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Self-glazed zirconia reducing the wear to tooth enamel

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ABSTRACT

The wear behaviors of a newly developed grade of self-glazed zirconia against the enamel of freshly extracted teeth were investigated under simulated oral stresses and chemical environment. It was revealed that an inherently formed enamel-like surface on self-glazed zirconia that is very smooth on micrometer scale yet with nanoscale roughness has almost the same frictional coefficient against tooth enamel as the well-polished zirconia surface. The wear scars observed on the worn surface of enamel against self-glazed zirconia and well-polished zirconia surface revealed that in both cases fatigue wear is the dominating wear mechanism. It was concluded that the friction and wear performances of both the well-polished and self-glazed zirconia ceramics against natural enamel were very similar, which bears a very strong implication for the clinical safe use of the full contour zirconia restorations, yet the self-glazed zirconia provides sufficiently improved aesthetic appearance that ensures its potential for direct clinical uses.

1. Introduction

Veneered zirconia restorations exhibited high failure rates due to veneering porcelain fractures [1–3]. As an alternative approach for overcoming this problem encountered often in clinical practice, milling of a pre-sintered blank to full-contour has been established for zirconia monolithic restorations [4,5]. In this way the common use of a thick ceramic veneer layer can be avoided, but a thin stain and glaze coating layer still need to be applied, otherwise the surface has to be well polished before use in order to reduce the wear of a rough surface to the enamel of the opposite teeth. Although these full-contour monolithic restorations are ideally suited to cases where only minimal space is available, the risk of clinical failures by the progressive occlusal height reductions initiated by the friction and wear between restorations and opposite natural teeth under mastication cycles in oral environment has to be considered [6–8].

In order to eliminate as many as possible manual operation steps in traditional dental labs such as sandblasting, polishing, veneering, staining and glazing, it is desirable to prepare strong, reliable and nature looking dental restorations completely by digital technology. By applying a precision wet-chemistry technology in which local plastic deformation is facilitated to pack nano-sized grains on the surface of dental restorations a new grade of self-glazed zirconia ceramics has been developed recently. The inherent formation of an enamel-like surface ensures no further need of veneering or glazing thereby avoiding breakable interfaces. This appears for the first time as a real monolithic restoration that can be directly applied with minimal clinical adjustment.

Zirconia restorations used to be blamed to increase the risk of wear to the opposite teeth due to their higher hardness compared to that of the silicate based veneer porcelains and the lithium disilicate glass-ceramics. Recent studies have, however, confidently demonstrated that the wear of a dental restoration to the opposite tooth mainly depends on the surface roughness of the former [6,8]. Different from the traditional blank-machined zirconia, which appears like a rough sand parchment, the self-glazed zirconia restorations have very smooth surfaces that may protect the opposite natural teeth from excessive wear. It is the aim of this work to investigate the wear behaviors of the self-glazed zirconia against natural enamel by in vitro friction tests under simulated oral stresses and chemical environment using zirconia ceramics with well-polished surface as a reference.

