

Functionally Graded Materials: An Overview of Dental Applications

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ABSTRACT

Background: Functionally graded material (FGM) is a smart configuration for designing composite materials to improve their long-term endurance. The main characteristic of an FGM is the gradual variation in composition and microstructure across its dimensions, resulting in enhanced properties. This approach has been applied in several fields of thermal barrier coatings, power industry, biomechanical, automotive, aerospace, mechanical, civil, nuclear, aeronautic, and naval engineering.

Aim: This article is intended to provide an overview of the FGMs, their application in dentistry, and the employment of the concept on the mechanical performance of dental restorative systems.

Results: A thorough review of FGM dental applications has found that several researchers have been able to develop different configurations based on FGM concept to avoid failures, such as infrastructure, abutment failures in implantology, and prosthodontics. These strategies result in a gradual transition between dissimilar materials decreasing residual stresses generated during fabrication and function.

Conclusion: Of late, multiple parallel researches based on the FGM concept are being carried out on dental implants coated with hydroxyapatite (HA), zirconia (Zr), and its oxides to improve osseointegration and reduce stress behavior. This review attempts to present a thorough understanding of FGMs, their characterization, manufacturing techniques and technology, and their dental applications.

Clinical significance: Throughout the past decade, the use of endosteal dental implants as a treatment modality of choice in oral rehabilitation cases has increased. Unfortunately, no artificial implants have biomechanical properties equivalent to that of the surrounding bone. Based on FGM concept, a graded composition between titanium and HA/Zr on the implant surface has now been achieved that has resulted in improved mechanical behavior and osseointegration.

Keywords: Biomaterials, Dental implants, Functionally graded materials, Graded microstructure composites, Powder metallurgy.

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BACKGROUND

Striving to achieve constant improvements of materials has always been the aim of material science researchers to satisfy the current global needs in various disciplines, such as engineering,¹ bioengineering,² aerospace,³ biomedical sciences,^{4,5} and dentistry.⁶ These improvements can be either in the form of enhancements/variations of material properties or production of new materials with desirable benefits. Lately, a newer concept called FGMs has emerged and is being studied for its various applications. Research and development of FGM have become a forefront of material science nowadays⁷ and are receiving worldwide attention. They serve for wide application including biomechanical, automotive, aerospace, mechanical, civil, nuclear, and naval engineering.⁸ New applications are continuously being discovered and developed.

The FGM concept originated in Japan in 1984 during the space plane project, in the form of a proposed thermal barrier material capable of withstanding a surface temperature of 2,000 K and a temperature gradient of 1,000 K across a cross section of <10 mm.^{9,10} Since then, FGM thin films have been comprehensively researched and are almost a commercial reality. In general, FGM is a composite material formed of two or more constituent phases with a continuously variable distribution which may be reflected either in their volume or weight fraction, orientation, and shape. In engineering studies, volume is a focus of many researchers where the phase volume fraction variations may be exclusively applied through the thickness of the structure surface coordinates of a plate or shell fraction.¹ The homogeneity cannot be achieved by conventional homogeneous materials, and whatever the approach is adopted for homogenization, the interaction between the particles should be of concern.¹¹

The concept of FGMs is to make a composite material of two or more constituent phases by varying the microstructure from one material to another material with a specific gradient end up, where the final product has the best

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Fig. 1: Schematic representation of a continuously graded microstructure



Fig. 2: Schematic representation of a stepwise graded microstructure

properties of both the parent materials.¹² For continuous grade structure, the change of composition and microstructure occurs continuously with position (Fig. 1), whereas stepwise microstructure feature occurs in stepwise manner producing multilayered structure with interface existing between discrete layers (Fig. 2). The smooth transition for the FGM can be measured from the measured variation in hardness, fracture toughness, and cellular adhesion across the cross section, e.g., FGM of HAp–Al₂O₃–yttria-stabilized Zr nanocomposite has increased bulk toughness combined with unrestricted surface bioactivity which is a suitable property for bone implant.⁵

REVIEW METHOD

An electronic search of publications was made using the electronic databases: ScienceDirect® and PubMed® through our institutional membership access. The inclusion criterion was abstract or full-text articles: Original

research, reviews, or systematic reviews. The keywords used for the search were selected listing the following five combinations: (1) Functionally graded materials, (2) biomaterials, (3) graded microstructure composites, (4) dental implants, and (5) powder metallurgy (PM).

