# Future Opportunities in Zirconia Applications in Dentistry: Functionally Graded Concept. Review

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Received: May 05, 2020; Published: August 11, 2020

## Abstract

Alumina and zirconia-based ceramic dental restorations are designed to repair functionality as well as esthetics of the failed teeth. However, these materials exhibited several performance deficiencies such as fracture, poor esthetic properties of ceramic cores (particularly zirconia cores) and difficulty in accomplishing a strong ceramic-resin-based cement bond. Therefore, improving the mechanical properties of these ceramic materials is of great interest in a wide range of disciplines. Consequently, spatial gradients in surface composition and structure can improve the mechanical integrity of ceramic dental restorations.

The aim of this article was to review the current status of the functionally graded dental prostheses inspired by the dentino-enamel junction (DEJ) structures and the linear gradation in Young's modulus of the DEJ, as a new material design approach, to improve the performance compared to traditional dental prostheses. This is a remarkable example of nature's ability to engineer functionally graded dental prostheses. The current article opens a new avenue for recent researches aimed at the further development of new ceramic dental restorations for improving their clinical durability.

Keywords: Dental Ceramics; Alumina; Zirconia; Dental Multilayer; Bioinspired Functionally Graded Materials

## Introduction

Since the beginning of dental practice, practitioners are faced with the challenge of the development and selection of biocompatible, long lasting, direct filling tooth restoratives and indirectly prosthetic materials capable of withstand the aggressive environment of the oral cavity. To improve performance of new updated materials compared to traditional homogeneous and uniform ones, the concept of functionally graded materials (FGM) has been conceived as a new material design approach [1].

Ceramics, including zirconia, have high hardness, low density and weight, brittleness, and excellent high-temperature fracture, creep, corrosion, radiation, wear, and thermal shock resistance. On the opposite side, metals are typically ductile, have high tensile strength, high toughness and high density. To take advantages of both, metal-ceramic FGMs can also be designed in order to achieve the desired function; the transition profile between the two materials must be designed. This makes the FGM a new class of materials, diverse from conventional homogeneous composite [2].

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In the literature functionally graded biomaterials development for implants for medical and dental applications has been reported [3]. To integrate dissimilar materials without formation of severe internal stress and combines diverse properties into a single material system the FGM concept is applied. FGM concept development has its origin in the sophisticated properties which arise from materials in nature, such as and bones and teeth [4].

For demonstration that functional gradation has been utilized by biological adaptation, the design of a bone with a change from dense, stiff external structure (the cortical bone) to a porous internal one (the spongy bone), is an example. A biologic evolution and optimization of the material's response to external loading are reflections to this bone structure. Therefore, for an artificial implant, optimized structure, should show similar gradation. In order to improve the implant performance in terms of biocompatibility and stress distribution, the same trend has been observed in the development of functionally graded dental implants with the introduction of surface coatings, porosity gradients and composite materials made essentially of metal and ceramics (e.g. hydroxyapatite) [5]. Similarly, several methods were proposed to mimic the DEJ using synthetic materials which were inspired by the microstructure and mechanical properties of natural teeth. It has been reported that the functionally graded glass/zirconia/glass structure as alternative for homogeneous zirconia. Increasing damage resistance, translucency, and allowing etching and silane application for reliable bonding mechanisms of a pre-sintered Y-TZP result from coating of its top and bottom with a slurry of glassy powder [6].

Recently, bioinspired functionally graded enamel structures in the design of dental multi-layers have been proposed, as alternative technique, aiming the enhancement of the overall performance of metal-ceramic and all ceramic dental restorative systems. This technique allows the production of a material with very different characteristics within the same material at various interfaces. Bioinspired functionally graded approach is an innovative material technology, which has rapidly progressed both in terms of materials processing and computational modeling in recent years. Bioinspired functionally graded structure allows the integration of dissimilar materials without formation of severe internal stress and combines diverse properties into a single material system [2,7]. The graded structure eliminates the sharp interface resulting from traditional core-veneer fabrication, eliminating the potential for delamination between the layers [8]. Graded transitions can also reduce stress concentrations at the intersection between an interface and a free surface [9]. Similarly, the local driving force for crack growth across an interface can be increased or reduced by altering the gradients in elastic and plastic properties across the interface [10].