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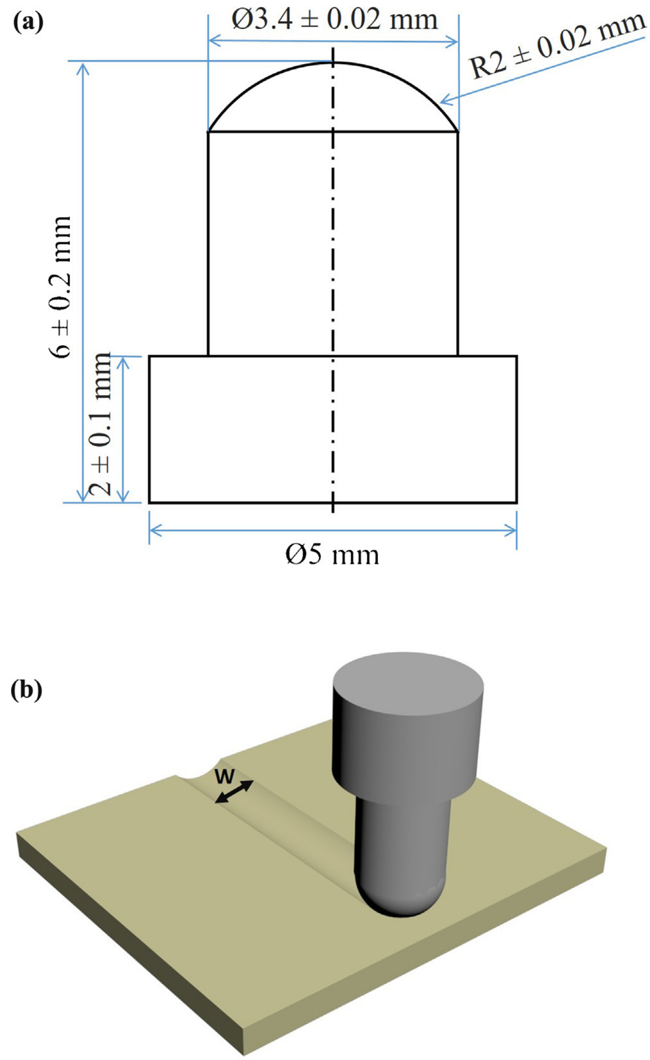

Fig. 1. A 2D (a) draw and a 3D (b) illustration showing the size and geometry of the cylindrical test sample and the setup of a wear pair in reciprocating ball on plat pattern style.

2. Experimental

2.1. Sample preparation

Two kinds of 3Y-TZP samples (ZrO_2 doped with 3 mol% Y_2O_3) were prepared for test, being the self-glazed zirconia with a smooth surface and the zirconia with a polished surface down to 1 μm finishing, with the latter one as a reference. As a 2D draw and a 3D illustration shown in Fig. 1 demonstrate, the cylindrical test samples were fabricated to have a hemisphere head at one end, with the radius of 2 mm, to simulate the dimension of natural tooth cusp with the radius of 2–4 mm. The hemispherical geometry was produced by CAD/CAM milling of a green body formed by a wet process in case of self-glazed zirconia sample and of a pre-sintered blank in case of zirconia with milled rough surface and polished surfaces, respectively. All the milled samples were pressure-less sintered at 1450 °C for 2 h in air atmosphere to achieve a relative density above 99.9%. After sintering the self-glazed zirconia sample was used directly for wear test without applying any additional post-sintering treatment, whereas one zirconia sample milled from pre-sintered blank was polished by diamond suspension down to 1 μm finishing before wear test. The smooth surface on self-glazed zirconia was inherently formed by local plastic deformation

Table 1
The chemical composition of artificial saliva.

NaCl(g)	0.4
KCl(g)	0.4
$CaCl_2{\cdot}2H_2O$(g)	0.795
$NaH_2PO_4{\cdot}2H_2O$(g)	0.78
$Na_2S{\cdot}9H_2O$(g)	0.005
Urea(g)	1
Distilled (ml)	1000

introduced during a precision wet-chemistry process that assured dense packing of nano-sized grains on the surface of dental restorations. The self-glazed zirconia sample reveled a finer grain structure than the blank-machined zirconia samples, being 100–250 nm versus 200–600 nm of the grain size for the former and later, respectively. In both cases, a newly developed 3Y-TZP powder is used as the precursor. The powder is a semi-product with the potential of being commercialized in the near future and, thus, the producer will not be disclosed at this point. The powder consists of 100–150 nm spherically-shaped particles. The single, secondary particle is composed of loosely aggregated primary nanoscaled crystallites of 27 nm in average size. The specific surface area of the powder is 18.2 m^2/g.

A premolar without obvious wear scar, extracted for orthodontic demand, was collected from a 13-year aged young male, and preserved in distilled water at 4 °C for sample preparation. The tooth was embedded in epoxy resin (SY-668-3, SenMeiYa, China) after pulpless, with the enamel of buccal surface (at least 5 × 5 mm area) exposed. The enamel surface was then grounded by carborundum sand papers in water, gradually from 180 to 1500 mesh, and polished by 1 μm diamond sand papers. The final dimension of the epoxy resin block was 30 mm in diameter and 10 mm in thickness. The sample was stored in distilled water during the whole test process.