CHARACTERIZATION OF FGMS

An FGM microstructure is designed to enhance various mechanical and thermal properties by reducing mismatch of inherent characteristics. A thorough understanding of the overall structure must be available to better understand the approach to configuration. For example, optical microscopy is one of the commonly employed methods to visualize the spatial distribution, transitions of the material, and microstructure of an FGM. Similarly, chemical composition, optical, thermal, and mechanical behavior, can be determined by various other characterization techniques and computer simulation analysis. Table 1

Table 1: List of techniques to analyze the FGM properties

<i>Properties analyzed</i>	<i>Techniques/equipment used</i>	<i>Evaluations</i>
Microstructural	Optical microscope Scanning electron microscope	Microstructure (spatial distribution and transitions)
Chemical	X-ray fluorescence Energy-dispersive X-ray Spectroscopy	
Phases identification	X-ray diffraction Raman spectroscopy	
Mechanical	Sonic resonance Micro-indentation Nano-indentation; atomic force microscope	Determine (E, v, G)*, Determine (E, H, S, Klc)* Determine (E, H, S, Klc)
Optical	Spectrophotometry	Determine (ΔE, L*, a*, b*)*
Adhesion	Shear bond test Scratch or nano-scratch test	Interface adhesion strength (MPa)
Computed behavior simulation	Finite element analysis	Residual stresses and thermal behavior prediction

ΔE: Color difference; L: Lightness difference; a* and b*: Chromaticity differences; E: Young's modulus; v: Poisson ratio; G: Shear modulus; H: Hardness; S: Stiffness; Klc: Fracture toughness

presents a list of some available techniques to investigate the whole FGM structures.

INCEPTION OF FGM IN DENTISTRY

The development of FGM coatings has attracted considerable interest of dental material scientists and engineers. A major application in the biomaterial field is the design of implant surface coating systems to avoid mismatch of mechanical and biological properties between implant materials and bone. In dentistry, ceramic FGMs were mainly studied for the use as implant abutments.^{13,14} For the success of a dental implant's osteointegration, several factors should be considered which include health and bone quality, surgical techniques, implant design, surface characteristic, and the biomaterial and biomechanical factors.¹⁵ Their use as dental implant abutment is due to the higher toughness and the lower modulus of elasticity, increased esthetics, and corrosion resistance. Osman and Swain⁶ have critically reviewed Zr and its oxides as dental implant materials and compared them with titanium. Yttria-stabilized tetragonal Zr polycrystalline material exhibits superior corrosion and wear resistance. They have made a few recommendations for better Zr implants, such as strict quality control during the manufacturing process and avoidance of stress concentration areas, such as sharp threads.

The use of FGM as dental implant has been a focus of research in many studies. The advantages of using FGM as dental implants are mainly due to reduced stress shielding effect on the surrounding bones which usually arises in the presence of fully metallic implants, improved biocompatibility, prevention of thermal-mechanical failure at the bone-implant interface, and achievement of biomechanical requirements during bone remodeling, thereby maintaining the bone's health status.^{16,17}

Other applications of FGMs were in the field of ceramometal technology. On conventional metal-ceramic restorations, a sharp transition occurs between infrastructure

and ceramic which induces a mismatch in the properties between the materials involved. A composite interlayer composed of both materials (metal + ceramic) can be used to create a transition material that mixes the properties of metal and ceramic, avoiding sharp transition in properties. Furthermore, the use of an FGM might significantly reduce failures between dissimilar materials. Henriques et al¹⁸ developed a composite interlayer to be applied at the interface between the metal infrastructure and the veneering ceramic, to impart a graded transition between the two materials (Fig. 3). They tested two types of metal-ceramic specimens: A conventional porcelain to metal specimens (PFM) and metal-ceramic specimens with a composite interlayer at the interface, designated as functionally graded restorations. They were able to improve metal-ceramic bond strength by ~140% relative to the conventional PFM restorations.

PROCESSING TECHNIQUES OF FGM

Functionally graded material production is divided into two main process classes. The first class is the constructive processes, where it produces gradients by stacking selectively two or more of the starting materials. This allows full and potentially automated control of compositional gradients. The second class is called "transport-based processes" that utilize natural transport phenomena to create compositional and microstructural gradients during the production of a component.¹⁹ These techniques can be classified into gas-based, liquid-phase, solid-phase, and biopolymeric-based processes.²⁰ The selection of these production methods is based mainly on the material combination, type of transition function required, and geometry of the desired component.²¹ Chemical vapor deposition (CVD), physical vapor deposition (PVD), ion plating, plasma spraying, and ion mixing are some examples of gas-based methods, whereas liquid-phase processes include centrifugal casting, combustion, tape casting, slip-casting, gel-casting, electrophoretic