#### Importance of functionally graded concept

Two billion \$ are the revenue of dental crowns per year, with 20% of these crowns being all-ceramic units [11]. Failure rate of dental crowns represents 3% per year [12] and in cases where ceramics are used as the top layer, more than 20% of them failed within the first 5 years of use [13]. This paid the attention of researchers to develop ceramic crowns that are more resistant to cracking under occlusal contact. Ceramic crown structures are idealized as flat multilayered structures that will deform if subjected to Hertzian contact loading [14], often leading to the pop-in of a subsurface radial crack due to the stress concentration in the subsurface regime in the top ceramic layer [15].

The opportunity to reduce the stress concentrations (at the bottom of the top ceramic layer in crown structures) was inspired by the nano-indentation measurements in a research and showed that the Young's modulus varies from 70 GPa for enamel to 20 GPa for dentin. Across the dentin-enamel junction (DEJ) Young's modulus variation was also shown to be approximately linear [16]. In a study inspired by the DEJ structure and the linear gradation in the Young's modulus of the DEJ, modeled the Hertzian contact-induced stress concentrations and showed that the bio-inspired functionally graded structure resulted in lower stresses in the subsurface region of the top ceramic layer [15]. Subsequent works also showed similar reductions in stress concentrations [17].

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The aim of producing FGM is to obtain a material with two different characteristics in its two opposite faces (Figure 1). The properties of this innovative FGM can mimic the natural gradients occur, including a graded elastic modulus in hard tissues such as the human enamel and dentin-enamel junction [18]. This novel technology is designed to improve the performance of the materials [19].

A		-
	Core	
	Core	
	Graded	
B		
	20 µm	

**Figure 1:** Morphology of the graded zone. (A) Schematic of graded structure. (B) Section view of graded zone of glass-infiltrated yttria stabilized zirconia.

## Methods used for bioinspired functionally graded approach

Although the efficacy of FGMs has been recognized since the early 1970s [20], the field of FGM did not take off until the mid-1980s, probably due to a lack of suitable fabrication methods until that time. This concept has been later expanded for different application such as coatings, packing, optical, biomedical, etc. In the biomedical field, several approaches have been used to develop functionally graded biomaterials for implants [21]. With established methods currently available to synthesize and process materials, gradations in composition, structure, and properties could be engineered over a wide range of length scales ranging from nanometers to meters. There are a wide range of process technologies that are now available for fabrication FGMs such as powder metallurgical process [22], layer stacking [23], glass infiltration [24], centrifugation [25], electrophoretic deposition [26], plasma spray [27], direct-write assembly [28] and rapid prototype color ink-jet printing [29].

Learning from nature, materials scientists increasingly aim to engineer graded materials that are more damage-resistant than their conventional homogeneous counterparts. This is particularly important at surfaces or at interfaces between dissimilar materials, where contact failure commonly occurs. Therefore, many engineered materials are graded in some manner, but functionally graded materials (FGMs) are often characterized by a gradient purposefully formed using compositional or microstructural design. FGM is a material with engineered gradients of composition, structure and/or specific properties aiming to become superior over homogeneous material composed of same or similar constituents [9,30].

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#### Dental prostheses mimic the dentino-enamel junction behavior

Among the previously mentioned processing methods, the glass infiltration technology is particularly suitable for the fabrication of all-ceramic restorations [31]. It combines anesthetic, low modulus, and low hardness glass "veneer" with a high strength ceramic "core", without a sharp interface between the materials (Figure 2). The lack of inter-face due to grading improves interfacial bond strengths, reduces residual stresses, and eliminates delaminations. The processing of these structures is simple and straightforward and can be readily adapted to CAD/CAM technology [32].



