Effect of Graphene Oxide Nanoparticles on the Flexural Strength of all Ceramics- An In-vitro Study

K YOGITHA¹, KASIM MOHAMED², PREETHA KRISHNAMURTHY³

(CC) BY-NC-ND

ABSTRACT

Dentistry Section

Introduction: Desirable properties of monolithic ceramics like strength, stability at high temperatures, high stiffness has made it useful for biomedical, electronic, automotive, industrial, defence and space applications. The brittle nature and mechanical unreliability of monolithic ceramics limit their use.

Aim: To evaluate, if there is an increase in flexural strength of all ceramics incorporated with graphene nanoparticles.

Materials and Methods: This in-vitro study was conducted in the Department of Prosthodontics and Crown and Bridge, Sri Ramachandra Institute of Higher Education and Research, Chennai, Tamil Nadu, India, from February 2021 to April 2021, where 20 specimens of all ceramic dimensions of $20 \times 5 \times 3$ (20 mm in length, 5 mm in width, 3 mm in thickness) according to American Dental Association (ADA) specification number 69, 1991 for flexural strength were grouped into group A (Control) with no Graphene Oxide (GO), group B with GO added. A universal testing machine with a cross head speed of 0.5 mm/min and a span length of 15 mm was used to load the specimen in the centre. To compare the flexural strength of the two groups, the Mann Whitney U-test was used. Normality was the data assessed using Shapiro-Wilk and Kolmogorov-Smirnov test.

Results: The results show that there was a significant difference (p=0.001) in the mean flexural strength between the groups. Group B had the highest mean flexural strength of 562.61 MPa, while Group A had the lowest of 458.61 MPa.

Conclusion: The results concluded that, all ceramics incorporated with graphene nanoparticles showed superior flexural strength.

Keywords: Crowns, Dental ceramics, Dental materials, Fixed partial denture, Mechanical properties

INTRODUCTION

Ceramics, due to their ability to mimic the optical characteristics of enamel and dentine as well as for their biocompatibility and chemical durability, are widely used in dentistry. In 1789, de Chemant patented the first porcelain tooth material [1]. Dr. Charles Land introduced one of the first ceramic crowns to dentistry in 1903. These crowns had excellent aesthetics, but the disadvantage of these all ceramics was chipping of the veneer porcelain from its core, which resulted in minute fractures in the restoration [1,2]. This occurred due to the low flexural strength of porcelain. Unfortunately, it also had issues such as a lifeless appearance and poor marginal fit. To address these issues, high-strength and aesthetically pleasing ceramics such as aluminum and zirconium oxide were developed [3].

While metal/ceramic bilayers are still considered the gold standard for Fixed Partial Dentures (FPDs), much research has been done to achieve the same level of excellence in all-ceramic systems [4-6]. The low biocompatibility and translucency of metals compared to ceramics have contributed to the use of it, as an infrastructure material for multi-layer restorations [5]. However, based on the systematic review by Sailer I et al., metal-ceramic FPD's exhibited significant higher survival rates at five years when compared with all-ceramic FPD's. The fracture of the veneering ceramic is the most common cause for failure of FPD's made out of glass-ceramics or glass-infiltrated ceramics [6]. This was a major limitation for their unrestricted use in Prosthodontics. Thus with advent of monolithic ceramics, the use of all ceramic aesthetic prosthesis had evolved. The advantageous properties like high stiffness, strength and stability at elevated temperatures, make monolithic ceramics favourable for biomedical, electronic, automotive, industrial, defence and space applications. The brittle nature and mechanically unreliability, thus, limits the use of monolithic ceramics [7].

With the advances in technology and material sciences, a number of techniques have been put forward to improve the performance of materials. Nanotechnology, which is called "Manufacturing technology of the 21st century," is one of the technologies that manipulate substances on a scale of 100 nm or less to create many materials with various physical properties and functions. In the last years, there has been a continuous research and progress in prosthetic material field for dental ceramics. Zirconia reinforced-Lithium Silicate ceramics (ZLS) were introduced to overcome the shortcomings of the lithium disilicate ceramics [7]. To date, data on mechanical properties and clinical performance of ZLS are still limited and often controversial [8-12].