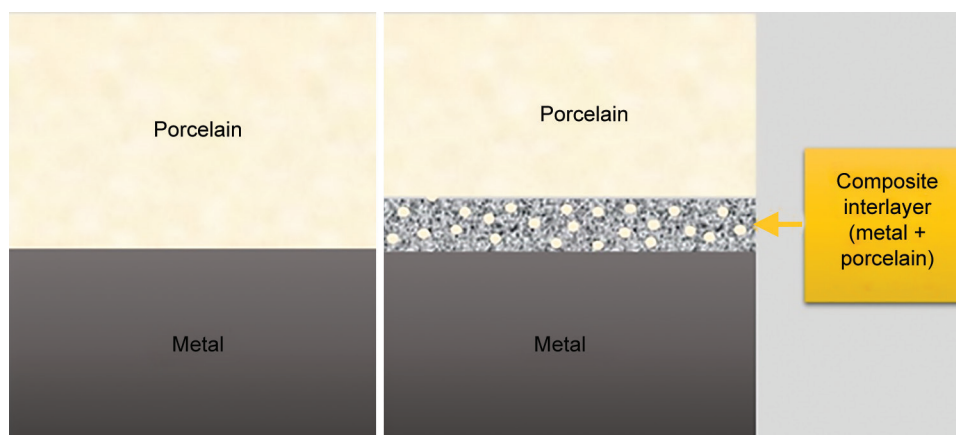


Fig. 3: Schematic representation of a conventional PFM and a FGR with a composite interlayer

deposition, chemical solution deposition, laser deposition, and directional solidification. Solid-phase processes include spark plasma sintering (SPS)⁵ and PM. Biopolymeric-based processes include freeze drying, compression molding, and electrospinning. Different processing techniques have different parameters that affect the final properties of the produced FGMs.²⁰

Functionally graded materials can be in the form of bulk composites, coating, or films.²² Several methods had been introduced to fabricate FGMs depending on the requirement, either for surface coatings or bulk FGM. Surface coatings are a thin FGM usually deposited by several vapor deposition techniques, such as CVD and PVD.²³ Other than that, surface coatings can also be prepared using techniques, such as plasma spraying,^{24,25} electrodeposition,²⁶ electrophoretics,^{27,28} ion beam-assisted deposition,²⁹ laser-engineered net shaping technology,³⁰ and self-propagating high-temperature synthesis.³¹ These techniques are generally slow, consume a lot of energy, and produce poisonous gas by-products. Therefore, it is uneconomic to produce surface-coated FGMs.³²

Bulk FGMs are FGMs with a gradient breadth in the order of millimeters to centimeters with continuous gradient profiles. This bulk FGM could be fabricated by PM technique,³³⁻³⁶ centrifugal method, solid freeform fabrication (SFF) method, laser cladding-based method, selective laser sintering (SLS), selective laser melting (SLM), three-dimensional (3D) printing, and few other techniques.^{37,38} Powder metallurgy technique involves weighing and mixing of powders, followed by stacking and ramming of pre-mixed powder and finally sintering³⁹ which gives a stepwise structure. The centrifugal method uses the force of gravity based on the difference of material densities through spinning. The fabrication of the FGMs by the centrifugal method can be classified into two categories depending on the processing temperature: The centrifugal solid-particle method and the centrifugal *in situ* method.⁴⁰ A new technique called centrifugal mixed-powder was introduced by Watanabe et al,⁴⁰ which could produce FGM-containing nanoparticles.

Mahamood et al²³ recommended the SFF technique in their overview of fabrication methods and application areas of FGM. Solid freeform fabrication method involves five basic steps: (1) Generation of computer-aided design (CAD) data from the software; (2) conversion of the CAD data to standard triangulation language (STL) file; (3) slicing of the STL into two-dimensional cross-section profiles; (4) building of the component layer by layer; and (5) removal and finishing.⁴¹ The advantages of SFF technique are higher speed of production with less energy consumption, maximum material utilization with minimal wastage, and ability to produce complex shapes

with design freedom (as parts are produced directly from CAD). This technique in conjunction with laser-based technology is among the most commonly used technique for FGM fabrication. Other techniques, such as laser cladding-based methods and SLM² are capable of producing fully dense components.