Figure 2: Schematic of the conventional sharp restoration and the new graded approach.

#### Researches done to investigate this concept

The ceramic crown structures under occlusal contact are idealized as flat multilayered structures that are deformed under Hertzian contact loading. Those multilayers consist of a crown-like ceramic top layer, an adhesive layer and the dentin-like substrate. Bio-inspired design of the adhesive layer proposed functionally graded multilayers (FGM) that mimic the dentin-enamel junction in natural teeth [33].

Investigation of the effects of FGM layer architecture on the contact-induced deformation of bio-inspired dental multilayers was performed by finite element modeling to explore the effects of thickness and architecture on the contact-induced stresses that are induced in bio-inspired dental multilayers. The conclusions of this study were: 1) The FGM structures reduce the stresses in the top zirconia ceramic crowns. Greater reductions in the stresses in the ceramic crowns were observed when the gradients in the moduli of the FGM were higher near the ceramic layer, 2) The actual FGM structures fabricated using nano composite layers of epoxy/ceramic mixtures had 20 - 40% higher critical pop in loads than flat conventional dental multilayers without FGM and 3) The bio-inspired, functionally graded, dental multilayers can be fabricated and incorporated into "tape" structures that can be used to reduce the top layer stresses in dental ceramic restorations. Further work is clearly needed to assess the clinical performance of such structures under cyclic loading conditions and environmental exposure relevant to occlusal conditions [33].

#### Graded glass-zirconia structures

Glass-zirconia structures with gradual elastic modulus maybe created by using infiltration method [31]. Comparing to the sintering temperature of zirconia, zirconia templates with somewhat low heat-treatment temperature are used for combining glass infiltration and

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zirconia densification into a single process [31,34]. This way the glass infiltration depth can be tailored by manipulating the porosity of the zirconia templates. Therefore, the grain growth and/or destabilizing of the tetragonal zirconia phase [35] associated with the post-sintering heat-treatment can be prevented. As coefficient of thermal expansion and Poisson's ratio of the infiltrating glass and zirconia (3Y-TZP) are relatively the same, no significant long-range thermal stresses are developed in the graded structure [36]. The resultant structure consists of a thin, outer surface residual glass layer followed by a graded glass-zirconia layer at both the top and bottom surfaces (Figure 3).



Figure 3: Cross-sectional view of a graded glass-alumina in (a) and graded glass-zirconia structure (b), respectively.

Investigators added bioinspired FGM layer between the zirconia dental ceramic and the dental cement and reported the effects of the functionally graded layer on the stress in the crown and its surrounding structures. They found that the functionally graded layer promotes significant stress reduction and improvements in the critical crack size. Their conclusion was that the low stress concentrations were associated with the graded distributions in the dentin-enamel junction [37]. Similar reductions in stress concentrations in simulations using a bio-inspired functionally graded material layer were shown subsequently, by other researchers. The processing of such functionally graded multi-layers and the increased critical loads in dental multilayer structures with FGM structures were demonstrated by their experimental work [38].

Recently, some authors patented three types of multilayered composite materials were produced to introduce newly designed functionally graded dental post using powders of zirconia ( $ZrO_2$ ), alumina ( $Al_2O_3$ ), hydroxyapatite (HA) and titanium (Ti) [39]. Those authors investigated the stress distribution of the newly designed functionally graded dental post which consisted of multilayer design of  $ZrO_2$ -Ti-

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HA and compared it to posts fabricated from homogeneous material such as titanium and zirconia. They reported that stress distribution in the new dental post exhibited several advantages in terms of the stress and strain distribution at the post-dentine interface of FGDP compared to posts fabricated from homogeneous material [40].