Graphene, a two dimensional single layer sp2 hybridised carbon atoms with hexagonal-packed configuration has been studied extensively to enhance performances of materials due to their unique properties [13]. Moreover, it possesses exceptional physicochemical, optical, and mechanical properties [14]. Since then, research efforts have focused on unearthing potential applications, including various biomedical applications such as drug carriers, contrast agents, biosensors, bimolecular analysis, and scaffolds for tissue engineering [15]. Its excellent mechanical properties, extreme chemical stability, superior biocompatibility, good antibacterial properties, and favourable tribological properties, all contribute to reduced wear and friction. It is only a single atomic layer thick, and therefore, the world's thinnest material, yet 200 times stronger than steel [16]. Latest studies have shown that graphene and graphene-based composites (especially GO) possess a series of merits like large surface area, excellent elasticity and ductility, good biocompatibility, and exceptional mechanical strength [14,16,17]. While research and development of graphene based dental biomaterials are at nascent stage, their idiosyncratic properties and the potential to be discreetly or combinedly functionalised with biomaterials pave way for several unique clinical applications. Therefore, the present study aims to evaluate the effect of addition of graphene nanoparticles on the flexural strength of dental ceramics.

K Yogitha et al., Effect of Graphene Oxide on the Flexural Strength of all Ceramics

MATERIALS AND METHODS

This in-vitro study was conducted in the Department of Prosthodontics and Crown and Bridge, Sri Ramachandra Institute of Higher Education and Research, Chennai, Tamil Nadu, India, where 20 from February 2021 to April 2021.

Inclusion criteria: All ceramic specimens, in accordance with the ADA specification number 69, 1991, for testing the flexural strength of dental ceramic, were included in the study.

Exclusion criteria: All other specimens of all ceramics not fulfilling the above criteria, were excluded from the study.

Study Procedure

A sample of 10 for each group was prepared. The samples were made with a wax patter of standardised dimensions measuring: $20 \times 5 \times 3$ (20 mm in length, 5 mm in breadth, 3 mm in width) in accordance with ADA specification number 69, 1991 for testing the flexural strength of dental ceramic [3,18]. The wax pattern was made with indentation in the centre measuring 2 mm in thickness and 10 mm in length [Table/Fig-1], so that pressed ceramic substructure can be layered. The control group was layered with ceramics and the test group was layered with ceramics incorporated with 0.10 grams of GO [Table/Fig-2] [17]. GO was obtained from BT Corp Generic Nano Pvt. Ltd., India, in powder form mixed along VITA VM 9 was used in the test group.





The twenty samples were divided into two groups: Group A (n=10)-Control (ceramic block) and Group B (n=10)-test sample (ceramic block incorporated with 0.10 grams of GO) [Table/Fig-3]. In the present in-vitro study, VITA PM 9 (ceramic ingots) was used as substructure and layered with VITA VM 9 for the control and the substructure was layered with VITA VM 9 incorporated with GO for the test group. All the specimens were subjected to three-point bend test performed with universal testing machine [Table/Fig-4]. The load was applied at the centre of the specimen with a cross head speed of 0.5 mm/min and a span length of 15 mm, recording the ultimate load resulting specimen fracture.

Groups	Material	No. of samples			
Group A	Ceramic substructure with ceramic layering	10			
Group B	Ceramic substructure layered with Graphene Oxide (GO) incorporated ceramics	10			
[Table/Fig-3]: Groups and materials.					



[Table/Fig-4]: Flexural strength tested with universal testing machine.

STATISTICAL ANALYSIS

The Mann Whitney U-test was used to compare the flexural strength between the two groups. Normality was the data assessed using Shapiro-Wilk and Kolmogorov-Smirnov test. The analyses were performed using IBM Statistical Package for the Social Sciences (SPSS) Statistics 20.0 (IBM Corp., Armonk, NY, USA).

RESULTS

The mean values for flexural strength in megapascals for all groups are shown in [Table/Fig-5]. There was a statistically significant difference in the flexural strength between lithium disilicate ceramic VITA VM 9 substructure, layered with VITA VM 9 for group A and substructure layered with GO incorporated ceramics for group B (p-value=0.001). Group B had the highest mean flexural strength, while the lowest is seen in group A.

Group	Mean (MPa)	N	Std. Deviation	p-value	
Group A	458.61	10	16.03229		
Group B	562.61	10	15.63454	0.001	
Total	510.61	20	55.53247		
[Table/Fig_5]. The mean flexural strength is expressed in MPa (Mega Pascal)					

[Table/Fig-5]: The mean flexural strength is expressed in MPa (Mega Pascal)

DISCUSSION

All ceramics are commonly used material in the fields of Prosthodontics. But, it shows weak physical and mechanical properties [19,20]. Many experiments have been undertaken to improve the flexural strength of all ceramics, in order to prevent the fracture and clinical failure [21,22]. There was a scope for improvement and experimentation as these tests showed inconclusive results which were clinically inapplicable.

