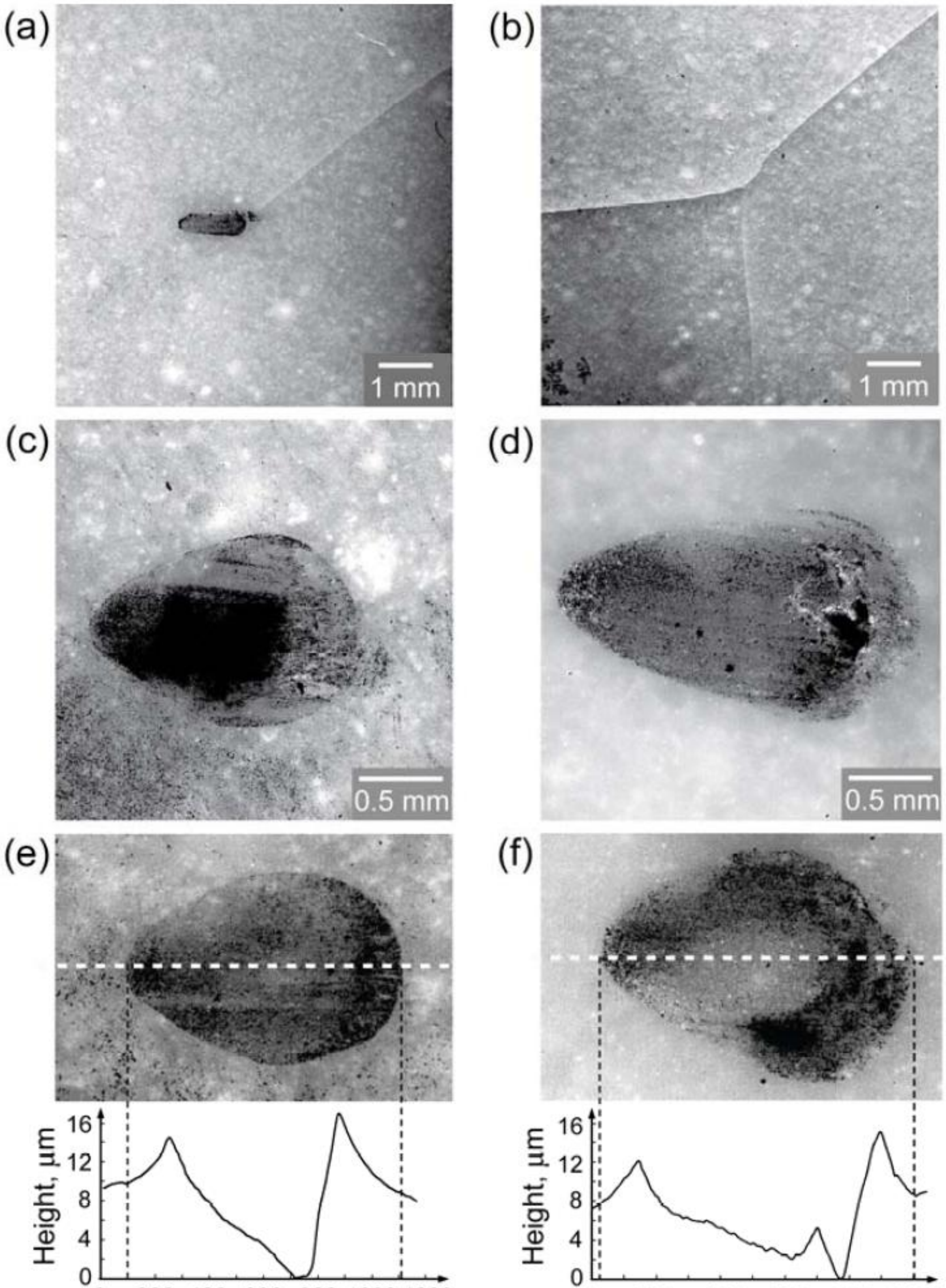

(a)
1 mm
(b)
1 mm
(c)
0.5 mm
(d)
0.5 mm
(e)
(f)
Height, µm
16
12
8
4
0
0 200 400 600 800 1000 1200
Scan distance, µm
Height, µm
16
12
8
4
0
0 200 400 600 800 1000
Scan distance, µm

Figure 3