#### Graded glass-alumina structures

Glass-alumina graded structures may be produced by infiltrating dense alumina surfaces with silica-based glasses [41]. Following a power law relationship, the transition of elastic modulus from the graded glass-alumina surface to the alumina core is continuous [42]. The resultant structure consists of a thin, outer surface residual glass layer followed by a graded glass-alumina layer, sandwiching a dense alumina core (Figure 3). Inspired by the microstructure and mechanical properties of natural teeth, synthetic functionally graded materials were proposed to mimic the DEJ.

Researchers described a procedure to produce a DEJ-like interface and enamel coating involved depositing slurries of oxide or glass powder by a draw-down blade method, drying at then higher temperature heating. They used alumina-glass or alumina-polymer composite to mimic the dentin and a calcium phosphate-based coating to mimic the enamel. Bonding between the two materials was accomplished by a eutectic melt in the CaO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> system. The inter penetration in this DEJ-like interface originates from a solidified melt phase penetrating into the dentin [43].

In a research authors added bioinspired FGM layer between the dental ceramic and the dental cement and investigated the effects of the functionally graded layer on the stress in the crown and its surrounding structures. From their results, the functionally graded layer was shown to promote significant stress reduction and improvements in the critical crack size. From their study, they concluded that the low stress concentrations were associated with the graded distributions in the DEJ. This provided new insights into the design of functionally graded crown architecture that can increase the durability of future dental restorations [37].

Graded dental restorations have been shown to display improved features relative to conventional ones, namely higher resistance to contact and sliding [44]; higher adhesion of porcelain to the substructure (metal or ceramic) [45]; improved esthetical properties and improved behavior under fatigue conditions. Another important point to which the FGM design can address is the reduction of thermal residual stresses that remains at the metal-ceramic interface during the cooling cycles after the porcelain firing. These stresses are further magnified when there is a significant difference between the thermal expansion behavior of the metal and the porcelain. Depending on the thermal residual stress level that remains in the crown and together with those arising from occlusal loads, a catastrophic failure of the restoration can occur. FGMs have been shown to decrease significantly the thermal residual stresses formed at the interface between metals and ceramics in other fields of applications [46].

Some studies demonstrated that when the contact surface of alumina or silicon nitride was infiltrated with aluminosilicate or oxynitride glass, respectively, they noticed that the graded glass/ceramic surfaces produced in this manner offered much better resistance to contact damage with and without a sliding action than either constituent ceramic or glass [47].

#### **Clinical implications**

The use of zirconia in crowns and bridges has increased over recent years, owing to esthetic and biocompatibility demands. However, the fact remains that porcelain-veneered zirconia restorations suffer unexpectedly high chipping rates, regardless of the manufacturer [48,49]. Also, aging populations will drive the demand for all types of dental restorations even higher [50]. If these chipping rates could be reduced, zirconia-based all-ceramic prostheses would become more widely used, addressing a quality of life issue [51]. Graded glass-zirconia structures offer a simple remedy.

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Zirconia cores are, however, only a portion of the all ceramic restoration. Alternative monolithic graded glass-zirconia restorations are recommended without porcelain veneer, which could be successfully and economically used in posterior applications. These restorations are suggested to eliminate the vulnerable porcelain veneer, while providing superior strength and esthetics. The color characterization of these graded glass-zirconia restorations is achieved by external residual glass and subsequent staining.

## Conclusion

Improving biological and mechanical properties of zirconia can be done through introducing a new method for fabrication of zirconia unitizing functionally graded concept. In order to replace the mechanical function of tooth from a restorative perspective, it is not only important to study its localized tissue properties but also its bulk structural behavior. Therefore, the functionally graded dental prostheses inspired by the DEJ have been reviewed. These prostheses such as "graded glass zirconia and graded glass alumina structures" offer better resistance to immediate flexural damage, better esthetics, and potentially better veneering and cementation properties over homogeneous ceramics materials. This new trend in zirconia fabrication is expected to improve biological and mechanical performance for zirconia, but it needs further investigation. The further development of the grading technology could potentially lead to superior long-term clinical performance for dental prostheses [52].

## **Conflict of Interest**

The authors declare that they have no conflict of interest.

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