2.2. Friction and wear tests

The wear pairs of the plate of natural tooth and hemispherical zirconia with two types of surfaces were tested by a micro friction and wear testing apparatus (UMT-2, CETR, USA) in a reciprocating ball on plate pattern style. Throughout the testing procedure the natural tooth was always immersed in artificial saliva, even during the cleaning of the samples before the experiment. The friction and wear tests were controlled by a computer. The relation between surface friction and displacement at every cycle was recorded. The frictional coefficient was automatically calculated and recorded by the UMT-2 control software. The average width of each wear track on enamel surface was calculated from five measurements in the middle part of the track. The spacing between neighbor measurements is 10% of the maximum track length. The Artificial saliva was used to simulate the actual oral condition and its composition was showed in Table 1 [9]. The enamel sample was tested with antagonist made by zirconia samples with well-polished and self-glazed surfaces, respectively, under constant static load, vertical load 4 N, and cyclic friction with back-and-forth movement pattern. Every sample was tested with four different antagonists, each for 5000 cycles, at frequency 2 Hz and sliding displacement 1 mm.

2.3. Microstructure characterization

The surface microstructure of the self-glazed zirconia, polished zirconia, and blank-machined zirconia ceramics was characterized by using a scanning electron microscope (SEM, JSM-7401F, JEOL, Tokyo, Japan). The samples were washed by water and acetone in an ultrasonic bath before loaded into the SEM and the SEM observation was carried out on the surface without any coating. Accelerating

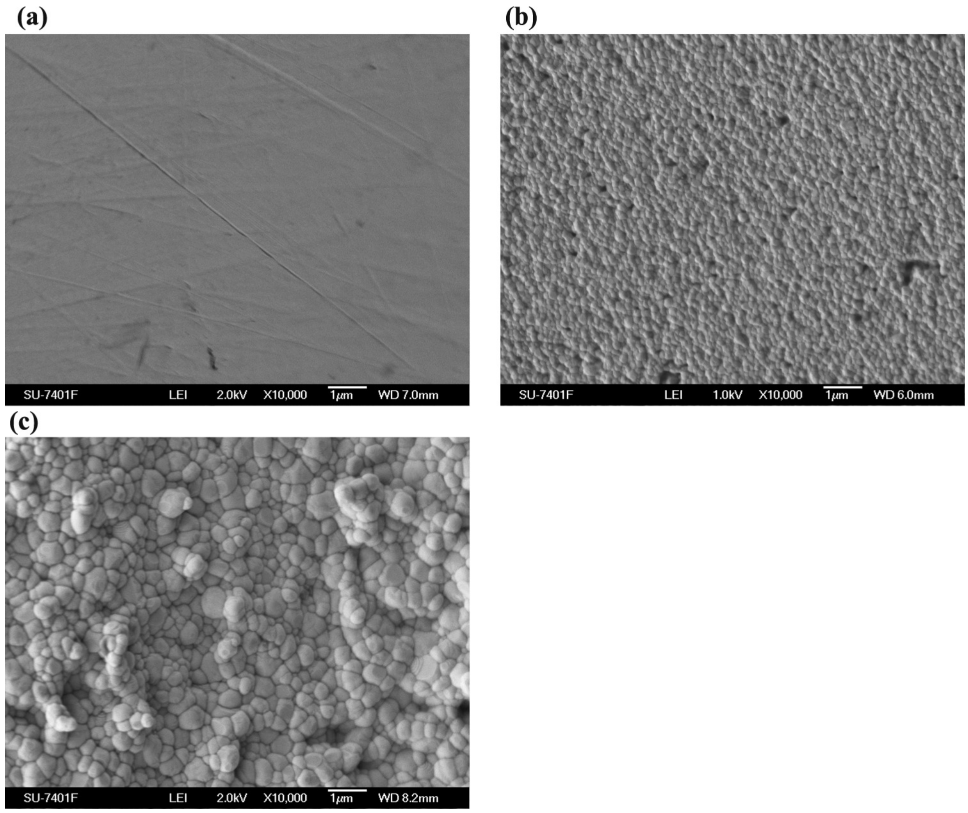

Fig. 2. SEM images taken on the surface of the polished zirconia (a), self-glazed zirconia (b), and blank-machined zirconia ceramics (c) as a reference.

voltages of 1 kV and 2 kV were applied in order to reduce the charging up of the samples.