Spark plasma sintering is another method used to consolidate powders prepared by mixing ZrO₂ (3Y-PSZ) with 10 to 50 vol% of AISI316L granules.⁴² The material was designed to remove elements of low fracture toughness of ceramics and degradation and toxicity of metals, which is important for artificial joints. The fabricated FGMs showed a higher fracture toughness than a monolithic ZrO₂ when the layers have a thickness of more than 2.0 mm. Functionally graded materials produced by SPS exhibited a low porosity level and consequently fully dense specimens.⁴³ To produce the best material, multiobjective optimization methods should be applied to solve multicriteria problems that might arise during the fabrication process. Sadollah et al¹⁷ used the multiobjective particle swarm optimization method to determine the Pareto front.

HYDROXYAPATITE AND ZIRCONIA FGMs

Hydroxyapatite is a naturally occurring mineral form of calcium apatite and usually written as Ca₁₀(PO₄)₆(OH)₂. It is well recognized as a potential material for tissue replacement and repair, as it supports new bone formation which is an important factor for implant osseointegration. Hydroxyapatite is also a good osteoconductive material that can support cell fate process.⁴⁴ However, HA cannot be implanted alone as monolithic material for load-bearing implants, such as tooth and artificial bones due to its low fracture strength and poor fracture toughness when compared with human bone. Therefore, a large number of studies have been done to enhance the mechanical properties of this material by doping it with different metals (Ti or Ti-6Al-4V alloys), ceramics (ZrO glass, etc.), polymers (low density polyethylene, ultra high molecular weight polyethylene, etc.), and carbon nanotubes.⁵ Zirconia has been extensively employed for such applications attributing to its bio-inertness and high fracture toughness (6–10 MPa m^{1/2}).⁴⁵ Functionally graded material Zr was successfully processed through SPS at 1200°C for 5 minutes. The stepwise functional variation of hardness and toughness along the cross section of FGM was effectively achieved with strong adherence. However, a desired variation in porosity could not be achieved. Successive cell culture provided clear evidence of cell adhesion and cell proliferation on the Zr FGM surface, indicating good cytocompatibility. The introduction of CAD/computer-aided manufacturing techniques has increased the general acceptance

of Zr in dentistry.⁴⁶ Furthermore, the advances in this technology are essential to research and development of high-strength polycrystalline ceramics, such as stabilized zirconium dioxide (ZrO₂), which could not have been practically processed by traditional laboratory methods.

POWDER METALLURGY METHOD

Powder metallurgy is a process of making components from metallic powders where the properties of the FGMs are dependent on the characteristics of the metal powders used. It depends on particle shape, particle size and distribution, flow rate, compressibility, apparent density, and purity. The manufacturing involves five steps which are: Manufacturing of metal powders, blending and mixing of powders, compacting, sintering, and finally, finishing. Hence, the fabrication processes involve compacting process, powder stacking, wet vibration, vibrating stacking process, centrifugal process, wet powder spray-forming process, sequential slip casting, and slurry dip slip casting and later is sintering.²⁰ Its wide advantages include lower costs, higher raw material availability, simpler processing equipment, lower energy consumption, and shorter processing times. By controlling the particles size in optimum mixing condition together with applying correct pressing technique, the sintering imbalance that occurs can be prevented. Controlling the sintering process and adding the activating phase are suggested. Powder metallurgy technique enables fabrication of materials with a good control of chemical composition and microstructure.²⁰

Powder preparation is done through chemical reactions, electrolytic deposition, grinding, or comminution. During powder processing, precision in weighing correct amounts and the dispersion of the mixed powders are the main consideration. The forming operations are performed at room temperature, while sintering is conducted at atmospheric pressure. The PM method is one of the most commonly employed techniques due to its wide range control on composition, microstructure, and shape-forming capability.⁴⁷ Bhattacharyya et al⁴⁸ synthesized and characterized Al/SiC and Ni/AlO FGMs where they reported that Al/SiC FGMs fabricated by PM technique exhibit homogeneous distribution of fragmented and clustered SiC particles microstructures. Chenglin et al,⁴⁹ who also investigated fabrication of HA-Ti FGM through PM technique, discovered that the bending strength and fracture toughness have a direct relationship with Ti content and it is much higher than human bone strength.