Nanoparticles are used based on the principle that reduction of filler size is known to increase the mechanical properties of all ceramics [23]. In the present research, GO is used to improve the flexural strength due to their dispersibility in the ceramic matrix. They show good dispersion and good adhesion to polymers, indicating chemical compatibility and creation of chemical bonds between the surface and polymer [23]. Incorporating GO into Polymethyl Methacrylate (PMMA), enhanced mechanical properties and increases in volumetric stability during polymerisation, clinically applicable drug-free antimicrobial adhesion properties of GO-PMMA complexes have been evaluated [24]. An Y et al., and Song J et al., studied the mechanical properties of a PMMA-based dental composite reinforced with GO incorporated into PMMA by ultrasonic dispersion in liquid phase followed by mechanical milling. The results showed that the presence of GO made PMMA harder and more resistant [25,26].

The GO is used since it reduces the adherence of biomolecules, aids in achieving better aesthetics, has antimicrobial properties and improves mechanical properties of all ceramics [27]. The amount of GO concentration used in the present study was restricted to 0.10 grams. The addition of GO has significant increase in the flexural strength of all ceramics. Better marginal adaptability of all ceramics crowns incorporated with GO could be observed due to increase in flexion, with no considerable change in compressive strength. Graphene possesses a combination of large surface area, two dimensional high aspect ratio sheet geometry, and outstanding mechanical properties making it the most promising nanofiller composite materials [28-32]. A number of studies using polymer-based matrices have shown that graphene fillers can significantly improve the mechanical properties of polymers at relatively low nanofiller loading [33-38].

The modification of ceramics by the addition of graphene nanoparticles, can be effectively brought about by powder incorporation, sintering consolidation and colloidal dispersion. Despite the different chemical properties of graphene, successful dispersion aids in strengthening [39-41]. A larger area of contact and possibly greater bond strength, between the graphene and the ceramic grains, reduces the crack propagation along grain boundaries attributes to the two dimensional sheet-like structure of graphene [23,40,41]. This property of GO had proved to strengthen the material, which attributes to significant improvement in the flexural strength of all ceramics. However, the dispersion of GO molecules can only be assessed with a scanning electron microscope. Additionally, the antimicrobial efficacy and the improvement in translucency conferred by GO, were not within the purview of the present study.

Limitation(s)

Despite the increase in the flexural strength, the tested samples were not subjected to other tests like fractographic analysis to examine the location, orientation, extent of cracking and dispersion of GO. Within the limitations, clinical trials with larger sample sizes are required to extrapolate these results for their clinical use.

CONCLUSION(S)

Graphene nanoparticles can be functionalised and combined with several biomolecules and biomaterials. It holds a great potential, in enhancing the existing biomaterials with superior properties and new capabilities. Addition of GO to all ceramics, brought about a significant improvement in the flexural strength, thus, diversifying its usage and reducing the incidence of marginal failures. Due to the lacunae of the "perfect" material, that can be applied universally to all clinical situations, GO could aid as a breakthrough in the field of material science. Ultimately, the use of engineered graphene-based nanomaterials in dentistry, could lead to reliable dental treatments in the near future and therefore, deserves profound examination.

REFERENCES

- Anusavice KJ. Phillips' Science of Dental Materials. 11th South Asian edition: Elsevier; 2011.
- [2] Zarone F, Russo S, Sorrentino R. From porcelain-fused-to-metal to zirconia: Clinical and experimental considerations. Dent Mater. 2011;27(1):83-96.
- [3] Chougule KJ, Wadkar AP. An in vitro comparative evaluation of flexural strength of monolithic zirconia after surface alteration utilising two different techniques. J Clin Diagn Res. 2017;11(8):ZC20.
- [4] Motta AB, Pereira LC, da Cunha AR. All-ceramic and porcelain-fused-to-metal fixed partial dentures: A comparative study by 2D finite element analyses. J Appl Oral Sci. 2007;15:399-405.
- [5] Silva LHD, Lima E, Miranda RBP, Favero SS, Lohbauer U, Cesar PF. Dental ceramics: A review of new materials and processing methods. Braz Oral Res. 2017;31(suppl 1):e58.