The surface topography of the self-glazed zirconia was characterized in a 2 μm × 2 μm area by an Atomic Force Microscope (AFM, XE-100, Park Systems, Suwon, Korea) in DC-EFM mode with scan rate of 0.8 Hz. The surface roughness was also characterized by white light interferometer. The sample was washed by water in ultrasonic bath and dried at room temperature before the observation.

The wear scars on the worn surfaces of teeth enamel were investigated both by an optical microscope (Nikon Eclipse LV100, Nikon Co., Ltd., Japan) and an environmental scanning electron microscope (SEM, SSX-550, Shimadzu, Japan). The widths of worn scars were measured on the optical microscope images.

3. Results and discussion

3.1. A unique surface of self-glazed zirconia

Fig. 2 shows the SEM microstructure taken on the surface of well-polished, self-glazed and blank milled zirconia ceramics, with the latter being quoted as a reference. It appears that the surface of self-glazed zirconia is very smooth on micrometer scale in comparison with a rough surface of the blank-milled zirconia ceramics, although the well-polished zirconia is the smoothest one with only nano-scale scratches on it. Materials are removed during the traditional milling of pre-sintered or partially sintered zirconia blanks of ~55% relative density by a local fracture mechanism, i.e., crystalline grains are peeled off one by one or in groups leaving behind a rough surface full of ditches in the scale of half to a few micrometers, see Fig. 2(c). Polishing would flatten this rough surface by removing materials in nano- and even atomic steps. The smooth surface on self-glazed zirconia is inherently formed by local plastic deformation introduced during a precision wet-chemistry process. A surface

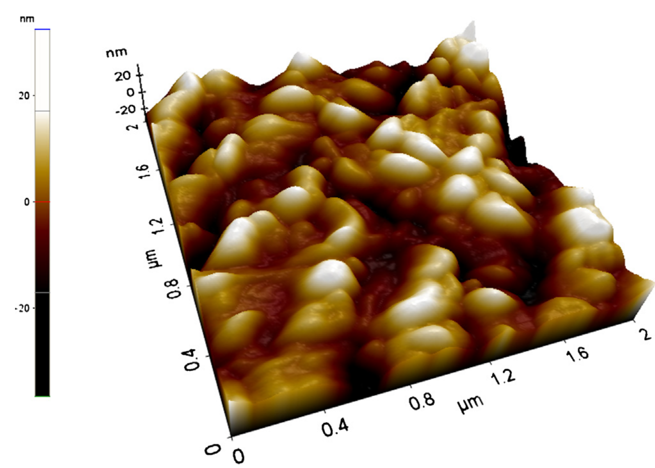

Fig. 3. An AFM image taken on the surface of the self-glazed zirconia ceramic sample.

roughness in the scale of 1/3 to 2/3 of the grain size does present on the surface of the self-glazed zirconia, which appears even more evident in the AFM image shown in Fig. 3. Nano-grains with average size in the range of 100–250 nm tend to coalesce to form aggregates of up to 1 μm, among which ditches in the scale of 1/3–2/3 of the size of nano-grains are formed.Under a lower magnification the characterization by white light interferometer revealed that on millimeter scale there are clear machining tracks with ridges of ~2 μm high between the neighboring tracks on the surface of blank milled zirconia ceramics, see Fig. 4(a), whereas, such ridges are not observable on the surface of self-glazed zirconia, see Fig. 4(b).