Oleiwi et al⁵⁰ had prepared FGM of Ti and HA using PM process by the layers pressing method. The metallic and ceramic pure powders were mixed in several proportions using ball mills with the rotational speed of 300 rpm

for 1 hour in several proportions. The mixture was later stacked layer-by-layer using the pressing method of cold uniaxial pressing in double action typically between 0.2 and 1 mm thickness with a concentration difference in components' content. The powder of layers was measured by a sensitive balance type (BEL), and sintering process was done using a mini-brute furnace at 400°C for 5 hours and at 1000°C for 2 hours. A review of FGM by El-Wazery and El-Desouky²¹ found that FGM can be used to avoid problems associated with the presence of an interface in material stress singularities due to elastic or thermal property mismatch, poor adhesion, or unwanted reflections at the interface.

The ceramic/metal FGMs can be designed to make use of the heat and corrosion resistances of ceramic combined with the mechanical strength, good machinability, and high toughness and bonding capability of metals. This could be achieved without severe internal thermal stresses. Higher fracture resistance parameters will result in higher toughness due to crack bridging in a graded volume fraction. Powder metallurgy method is the most suitable technique certainly for mass production and upscaling of the FGMs.²¹

FUTURE OF FGM RESEARCH IN DENTISTRY—SIMULATION STUDIES

Several theoretical studies had been conducted using a variety of techniques in producing the best FGMs. Ogawa et al⁵¹ carried out computer simulation for FGM using the centrifugal solid particle method. In this method, the centrifugal force was applied to a mixture of molten metal and solid particles where the particles are arranged in a graded manner along the centrifugal force direction. Computer simulation was carried out for an estimation of the effect of centrifugal force on the formation of graded distribution and the difference of migration rate between two kinds of spherical particles with different diameters and density. They observed that the large particles with low density can have higher migration rate compared with small particles with high density, if the difference in diameter is significantly large.

In another study, a buccolingual sectional model consisting of a single unit implant and four other adjacent teeth was constructed from computerized tomography scan images. Bone remodeling induced by the use of various FGM dental implants was calculated over 4 years. Based on remodeling results, response surface method was adopted to develop a multiobjective optimal design for FGM implantation FGM designs.¹³

In a review by Birman and Byrd,¹¹ they found that material properties evaluated according to theoretical models often disagree with measured values of FGM

constants. The thermal residual stresses should be accounted in the analysis of FGMs, since they may affect local strength and fracture resistance of these materials. Furthermore, they suggested to imply necessity in balancing the advantages of grading against current disadvantages, such as the asymmetry introduced by grading. Other researchers have theoretically studied the cosedimentation technique to fabricate FGMs.^{52,53} A compositional gradient was produced by the settling of particles under gravity in a vessel.³⁶ In other study, a compositionally graded W–Mo composite which was formed through the settling of the W and Mo particles was studied. The density gradient was distributed in the initial clear liquid along the settling direction.^{53,54}

A study by Jamaludin et al⁵⁵ was conducted to assess the grading parameter domination on the displacement and stress distribution of the HA/Ti FGM plates. The simulation of the thermoelastic behaviors was done using a 3D thermomechanical model of a 20-node brick quadratic element, and the plates were subjected to constant and functional, thermal, mechanical, and thermomechanical loadings. The numerical simulations were carried out using the general-purpose finite element software package, which was divided into three parts. First part was temperature field calculation using a thermal analysis preference; second part demonstrated the structural analysis for the displacement and stress field calculation, and finally, the last part combined both analyses to a thermal–structural coupled analysis. They conclude that the HA/Ti FGM has a potential application as a high thermal resistance material because it can withstand the extreme surface temperature and thermal stress.

CONCLUSION

Of late, multiple parallel researches based on the FGM concept are being carried out on dental implants coated with HA, Zr, and its oxides to improve osseointegration and reduce stress behavior. The FGM model could bring better biomechanical, microstructural, and compositional compatibility with natural bone. Another application of FGM concept is in PFM restorations, wherein an FGM composite interlayer significantly reduces failure between metal ceramic and improves the metal–ceramic bond strength. This review attempts to present a thorough understanding of FGMs, their characterization, manufacturing techniques and technology, and their dental applications.

CLINICAL SIGNIFICANCE

Throughout the past decade, the use of endosteal dental implants as a treatment modality of choice in oral rehabilitation cases has increased. Unfortunately, no artificial implants have biomechanical properties equivalent

to that of the surrounding bone. Therefore, the introduction of newer materials that can withstand such applications while maintaining long-term mechanical stability was required. In the past, pure titanium and titanium-based implants with a bioactive HA coating have been used. However, the major concerns about implants coated with HA or Zr, for example, lie in the possible defects and debonding of the coated layer. Based on FGM concept, a graded composition between titanium and HA/Zr on the implant surface has now been achieved that has resulted in improved mechanical behavior and osseointegration.

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