K Yogitha et al., Effect of Graphene Oxide on the Flexural Strength of all Ceramics

- [6] Sailer I, Pjetursson BE, Zwahlen M, Hammerle CH. A systematic review of the survival and complication rates of all-ceramic and metal-ceramic reconstructions after an observation period of at least 3 years. Part II: Fixed dental prostheses. Clin Oral Implants Res. 2007;18:86-96.
- [7] Porwal H, Grasso S, Reece MJ. Review of graphene-ceramic matrix composites. Adv Appl Ceram. 2013;112(8):443-54.
- [8] Zarone F, Di Mauro MI, Ausiello P, Ruggiero G, Sorrentino R. Current status on lithium disilicate and zirconia: A narrative review. BMC Oral Health. 2019;19(1):01-04.
- [9] de Carvalho Ramos N, Campos TM, de La Paz IS, Machado JP, Bottino MA, Cesar PF, et al. Microstructure characterization and SCG of newly engineered dental ceramics. Dent Mater. 2016;32(7):870-78.
- [10] Fathy SM, Swain MV. In-vitro wear of natural tooth surface opposed with zirconia reinforced lithium silicate glass ceramic after accelerated ageing. Dent Mater. 2018;34(3):551-59.
- [11] Zimmermann M, Koller C, Mehl A, Hickel R. Indirect zirconia-reinforced lithium silicate ceramic CAD/CAM restorations: Preliminary clinical results after 12 months. Quintessence Int. 2017;48(1):19-25.
- [12] Saavedra GSFA, Rodrigues FP, Bottino MA. Zirconia-reinforced lithium silicate ceramic-a 2-year follow-up of a clinical experience with anterior crowns. Eur J Prosthodont Restor Dent. 2017;25(1):57-63.
- [13] Geim AK, Novoselov KS. The rise of graphene. Nat Mater. 2007;6(3):183-91.
- [14] Sur UK. Graphene: A rising star on the horizon of materials science. International Journal of Electrochemistry. 2012;2012:237689.
- [15] Zhang Y, Nayak TR, Hong H, Cai W. Graphene: A versatile nanoplatform for biomedical applications. Nanoscale. 2012;4:3833-42.
- [16] Dong HS, Qi SJ. Realising the potential of graphene-based materials for biosurfaces-A future perspective. Biosurface and Biotribology. 2015;1(4):229-48.
- [17] Han S, Sun J, He S, Tang M, Chai R. The application of graphene-based biomaterials in biomedicine. Am J Transl Res. 2019;11(6):3246.
- [18] American National Standard/American Dental Association Specification number 69 for Dental Ceramic.
- [19] Rizkalla AS, Jones DW. Indentation fracture toughness and dynamic elastic moduli for commercial feldspathic dental porcelain materials. Dent Mater. 2004;20(2):198-206.
- [20] Shenoy A, Shenoy N. Dental ceramics: An update. Journal of conservative dentistry. J Conserv Dent. 2010;13(4):195.
- [21] Bajraktarova-Valjakova E, Korunoska-Stevkovska V, Kapusevska B, Gigovski N, Bajraktarova-Misevska C, Grozdanov A. Contemporary dental ceramic materials, a review: Chemical composition, physical and mechanical properties, indications for use. Open access Maced J Med Sci. 2018;6(9):1742.
- [22] White SN, Miklus VG, McLaren EA, Lang LA, Caputo AA. Flexural strength of a layered zirconia and porcelain dental all-ceramic system. J Prosthet Dentistry. 2005;94(2):125-31.
- [23] Walker LS, Marotto VR, Rafiee MA, Koratkar N, Corral EL. Toughening in graphene ceramic composites. ACS Nano. 2011;5(4):3182-90.
- [24] Jin J, Zhang L, Shi M, Zhang Y, Wang Q. Ti-GO-Ag nanocomposite: The effect of content level on the antimicrobial activity and cytotoxicity. Int J Nanomedicine. 2017;12:4209.
- [25] An Y, Liu B, Yan X, Pet J, Liu W. The experimental study on wear resistance of the denture base material reinforced with graphene oxide. Tribology. 2013;33:222-28.
- [26] Song J, Zhang J, Lin C. Influence of graphene oxide on the tribological and electrical properties of PMMA composites. J Nanomater. 2013;2013:846102.
- [27] Mohammed H, Kumar A, Bekyarova E, Al-Hadeethi Y, Zhang X, Chen M, et al. Antimicrobial mechanisms and effectiveness of graphene and graphenefunctionalised biomaterials. A scope review. Front Bioeng Biotechnol. 2020;8:465.
- [28] Yu A, Ramesh P, Sun X, Bekyarova E, Itkis ME, Haddon RC. Enhanced thermal conductivity in a hybrid graphite nanoplateletcarbon nanotube filler for epoxy composites. Adv Mater. 2008;24:4740-44.
- [29] Lee C, Wei X, Kysar JW, Hone J. Measurement of the elastic properties and intrinsic strength of monolayer graphene. Science. 2008;321(5887):385-88.
- [30] Wang X, Zhi L, Müllen K. Transparent, conductive graphene electrodes for dyesensitized solar cells. Nano letters. 2008;8(1):323-27.
- [31] Balandin AA, Ghosh S, Bao W, Calizo I, Teweldebrhan D, Miao F, et al. Superior thermal conductivity of single-layer graphene. Nano Letters. 2008;8(3):902-07.
- [32] Rafiee MA, Lu W, Thomas AV, Zandiatashbar A, Rafiee J, Tour JM, et al. Graphene nanoribbon composites. ACS Nano. 2010;4(12):7415-20.
- [33] Watcharotone S, Dikin DA, Stankovich S, Piner R, Jung I, Dommett GH, et al. Graphene-silica composite thin films as transparent conductors. Nano letters. 2007;7(7):1888-92.
- [34] Khan U, May P, O'Neill A, Coleman JN. Development of stiff, strong, yet tough composites by the addition of solvent exfoliated graphene to polyurethane. Carbon. 2010;48(14):4035-41.
- [35] Rafiee MA, Rafiee J, Wang Z, Song H, Yu ZZ, Koratkar N. Enhanced mechanical properties of nanocomposites at low graphene content. ACS Nano. 2009;3(12):3884-90.
- [36] Rafiee MA, Rafiee J, Srivastava I, Wang Z, Song H, Yu ZZ, et al. Fracture and fatigue in graphene nanocomposites. Small. 2010;6(2):179-83.
- [37] Ramanathan T, Abdala AA, Stankovich S, Dikin DA, Herrera-Alonso M, Piner RD, et al. Functionalised graphene sheets for polymer nanocomposites. Nat Nanotechnol. 2008;3(6):327-31.
- [38] Stankovich S, Dikin DA, Dommett GH, Kohlhaas KM, Zimney EJ, Stach EA, et al. Graphene-based composite materials. Nature. 2006;442(7100):282-86.
- [39] Li Z, Khun NW, Tang XZ, Liu E, Khor KA. Mechanical, tribological and biological properties of novel 45S5 Bioglass® composites reinforced with in situ reduced graphene oxide. J mech behave Biomed Mater. 2017;65:77-89.