It has been established that due to the strong reflection of light by a well-polished surface the polished zirconia restorations reveal a pearl-like optical appearance, thus are not favorable for clinical

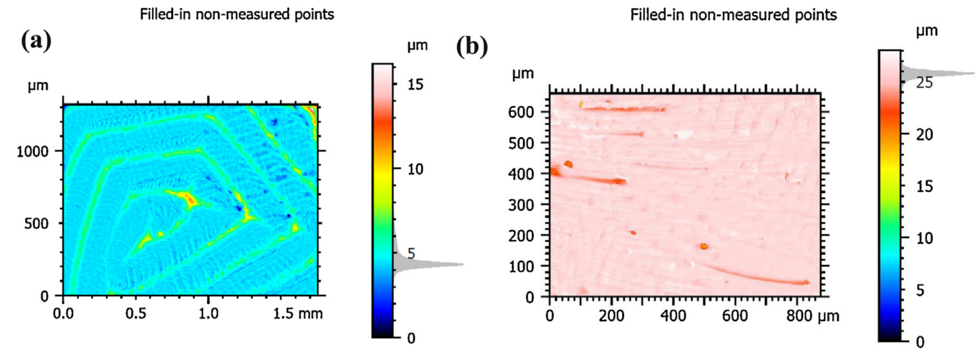

Fig. 4. Surface roughness of blank-machined zirconia (a) and self-glazed zirconia (b) characterized by the light interferometer on millmeter scale.

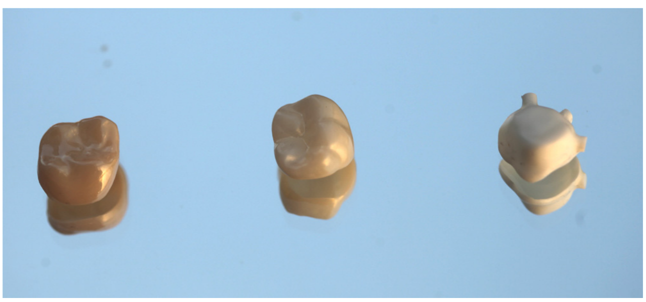

Fig. 5. A photo revealing the optical appearance difference among zirconia crowns with well-polished, self-glazed and blank-machined surfaces (from left to right).

applications [10]. However, the characteristic surface microscopic morphology of the self-glazed zirconia, i.e., micro-smooth yet with nanometer scale roughness viewed as densely packed grain structure, ensures this surface can scatter light besides reflecting it. It imitates the surface morphology of the natural dental enamel on which enamel prisms of 4–6 µm in size are clearly visible, cf. Fig. 8 below. A simultaneous reflection and scattering of light make the self-glazed surface having a sufficiently improved aesthetic appearance, see Fig. 5.

3.2. Friction behaviors

Fig. 6 shows the time-dependent frictional coefficient of natural enamel against the surfaces of well-polished zirconia and self-glazed zirconia ceramics. Although the self-glazed surface shows a slight low frictional coefficient at the beginning, both surfaces achieve the same steady state frictional coefficient, around 0.5. The high strength and toughness of zirconia enabled it to resist surface damage under stress. So the surface of polished zirconia antagonist kept its fineness during long time frictional process, presenting a stable frictional coefficient over time.

Frictional coefficient is a very important parameter that reflects the intrinsic interaction characteristics of tribology. An ideal dental restoration should have appropriate frictional coefficient with natural teeth for the benefit of chewing food without excessive abrasion of natural teeth. In the present study, the enamel sample and antagonists were tested in artificial saliva under similar stress and sliding distance with human teeth to mimic the oral chemical and mechanical environment [11]. The steady state frictional coefficient of the well-polished zirconia sample (around 0.5) in the present study is comparable to the numbers obtained in our previous study for both the well-polished zirconia and porcelain samples (both of them are around 0.55) [6]. Veneer porcelain is the common material used in fixed and implant dental restorations. A combination of the present and our past results shows that both

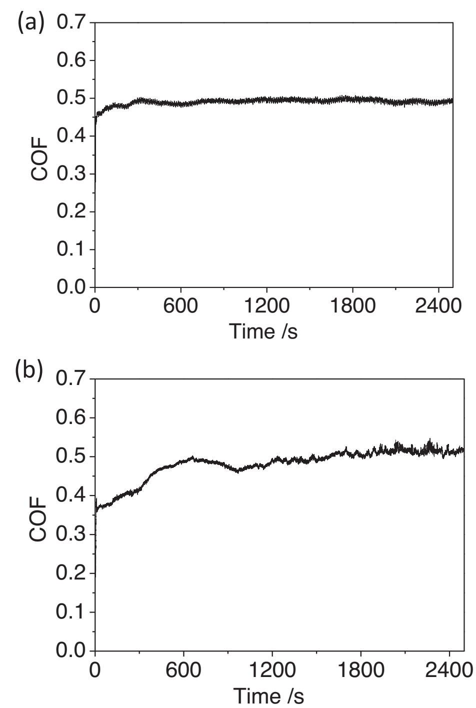

Fig. 6. The frictional coefficient of natural enamel against the surface of well-polished (a) and self-glazed zirconia ceramics (b), plotted versus the time.

self-glazed and well-polished zirconia restorations have the similar friction behaviors with porcelain veneered restorations [6]. The fluctuation of frictional coefficient by time observed on self-glazed zirconia ceramics can be ascribed to the presence of nanometer scale roughness.

3.3. Wear behaviors

Fig. 7 shows an optical microscope image revealing the wear tracks generated by the wear of dental enamel against the well-polished zirconia and self-glazed zirconia antagonist. The average width of the wear track on the enamel surface is determined to be 372 ± 6 µm and 349 ± 8 µm for the former and latter cases, respectively. The width and length of wear scars on the enamel surface by well-polished and self-glazed zirconia antagonists are almost the same. Therefore, it is reasonable to estimate that the wear loss volume of dental enamel is similar in both cases.

Fig. 8 shows the morphology of the original polished and the worn surface of the natural enamel after wear test against the well-polished zirconia and the self-glazed zirconia antagonist, respectively. Under the environmental SEM the well-assembled

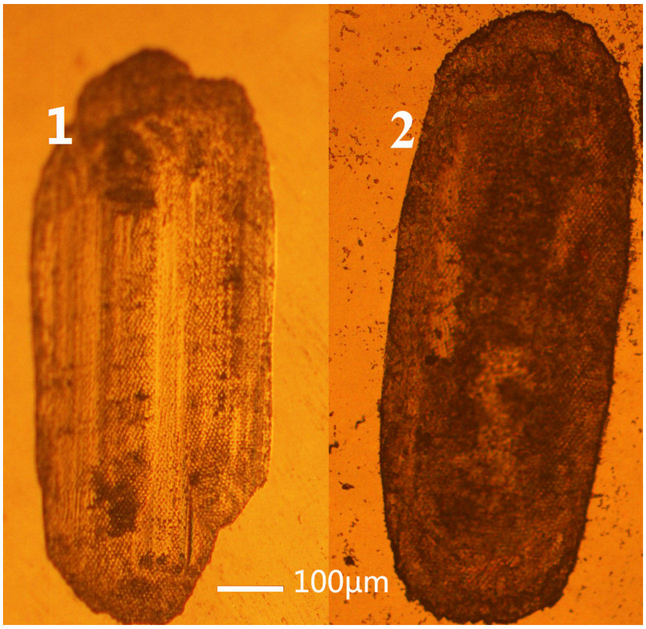

Fig. 7. The width of the wear tracks on natural enamel viewed an optical microscopic image; against well-polished zirconia (1) and self-glazed zirconia (2).

enamel prisms in the range of 4–6 µm are clearly visible on the original polished surface of the natural enamel as shown in Fig. 7(a and b). This is the section view of the enamel prisms, perpendicular to which every enamel prism is constituted by a bunch of elongated and well-assembled hydroxyapatite nano-crystals. Between the prisms, there is an interprismatic substance containing more organics and less mineral that is well preserved in wet condition under the environmental SEM [12,13]. Such well-arranged enamel prism structure is hard to be observed by conventional SEM carried out under high vacuum due to the dehydration and the conductive coating that may either destroy the prisms or hide their surfaces.