K Yogitha et al., Effect of Graphene Oxide on the Flexural Strength of all Ceramics

[40] Xie H, Cao T, Rodríguez-Lozano FJ, Luong-Van EK, Rosa V. Graphene for the development of the next-generation of biocomposites for dental and medical applications. Dent Mater. 2017;33(7):765-74. [41] Baradaran S, Moghaddam E, Basirun WJ, Mehrali M, Sookhakian M, Hamdi M, et al. Mechanical properties and biomedical applications of a nanotube hydroxyapatite-reduced graphene oxide composite. Carbon. 2014;69:32-45.

PARTICULARS OF CONTRIBUTORS:

- 1. Postgraduate, Department of Prosthodontics and Crown and Bridge, Sri Ramachandra Institute of Higher Education and Research, Chennai, Tamil Nadu, India.
- 2. Head, Department of Prosthodontics and Crown and Bridge, Sri Ramachandra Institute of Higher Education and Research, Chennai, Tamil Nadu, India.
- 3. Postgraduate, Department of Prosthodontics and Crown and Bridge, Sri Ramachandra Institute of Higher Education and Research, Chennai, Tamil Nadu, India.

NAME, ADDRESS, E-MAIL ID OF THE CORRESPONDING AUTHOR: Dr. Kasim Mohamed,

Head, Department of Prosthodontics and Crown and Bridge, Sri Ramachandra Institute of Higher Education and Research, Chennai, Tamil Nadu, India. E-mail: yogitajuly93@gmail.com

AUTHOR DECLARATION:

- Financial or Other Competing Interests: None
- Was Ethics Committee Approval obtained for this study? NA
- Was informed consent obtained from the subjects involved in the study? NA
- For any images presented appropriate consent has been obtained from the subjects. NA

PLAGIARISM CHECKING METHODS: [Jain H et al.]

- Plagiarism X-checker: Jun 30, 2022
- Manual Googling: Nov 01, 2022
- iThenticate Software: Nov 24, 2022 (18%)

Date of Submission: Jun 25, 2022 Date of Peer Review: Jul 30, 2022 Date of Acceptance: Nov 25, 2022 Date of Publishing: Feb 01, 2023

ETYMOLOGY: Author Origin

www.jcdr.net