As shown in Fig. 8(a–d), part of the enamel prisms are hidden in a very similar way by a deposition layer most probably formed during initial grinding and polishing of the freshly extracted tooth. No noticeable increase of such deposition layer and the formation of scratches are observed on the worn surface of the dental enamel in both cases of wear of enamel against well-polished zirconia and self-glazed zirconia antagonists. It indicates that the observed wear in both cases is not the abrasive wear type but a moderate type of surface fatigue.

The implication of the present results is the same as that of our previous results [6]. When zirconia slid over the enamel surface a compression zone is generated ahead the zirconia antagonist, whereas behind the antagonist plastic deformation of enamel creates a zone of tension. Micro-cracks thus would nucleate in the subsurface of enamel and propagate [14]. The micro-cracks initiated in enamel under high stress prefer to form a reticular framework instead of prolonging straight in depth direction. Eventually, materials surrounded by the cracks propagating to the surface would get lost. This micro-fracture character is strongly determined by the unique structure of dental enamel that is composed by well-assembled enamel prisms and interprismatic substance [15]. Therefore, the wear behaviors of the dental enamel against the self-glazed zirconia restoration is similar to that of the well-polished zirconia restorations commonly used in clinic nowadays, whereas the former demonstrates sufficiently improved aesthetic appearance than the later.

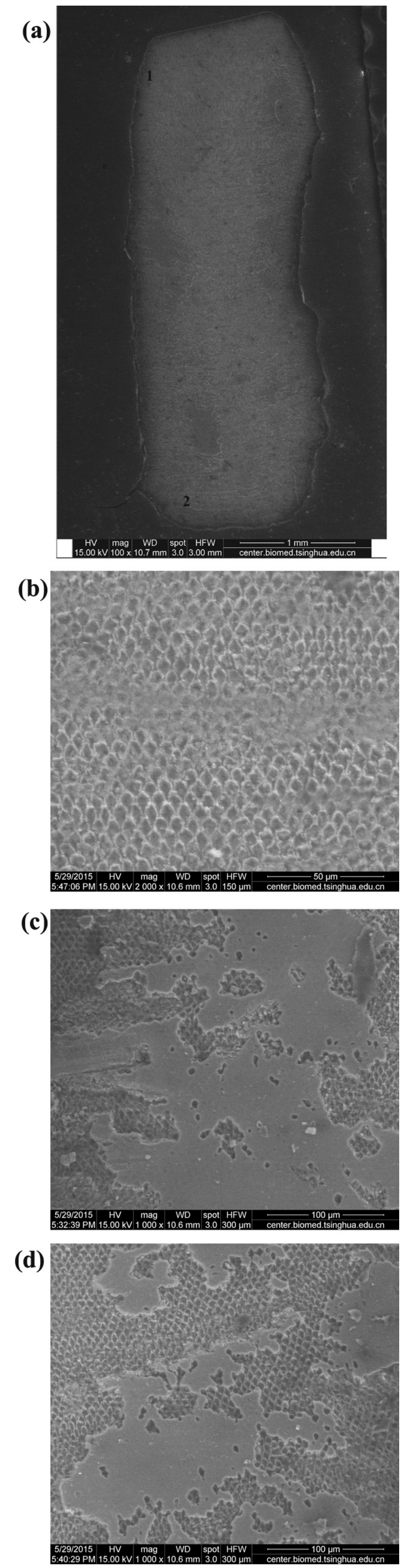

Fig. 8. SEM images revealing the microscopic morphology of the natural enamel (a and b) and the worn surface of the natural enamel after wear test for 5000 cycles against the well-polished zirconia (c) and the self-glazed zirconia (d), respectively. The low magnification image shown in (a) gives an overview of the embedded tooth and its surface morphology inside the epoxy resin block in which "1" and "2" indicate the location of wear tracks generated by wearing against well-polished zirconia and self-glazed zirconia, respectively.

4. Conclusions

1) The measured frictional coefficient of the tested zirconia samples against the enamel of freshly extracted tooth confirms that self-glazed zirconia has almost the same frictional coefficient as well-polished zirconia surface. The fluctuation of frictional coefficient by time observed on self-glazed zirconia ceramics can be ascribed to the presence of nanometer scale roughness.
2) The wear scars observed on the worn surface of enamel against self-glazed zirconia and well-polished zirconia surface reveal fatigue wear style.
3) Comparing to well-polished zirconia ceramics, the self-glazed zirconia ceramics shows similar friction and wear performance against natural tooth while provides sufficiently improved aesthetic appearance, revealing its potential for direct clinical use.

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References

[1] F. Beuer, M. Stimmelmayr, W. Gernet, D. Edelhoff, J.F. Guh, M. Naumann, Prospective study of zirconia-basedrestorations: 3-year clinical results, Quintessence Int. 41 (2010) 631–637.
[2] Y.H. Liu, H.L. Feng, Y.W. Bao, Y. Qiu, N. Xing, Z.J. Shen, Fracture and interfacial delamination origins of bilayer ceramic composites for dental restorations, J. Eur. Ceram. Soc. 30 (6) (2010) 1305–1927.
[3] Y.H. Liu, G.H. Liu, Y. Wang, Z.J. Shen, H.L. Feng, Failure modes and fracture origins of porcelain veneers on bilayer dental crowns, Int. J. Prosthodont. 27 (2) (2014) 147–150.
[4] V. Preis, M. Behr, S. Hahnel, G. Handel, M. Rosentritt, In vitro failure and fracture resistance of veneered and full-contour zirconia restorations, J. Dent. 40 (11) (2012) 921–928.
[5] V. Preis, M. Schmalzbauer, D. Bougeard, S. Schneider-Feyrer, M. RosentrittSurface, Properties of monolithic zirconia after dental adjustment treatments and in vitro wear simulation, J. Dent. 43 (1) (2015) 133–139.
[6] L. Wang, Y.H. Liu, W.J. Si, H.L. Feng, Y.Q. Tao, Z.Z. Ma, Friction and wear behaviors of dental ceramics against natural tooth enamel, J. Eur. Ceram. Soc. 32 (11) (2012) 2599–2606.
[7] J. Sripetchdanond, C. Leevailoj, Wear of human enamel opposing monolithic zirconia, glass ceramic, and composite resin: an in vitro study, J. Prosthodont. Dent. 112 (5) (2014) 1141–1150.
[8] B.J. Chong, A.K. Thangavel, S.B. Rolton, M. Guazzato, I.J. Klineberg, Clinical and laboratory surface finishing procedures for zirconia on opposing human enamel wear: a laboratory study, J. Mech. Behav. Biomed. Mater. 50 (2015) 93–103.
[9] C.D. Mayworm, S.S. Camargo, F.L. Bastian, Influence of artificial saliva on abrasive wear and microhardness of dental composites filled with nanoparticles, J. Dent. 36 (2008) 703–710.
[10] H.K. Kim, S.H. Kim, Effect of the number of coloring liquid applications on the optical properties of monolithic zirconia, Dent. Mater. 30 (9) (2014) 229–237.
[11] J. Zheng, Z.R. Zhou, J. Zhang, et al., On the friction and wear behavior of human tooth enamel and dentin, Wear 255 (2003) 967–974.
[12] C.F. Li, S. Risnes, SEM observations of Retzius lines and prism cross-striations in human dental enamel after different acid etching regimes, Arch. Oral Biol. 49 (1) (2004) 45–52.
[13] J. Seong, A. Virani, C. Parkinson, N. Claydon, N. Hellin, R.G. Newcombe, N. West, Clinical enamel surface changes following an intra-oral acidic challenge, J. Dent. 43 (8) (2015) 1013–1020.
[14] H. Mair, T.A. Stolarskij, R.W. Vowles, C.H. Lloyd, Wear: mechanisms, manifestations and measurement, J. Dent. 24 (1996) 141–148.
[15] S. Bechtle, S. Habelitz, A. Klocke, et al., The fracture behaviour of dental enamel, Biomaterials 31 (2010) 375–384.